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Metal Concentrations in Littoral Sediments and Aquatic Macroinvertebrates in the St. Lawrence River, near Cornwall, Ontario

Concentrations de métaux dans les sédiments littoraux et les macroinvertébrés aquatiques dans le Fleuve Saint-Laurent, près de Cornwall, Ontario

Thesis submitted to the
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Abstract

Concentrations of Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn were measured in surficial sediments and in five aquatic macroinvertebrate taxa from shallow littoral areas of the St. Lawrence River, near Cornwall, Ontario. The principal objectives were to assess metal levels in littoral sediments and littoral macroinvertebrates, and to determine the influence of local point sources relative to upstream inputs. Metal concentrations in littoral sediments were generally below the lowest effect level of the Ontario Provincial Sediment Quality Guidelines and were positively related to the amount of silt and clay (< 63 μm) and of organic matter in the sediments. Hg concentrations were, however, greatly enriched at a site 1 km downstream of two historical point sources of Hg (Domtar Fine Papers/ICI Forest Products). Littoral sediments had lower metal levels than contaminated areas of the Great Lakes and deeper sites in the Cornwall area. Differences in metal concentrations among littoral sediments and those of deeper sites reflected mainly the sorting of fine-grained sediments towards the deeper parts of the river. Results of these comparisons, however, suggested a problem of Hg contamination in the Cornwall area. Metal levels in littoral macroinvertebrates were assessed for the amphipod *Gammarus fasciatus*, the decapod *Orconectes* sp., the gastropod *Bythinia tentaculata*, the Chironomidae (midges) and the Oligochaeta (worms). Metal concentrations in macroinvertebrates were found to differ among taxonomic groups, and these differences were metal-specific. When compared to concentrations in sediments, invertebrates had lower levels of Cd (except for *B. tentaculata*), Cr, Fe, Ni and Pb, and higher concentrations of Cu and Zn. Concentrations of Hg were similar to levels in the sediments, except for slightly higher concentrations in *B. tentaculata*. Metal concentrations in littoral macroinvertebrates were generally similar or lower than levels described for the same or related taxa from low to moderately contaminated aquatic environments. However, the high Hg concentration in sediments at a site 1 km downstream of two historical point sources of Hg were paralleled by elevated levels in all macroinvertebrate taxa, except *B. tentaculata*. Concentrations of Cr, Fe, Ni and Zn in chironomids and oligochaetes were similar or higher than levels reported for contaminated areas of the Great Lakes and deeper sites in the Cornwall area, and this despite the relatively much lower concentrations in littoral sediments. Comparisons of metal concentrations in sediments and in macroinvertebrates among reference, most impacted, and least impacted sites, and in sediments among downstream and upstream sites, showed that local sources of Hg and Zn contributed to the contamination of littoral sediments and littoral macroinvertebrates. However, the relatively low concentrations suggest that these local sources generally had only a small impact on the littoral zone, except for the elevated Hg levels in sediments and invertebrates immediately downstream of Domtar Fine Papers/ICI Forest Products. Results for other metals reflected the influence of upstream sources (Lake Ontario) or natural levels.

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Introduction

Metals of natural and anthropogenic origins in surface waters of aquatic systems can exist in dissolved forms or associated with suspended particulate materials (inorganic particles, planktonic living organisms, and detritus), but eventually the bulk of these metals accumulates in bottom sediments. In many areas of the Laurentian Great Lakes and of the St. Lawrence River, sediments show high metal concentrations as a result of the industrial and urban development that has occurred during the last century (e.g., Hart et al., 1986; Sloterdijk, 1991; Jaagumagi and Persaud, 1992; Krantzberg and Boyd, 1992). In particular, sediments in the Cornwall (Ontario) region of the St. Lawrence River have been found to have elevated concentrations of cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb), nickel (Ni), and zinc (Zn) (Jaagumagi and Persaud, 1992; St. Lawrence RAP Team, 1992; Richman, 1994). For most metals, those concentrations are higher than pre-industrial levels estimated from core samples from the eastern depositional basin of Lake St. Francis (Carignan et al., 1994).

The accumulation of elevated metal concentrations in sediments of aquatic environments can result in biological impacts. In particular, numerous studies have shown that, in contaminated freshwater systems, aquatic macroinvertebrates can accumulate significant quantities of metals (Hart et al., 1986; Alikhan et al., 1990; van Hattum et al., 1991; Cain et al., 1992; Kiffney and Clements, 1993). Metal bioaccumulation can occur through feeding, absorption from the water, and adsorption to external surfaces, and once in the animal's tissues, metals can cause chronic or acute toxic effects (Luoma, 1983; Hare, 1992). This is of concern because aquatic macroinvertebrates play essential ecological roles in aquatic ecosystems. These organisms are involved in the decomposition of organic matter, in nutrient cycling and in the trophic transfer of energy (Luoma, 1989; Plante and Downing, 1989; Woodward et al., 1994). Aquatic macroinvertebrates are an important food resource for many aquatic and terrestrial organisms, including fish and waterfowl, and can therefore play an important role in metal transfer to the food chain (Allard and Stokes, 1989; Moore et al., 1991; Woodward et al., 1994).

In part because of the possibility of biological impacts of sediment contamination, remedial measures are being considered for the Cornwall-Lake St. Francis region of the St. Lawrence River under the St. Lawrence River Remedial Action Plan (St. Lawrence RAP Team, 1992) and the Ecosystem Recovery on the St. Lawrence research project (Crabbé et al., 1992). The St. Lawrence Recovery project complements previous and ongoing research done towards the development of the St. Lawrence Remedial Action Plan. The development of effective remedial actions, however, requires a good understanding of the extent and degree of sediment contamination, of biological impacts, and of the role of local sources compared to upstream inputs.

Much information exists regarding metal levels in surficial sediments of deep open-water and relatively deep near-shore areas in the Cornwall region. Several recent environmental investigations have shown that the concentrations of Cd, Cr, Cu, Hg, Pb, Ni, and Zn at several locations exceed the lowest effect level of the Ontario Provincial Sediment Quality Guidelines (Persaud et al., 1993) for the protection of benthic communities, and that in one area immediately downstream of a historical point source, Cu, Hg, Pb, and Zn exceed the severe effect level (Anderson, 1990; Jaagumagi and Persaud, 1992; Poulton, 1994; Richman, 1994). Hg concentrations in sediments are particularly very high in this stretch of the St. Lawrence River as a few other locations also exceed the severe effect level of the sediment quality guidelines (Richman, 1994). Based on the spatial distribution of grain-size corrected metal concentrations in sediments and on effluent monitoring data, Anderson (1990) and Richman (1994) concluded that local sources had contributed to the Cu, Hg, Pb, and Zn contamination of sediments at one (Cu, Pb) or several locations (Hg, Zn), while Cd, Cr, Fe, and Ni levels reflected upstream inputs from Lake Ontario. The spatial pattern of Hg concentrations in Lake St. Francis sediments also points to a significant Hg source in the Cornwall region (Sloterdijk, 1991).

Two of the above studies have also assessed metal bioaccumulation in benthic macroinvertebrates. Jaagumagi and Persaud (1992) reported, for several sites, the bioaccumulation of Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn in macroinvertebrate samples mostly composed of Chironomidae and Oligochaeta. Richman (1994) described Hg levels in Chironomidae, and concluded that concentrations at one location appeared to be influenced by the proximity of a local source of Hg. In another study, Kauss et al. (1988) described, for a few sites in the area, metal concentrations in pooled samples of different combinations of taxa.

In river ecosystems, however, the most biologically productive areas are the shallow littoral zones. Yet, in the Cornwall region, metal concentrations in sediments as well as bioaccumulation in aquatic macroinvertebrates have not been assessed in these important areas. Accordingly, it is not known if local sources compared to upstream inputs (natural or anthropogenic) have a significant impact on metal levels in littoral sediments and littoral macroinvertebrates.

In this study, I measured the concentrations of Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn in surficial sediments and in aquatic macroinvertebrates of shallow littoral areas in the St. Lawrence River. The principal objectives were to assess metal levels in littoral sediments and littoral macroinvertebrates, and to determine the influence of local point sources relative to upstream inputs. To accomplish these objectives, I examined the effect of sediment composition (proportion of silt and clay, and quantity of organic matter) on metal concentrations in sediments, and of body size (body weight) on metal bioaccumulation in macroinvertebrates, as well as taxon-specific differences in metal bioaccumulation. Metal concentrations in sediments and organisms were compared to quality guidelines (sediments only), with results of recent provincial investigations, and with results reported for contaminated areas in the Great Lakes-St. Lawrence River system and for other aquatic systems. The influence of local sources of metals was assessed by comparing concentrations in sediments and macroinvertebrates among sites grouped relative to their position to important local sources, and by comparing concentrations in sediments with levels observed immediately upstream of the Cornwall area.

Materials and Methods

Study area and study sites

The St. Lawrence River is the natural outlet of the Laurentian Great Lakes, one of the most industrialized hydrographic basins in North America. In the Cornwall (Ontario) area (Fig. 1), 200 km downstream, 95% of the river flow is contributed by Lake Ontario (St. Lawrence RAP Team, 1992). Immediately upstream of the city of Cornwall, the St. Lawrence River is impounded by the Moses-Saunders Dam to form Lake St. Lawrence. Below the dam, the river is characterized by a strong water flow and divides around Cornwall Island into a north and a south channel. Further downstream, the river widens to form Lake St. Francis, whose water levels are controlled by the Beauharnois Dam. A more detailed description of the physical characteristics of this stretch of the St. Lawrence River can be found in the Remedial Action Plan Stage 1 Report (St. Lawrence RAP Team, 1992).

In the Cornwall area, Domtar Fine Papers, ICI Forest Products (formerly CIL), Courtaulds Films (closed in 1989), and Courtaulds Fibers (closed in 1992) (Fig. 1) represent important historical point sources of metals to the St. Lawrence River (St. Lawrence RAP Team, 1992). Domtar Fine Papers is a pulp and paper mill and, until 1970, it was a significant source of Hg because of the use of mercury-based slimicides. ICI Forest Products, a chlor-alkali plant using a mercury cell process, was until the early 1980's a major source of Hg to the river. Plant modifications were then brought which allowed the company to meet discharge limits. Finally, in 1994 the mercury cell process was abandoned and the associated plant decommissioned. Courtaulds Fibers and Courtaulds Films were rayon and cellophane manufacturers, respectively. The production of rayon required the use of Zn, and because of that Courtaulds Fibers was responsible for the discharge of very high amounts of Zn to the river. Courtaulds was also a major source of Hg and an appreciable source of Pb.

In this study, sediments and aquatic macroinvertebrates were sampled from 16 shallow littoral sites (depths of 0.5 - 1.1 m) situated in embayments and other calm reaches, from downstream of the Moses-Saunders Dam to Thompson Island in Lake St. Francis (Fig. 1). Study sites were generally vegetated with macrophytes and coarse substrate sediments were avoided.

The goal in site selection was to obtain a gradient in metal concentrations, based on the location of local sources. Three sites (D11 - D13) were situated upstream of local metal sources, 2 sites (D14 and D15) were in the north channel downstream of Domtar Fine Papers and ICI Forest Products, but upstream of Courtaulds Fibers and Courtaulds Films, and 11 sites (D1 - D10, and D16) were downstream of these local sources. For the purpose of assessing the influence of local metal sources, sediments were also sampled at an additional 8 shallow littoral sites (U17 - U24) located upstream of the Moses-Saunders Dam in Lake St. Lawrence (Fig. 1).

Determination of metal concentrations in sediments

In order to account for sediment heterogeneity, 3 sampling stations were established at each study site (2 at site D16). Sampling stations were usually less than 50-100 m apart. At each station, the uppermost 2.5 cm of sediments were sampled using a Plexiglas coring tube. Three samples were collected per station in order to get enough material for the various analyses (only 1 sample per station at site D1 and D2, all 3 stations pooled). Sediment samples were placed in polyethylene sampling bags (1 bag/station), kept on ice in the field and frozen the same day at -18°C until analysis. Sediment sampling was carried out over a one month period from mid-July to mid-August 1994.

In the laboratory, frozen sediment samples were thawed at 4°C and prepared within one week. After removing excess water, the samples were thoroughly mixed and several subsamples were taken for different analyses. To minimize variability among subsamples, plants, visible shells and other debris were avoided. To determine the sediment dry weight to wet weight ratio, 1 or 2 subsamples were dried at 60°C. For the determination of metal concentrations, approximately 1 g equivalent dry weight of wet sediments was transferred to a 50 ml centrifuge tube. Prepared samples were delivered to the Analytical Method Development Section at the Geological Survey of Canada (Ottawa, Canada) and extracted by aqua-regia ($3HCl:1HNO_3$, v/v). 4 ml of 16 M HNO_3 followed by 12 ml of 12 M HCl were added to the samples, which were then left to sit overnight at room temperature. Uncapped samples were heated to 90°C in a water bath and maintained at that temperature for 3 hrs. After cooling, the volume of acid was made up to 15 ml with cooked aqua-regia and centrifuged. Finally, the acid was decanted into 15 ml centrifuge tubes and diluted to 10% aqua-regia. Metal concentrations in the extracts were analyzed by flame atomic absorption spectrometry (FAAS: Cu, Fe, Ni, Pb, and Zn), inductively coupled plasma-mass spectrometry (ICP-MS: Cd), hydride-generation ICP-MS (Hg) and inductively coupled plasma-atomic emission spectrometry (ICP-AES; Cr). For any extract that had a concentration value of a metal below the analytical detection limit, a figure of ½ times the detection limit was entered as the concentration value for that metal. This occurred in some cases for Cu and Pb (including all samples at site D13) downstream and for Pb upstream. The particle size distribution of sediments was determined by a modification of the wet-sieving method described in Lewis (1984). Three sediment fractions were determined: gravel (>2 mm), sand (0.063 - 2 mm), and silt and clay (<0.063 mm). The organic matter content of sediments (downstream) was estimated by loss of weight on ignition. Previously dried subsamples were placed in a cold muffle furnace and brought up to 475°C over a period of ½-¾ hr., then maintained at this temperature for approximately 4 hours.

Quality assurance of the analytical procedures was verified with the analysis of reference lake sediments LKSD-4 (Natural Resources Canada), of replicates, and of acid blanks. Metal concentrations in blanks were below detection, except for Cd in the analyses of upstream sediments. Therefore, all upstream Cd concentrations (including the reference material) were corrected (corrected concentration value = observed concentration value - blank contamination value + 0.5 * the detection limit). In the analyses of downstream sediments, recovery of Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn from LKSD-4 sediments ($n = 4$) was 115, 86, 101, 96, 115, 86, 93, and 98%, respectively. Measured concentrations were within ± 1 SD of the provisional mean value for each element, except Cr and Hg. In the analyses of upstream sediments, recovery of Cd, Cr, Cu, Fe, Hg, Ni, Pb, and Zn was 97, 92, 110, 93, 101, 80, 92, and 96%, respectively ($n = 3$). Measured concentrations were within ± 1 SD of the provisional mean value for each element, except Cu and Ni. In both upstream and downstream analyses, however, measured concentrations were all within ± 2 SD of the provisional value. Measured concentrations in reference sediments also varied little among samples (CV = 3 - 9%). Replicate analyses gave similar results, except in two cases for Pb (the replicates differed considerably). Also, during data verification, 2 extreme Pb concentration values (for one sample at site D3 and one at D12) were deleted from the dataset.

When sample values at those sites were compared among each other, these extreme Pb concentrations did not concur with values obtained with 2 weak acid extractions (0.5N HCl and 1N HCl; results not included here).

Determination of metal concentrations in macroinvertebrates

At each downstream study site, aquatic macroinvertebrates were sampled from surficial sediments and macrophytes using a dip net. Organisms were sorted from sediments, plants and debris in the field (sorting of organisms at site D1 was only partly done in the field and was completed in the laboratory the next day) and separated into different taxonomic groups. The 3 stations were sampled repeatedly until a sufficient number of individuals of each taxon was collected. Criteria for collecting individuals of a given taxon were their widespread distribution to allow comparison of metal bioaccumulation among study sites and their relative abundance and large size to obtain sufficient biomass for metal analysis. Apart from a few exceptions, collected organisms belonged to 5 taxa: the amphipod *Gammarus fasciatus*, the decapod *Orconectes* sp., the gastropod *Bythinia tentaculata*, the Chironomidae (midges) and the Oligochaeta (worms). The two latter taxa were not separated beyond the family and order levels, respectively. Collected organisms were placed in appropriate containers (according to their size) and excess water was removed. Samples were kept on ice in the field and frozen the same day at -18 °C until analysis. Because littoral macroinvertebrates may play an important role in metal transfer to organisms of higher trophic levels through foodweb relationships, their digestive tract was not depurated as this undigested material also represents a metal source to those organisms. Besides one site sampled in late June, the sampling of macroinvertebrates was carried out over a five week period from mid-July to mid-August 1994. During that period, some study sites were revisited to collect more samples.

In the laboratory, macroinvertebrate samples were prepared for metal analysis following a method modified from Cain et al. (1992). Frozen samples were thawed at room temperature and rinsed thoroughly in deionized distilled water to remove any particles and detritus from external surfaces. Gastropods were separated from their shells, as only the soft tissues were analyzed for metal content. Excess water was removed and the organisms were dried at 80°C for 48 hours.

Organisms of the same taxon were sorted by size and, in order to meet analytical detection limits for trace metal analysis, similar-sized organisms were pooled to a total dry weight of usually at least 50 mg. Macroinvertebrate body size was measured as the mean dry weight of the individuals composing a sample. Furthermore, for most sites, organisms from the 3 stations were pooled. However, in several cases individuals of different sizes had to be pooled to get enough material for metal analysis. Dried macroinvertebrate samples were stored at room temperature until acid digestion. Samples of *Bythinia tentaculata* and Oligochaeta were digested soon after preparation, while samples of *Orconectes* sp., Chironomidae and *Gammarus fasciatus* were digested up to 4 months later.

The samples were digested with concentrated nitric acid (quality Baker "instra-analysed") in closed Teflon vessels using a 630W pressure controlled microwave sample preparation system (Model MDS-2000 of CEM Corporation). Two ml of nitric acid were used to digest samples of *Orconectes* sp., Chironomidae and *G. fasciatus*, and 3.75 ml for samples of *B. tentaculata* and Oligochaeta (bigger samples). Eight samples at a time were digested in the microwave system, including the acid blank and, at approximately every second run, the reference sample. Samples were heated at 100% power in two steps: 40 minutes at 85 psi, followed by 30 minutes at 130 psi. The period of time indicated for each step includes the time necessary to increase the pressure inside the vessels to that value. Once the digestion terminated, the vessels and their samples were allowed to cool down for a minimum period of 15 minutes. Digested samples were then transferred to small polypropylene centrifuge tubes, diluted to 15 ml with deionized distilled water and later delivered to the Analytical Method Development Section at the Geological Survey of Canada for metal analysis. The concentrations of Cd, Cr, Cu, Fe, Ni, Pb, and Zn were determined by ICP-MS and Hg concentrations by hydride-generation ICP-MS. For any digested sample that had a concentration value of a metal below the analytical detection limit, a figure of ½ times the detection limit was entered as the concentration value for that metal. This situation occurred in some cases for Ni and Cd concentrations in most taxa.

Quality assurance of the analytical methods was monitored by the analysis of NRCC certified reference material (lobster hepatopancreas TORT-2), of repeats, and by the inclusion of acid blanks. Contamination of acid blanks during digestion was detected at least once for most metals. In many cases, no corrections were needed. However, in other cases, the data had to be corrected, including reference material (corrected concentration value = observed concentration value - blank contamination value + 0.5 * the detection limit). The only important contamination problem occurred with Cr and Ni in most *B. tentaculata* and Oligochaeta samples. Other contamination problems that required corrections implicated Cr in many *G. fasciatus* samples and a low systematic contamination by Hg for all *G. fasciatus*, *Orconectes* sp. and Chironomidae samples. For the digestion procedure specific to *Orconectes* sp., the Chironomidae and *G. fasciatus*, recovery of Cd, Cr, Cu, Fe, Hg, Ni, and Zn from TORT-2 samples (n = 19) was 98, 128, 86, 98, 97, 90, and 92%, respectively. Recovery for Pb was 71% (n = 3, other samples were below the detection limit). Measured concentrations were within the 95% confidence interval of the certified mean value for each element, except Cr, Cu, Ni, and Zn. Measured concentrations in the certified reference material varied little for Cd, Cu, Hg, Ni, Zn (CV = 3 - 6%). However, Cr, Fe, and Pb concentrations were more variable (CV = 13 - 17%). For the digestion procedure specific to *B. tentaculata* and the Oligochaeta, recovery of Cd, Cr, Cu, Fe, Hg, Ni, and Zn was 98, >200, 88, 118, 72, 100, 96%, respectively (n = 3). Recovery of Pb could not be determined as concentrations were below detection limit. Measured concentrations were within the 95% confidence interval for Cd and Ni only. Measured concentrations in the certified reference material varied little for Cd, Cu, Hg, Zn (CV = 1 - 6%). However, Fe concentrations were more variable (CV = 16%), while Cr and Ni varied extremely (CV = 68 - 152%). For these later 2 metals, since measurements were highly imprecise, Cr and Ni concentrations in affected organisms were therefore considered unreliable and not presented here.

Statistical analyses

Prior to all parametric analyses, metal concentrations, sediment composition variables and body weight of organisms were log-transformed, and the data were plotted to confirm homogeneity of variance. Statistical results were deemed significant at $p < 0.05$. All statistical analyses were performed using SYSTAT® version 5.0.

Simple linear regressions were performed to determine the effect of the amount of silt and clay and of organic matter on sediment metal concentrations, as well as the effect of body size (measured as body weight) on metal concentrations in macroinvertebrates. The association between the proportion of silt and clay and the quantity of organic matter in sediments was examined by using a Pearson's correlation analysis. A Spearman's rank correlation analysis was performed to examine if metal concentrations exhibited a similar spatial distribution in sediments. Parametric and Kruskal-Wallis one-way analyses of variance (ANOVA) were used to investigate differences in metal bioaccumulation among macroinvertebrate taxa. The second of these two types of ANOVA was used when the log-transformed data were heteroscedastic.

To determine the influence of local sources, metal concentrations in sediments and macroinvertebrates were compared among study sites classified into three zones of potential impact (reference, most impacted and least impacted) according to their position relative to each of the following 2 groups of local industries: Domtar Fine Papers/ICI Forest Products and Courtaulds Fibers/Courtaulds Films (Table 1). The most impacted zone covers the area from downstream of either group of industries to the western end of Lake St. Francis, north of the St. Lawrence Seaway, while the least impacted zone covers the river reach south of the St. Lawrence Seaway from Ile Saint-Régis to Thompson Island in Lake St. Francis (Fig. 1). Study sites in the most impacted zone are located either directly in the north channel or in waters originating mostly from the north channel, while sites in the least impacted zone are situated in waters originating from both the north and south channels. It is important to note that study sites within the most impacted zone, when it is defined relative to the position of Domtar Fine Papers/ICI Forest Products, are also impacted by Courtaulds Fibers/Courtaulds Films, except for sites D14 and D15. Comparisons of metal concentrations among zones were performed using analyses of covariance (ANCOVA). In order to take into account any effect sediment composition (proportion of silt and clay, and quantity of organic matter) and organism size (body weight) can have on metal concentrations in sediments and macroinvertebrates, respectively, these variables were treated as covariates in the statistical analyses. The analyses of covariance were performed twice, first relative to the position of Domtar Fine Papers/ICI Forest Products, and then relative to the position of Courtaulds Fibers/Courtaulds Films. Also in order to evaluate the effect of local sources, analyses of covariance, with the proportion of fine inorganic particles as a covariate, were used to compare metal concentrations in downstream littoral sediments with concentrations measured upstream of the Moses-Saunders Dam. In all analyses of covariance, the complete ANCOVA model was first computed to check for the absence of interaction between the main factor and the covariate, and then the reduced model without the interaction term was computed. Statistical results presented in the text and in tables are based on the reduced model.

Results

Metal concentrations in sediments

In shallow littoral areas, sediments were mainly composed of sand, with the silt and clay fraction representing only a small proportion (Table 2). The mean amount of sand per site ranged from 38 to 87%, with a median of 78%, whereas the mean proportion of fine inorganic particles (silt and clay) varied between 11 and 60%, with a median of 17%. Only four sites had relatively high amounts of silt and clay: 3 with mean values of around 40% and 1 with 60%. The organic matter content of littoral sediments was also generally low (Table 2). Mean values per site ranged from 1.3 to 18.0%, with a median of 2.9%. All study sites had a mean quantity of organic matter lower than 6.2%, except 2 sites with 13.1% and 18.0%. Organic matter content and the proportion of fine inorganic particles were strongly and positively correlated ($r = 0.78$, $p = 0.0004$, $n = 16$; Fig. 2). Sediment composition data for all samples are included in Appendix 1.

Metal concentrations in littoral sediments were generally below the lowest effect level of the Ontario Provincial Sediment Quality Guidelines (Persaud et al., 1993) (Fig. 3). The median of mean concentration values per site was 0.23 µg/g for Cd, 10.2 µg/g for Cr, 8.5 µg/g for Cu, 9145 µg/g for Fe, 0.082 µg/g for Hg, 8.9 µg/g for Ni, 7.3 µg/g for Pb, and 44 µg/g for Zn. Sediments at a few study sites, however, had mean concentrations of one or several metals (Cd, Cu, Fe, Hg, Ni and Zn) just above the lowest effect level, and sediments at one site in particular (D14) had a high Hg concentration (mean of 1.12 µg/g) (Fig. 3). The Hg level at site D14, which is situated 1 km downstream of a known local point source of Hg, was at least one order of magnitude higher than the median of mean Hg concentration values per site (0.082 µg/g). Apart from site D14, these higher metal concentrations were observed at sites with the highest amounts of silt and clay or of organic matter. The highest mean concentrations of Cr and Pb, although below the lowest effect level, were also observed at one or several of those sites (Fig. 3). In addition, mean sediment metal concentrations were positively and often strongly correlated with one another (Table 3). Only the correlations between Fe-Cd, Ni-Cd and Pb-Ni were not significant. Most correlation coefficients (r_s) were above 0.6, and half were greater than 0.7. Thus, all metals showed a relatively similar spatial distribution. Sediment metal concentrations for all samples are given in Appendix 1.

Metal concentrations in littoral sediments were positively related to both the proportion of silt and clay (Fig. 4) and the quantity of organic matter (Fig. 5) (most relationships: $p < 0.0001$). 48 to 78% of the variation in Cd, Cr, Cu, Hg and Pb concentrations and 23 to 38% of the variation in Fe, Ni and Zn concentrations among sediment samples could be significantly explained by one of the two physico-chemical variables. The organic matter content of sediments explained the greatest amount of variation in Cd, Cu, Hg, Pb and Zn concentrations, while the proportion of fine inorganic particles explained the largest amount of variation for Cr and Fe. Both variables explained the same amount of variation in Ni concentrations. However, a statistical comparison for each metal of the residual mean square (RMS) of both regression models has shown, except for Cd, that the proportion of silt and clay predicted metal levels as well as the quantity of organic matter (F tests, $p > 0.05$; Cd: $p < 0.05$). Examination of the residual values of the regression models revealed that the Hg level observed at site D14 was much higher than expected from the two models (Fig. 4 and 5). Based on mean percent fine inorganic particles and mean percent organic matter, the predicted Hg concentration at site D14 would be 0.067 μg/g and 0.122 μg/g, respectively, instead of the observed mean value of 1.12 μg/g. Pb concentration at site D16 (1 sample) and Zn concentrations at sites D1, D2 and D16 (1 sample) were also higher than expected when metal concentrations are regressed against the quantity of organic matter (Fig. 5).

Metal concentrations in macroinvertebrates

Metal bioaccumulation in littoral macroinvertebrates was assessed for 5 taxa: the amphipod *Gammarus fasciatus*, the decapod *Orconectes* sp., the gastropod *Bythinia tentaculata*, the Chironomidae (midges) and the Oligochaeta (worms). Except for *G. fasciatus*, no taxon was sampled or represented in sufficient numbers or adequate size for metal analysis at all study sites, and due to analytical problems, no metal concentration data are available for any taxon at site D3.

Metal concentrations in each macroinvertebrate taxon varied among study sites (Table 4), except for Cu and Zn which showed less variability for *G. fasciatus*, *Orconectes* sp., and the

Chironomidae. As with sediments, Hg concentrations in invertebrates were high at study site D14 relative to any other site. For *G. fasciatus*, *Orconectes* sp., and the Chironomidae, Hg levels at that site were 10 times higher than the median of mean concentration values per site of 0.070, 0.086, and 0.098 µg/g, respectively. Although also elevated at site D14, Hg levels in the Oligochaeta were highest at site D15 situated a few kilometers downstream from site D14, possibly due to an extreme concentration value for one sample. In contrast, Hg levels in *B. tentaculata* were surprisingly low at site D14 ($n = 1$). No particularly elevated levels relative to other sites were observed for any other metal, except possibly for Pb concentrations in *Orconectes* sp. at site D7 and D16 ($n = 1$), and Cd levels in the Oligochaeta at site D5. Bioaccumulation data for all samples are presented in Appendix 2.

Compared to metal concentrations in sediments, littoral macroinvertebrates were generally found to have lower levels of Cd (except for *B. tentaculata*), Cr, Fe, Ni and Pb, and higher concentrations of Cu and Zn. Hg concentrations in invertebrates were similar to levels in the underlying sediments, except for slightly higher concentrations in *B. tentaculata* (Fig. 6).

Metal concentrations in invertebrates differed among taxonomic groups and these differences were metal specific (Fig. 6). The median of mean concentration values per site varied among taxa from 0.11 to 0.45 µg/g for Cd, 1.6 to 8.5 µg/g for Cr, 14 to 292 µg/g for Cu, 790 to 5044 µg/g for Fe, 0.070 to 0.155 µg/g for Hg, 1.3 to 4.8 µg/g for Ni, 0.7 to 5.0 µg/g for Pb, and 66 to 271 µg/g for Zn. The gastropod *B. tentaculata* accumulated the highest concentrations of Cd, Cu, Hg, and Zn, and with the Chironomidae and Oligochaeta, accumulated the highest levels of Fe and Pb. The Chironomidae also accumulated the greatest amounts of Cr and Ni. In contrast, the Chironomidae and Oligochaeta accumulated relatively low to intermediate levels of Cd, Cu, Hg, and Zn. Generally, the amphipod *G. fasciatus* and the decapod *Orconectes* sp. accumulated relatively low to intermediate metal concentrations. Chironomids and oligochaetes generally accumulated relatively similar metal concentrations, as did decapods and amphipods. These observed differences in metal bioaccumulation among taxonomic groups were found to be highly significant (one-way ANOVA, all metals except Hg: $p < 0.00001$; Kruskal-Wallis one-way ANOVA, Hg: $p = 0.0086$). However, since no metal concentration data were available for all study sites for any of the taxa besides *G. fasciatus*, the analysis was repeated with the data of sites

D8 and D14 (the only 2 study sites where metal levels were measured for all 5 taxa). In both cases, metal levels differed significantly among taxa (parametric or Kruskal-Wallis one-way ANOVA: site D8, $p < 0.00002$; site D14, $p < 0.009$) and the relative differences among taxonomic groups were generally very similar to those observed when all study sites are included (Fig. 6), except for unexpectedly low Hg concentrations in *B. tentaculata* at site D14 ($n = 1$).

The effect of organism size (measured as body weight) on metal concentrations in littoral macroinvertebrates was examined for *Orconectes* sp., the Chironomidae and *G. fasciatus*. These were taxa for which there were one (*Orconectes* sp., the Chironomidae) or several sites (*G. fasciatus;* 4 sites) with a sufficient number of samples to perform linear regressions of metal concentrations on body weight. Except for Zn, body weight usually had little effect on metal concentrations in those macroinvertebrates (Table 5). At the one site examined for *Orconectes* sp., organism size had only a significant negative effect on Cu levels. Similarly, body size of Chironomidae had only a significant negative effect on Zn concentrations. For *G. fasciatus*, body weight at 3 out of the 4 sites examined had only a significant positive effect on Zn concentrations. However, all relationships, except Cr, were significant at the fourth site (D8). A point to note is that, for any given metal, the slope of the relationship between concentrations and body weight was generally the same (positive or negative) at all four sites (results not shown).

Influence of local point sources

To determine the influence of Domtar Fine Papers/ICI Forest Products and Courtaulds Fibers/Courtaulds Films on metal levels in littoral sediments and macroinvertebrates, mean metal concentrations per site were compared among reference, most impacted and least impacted zones, as defined relative to each group of point sources (Table 1). These comparisons show that local sources had only an impact on sediment and macroinvertebrate Hg and Zn concentrations.

In sediments, Hg concentrations were higher in the zone most impacted by Domtar Fine Papers/ICI Forest Products (zone also impacted by Courtaulds Fibers/Courtaulds Films) (ANCOVA, proportion of silt and clay as a covariate: $p = 0.037$, organic matter content as a covariate: $p = 0.0097$; Fig. 7a), while Zn levels were higher in the zone most impacted by

Courtaulds Fibers/Courtaulds Films (with the two sites immediately below Domtar/ICI considered as reference) (ANCOVA, proportion of silt and clay as a covariate: $p = 0.016$, organic matter content as a covariate: $p = 0.0097$; Fig. 7b). To verify that the observed difference in Hg levels among zones was not only a result of the high concentration at site D14, I repeated the analysis without that site and found the difference in Hg levels to be still significant ($p < 0.01$). Concentrations of other metals differed little among zones ($p > 0.05$), except for higher Fe levels in the reference and most impacted zones relative to Courtaulds (ANCOVA, proportion of silt and clay as a covariate, $p = 0.041$).

For macroinvertebrates, Hg concentrations in Oligochaeta and Zn concentrations in Chironomidae were higher in the zone most impacted by Domtar Fine Papers/ICI Forest Products (zone also impacted by Courtaulds Fibers/Courtaulds Films), whereas Cr levels in *G. fasciatus* were higher at reference sites (Table 6). Although differences were not significant, Hg concentrations in other taxa were generally higher in the most impacted zone. Relative to Courtaulds Fibers/Courtaulds Films, Hg concentrations in *B. tentaculata* and Zn concentrations in *B. tentaculata* and *G. fasciatus* were higher in the most impacted zone (Table 6). For *B. tentaculata*, however, the Zn concentration at one ($n = 1$) out of the three reference sites was as high as levels from some of the sites in the most impacted zone. Zn levels in the Oligochaeta were higher in the most impacted zone; however, differences were not significant. Concentrations of other metals in macroinvertebrates differed little among zones ($p > 0.05$).

Metal concentrations in sediments were also compared to levels measured at eight littoral sites located upstream of the Moses-Saunders Dam in Lake St. Lawrence (Fig. 8). Since the purpose of this comparison was to determine the influence of local sources on metal concentrations in downstream sediments, the 3 downstream reference sites (D12, D13, and D11), which are located above local point sources but below the dam, were excluded. The effect of the proportion of silt and clay on metal levels was taken into account in the comparisons. Only Hg concentrations were found to be significantly higher downstream, while Cr and Fe levels were significantly higher upstream (ANCOVA, Hg: $p = 0.020$, Cr: $p = 0.0014$, Fe: $p = 0.0047$). To verify that the observed difference in Hg levels among upstream and downstream sediments was not only a result of the high concentration at downstream site D14, I repeated the analysis without that site. Without site D14, Hg concentrations were still significantly higher downstream ($p < 0.010$). Upstream sediment metal concentrations are given in Appendix 3.

Discussion

Metal levels in littoral sediments

Metal concentrations in sediments of shallow littoral areas were generally low and these low levels reflect the low fine particulate material (inorganic and organic) content of the largely sandy sediments. The occurrence of highly significant and often strong positive relationships between metal concentrations and the proportion of silt and clay and organic matter content strongly suggests that metals were associated with these particulate materials, and that, consequently, sediment composition played a dominant role in metal accumulation in littoral sediments. Other studies in the Great Lakes-St. Lawrence River system, including the Cornwall-Lake St. Francis area, have also shown a link between sediment composition and metal concentrations (e.g., Mudroch and Duncan, 1986; Poulton et al., 1988; Anderson, 1990; Carignan et al., 1994; Flessas, 1994; Richman, 1994). This is consistent with the traditional concept that, in aquatic sediments, metals are predominantly associated with the fine-grained fractions (< 63 μm) (Salomons and Förstner, 1984; Förstner, 1987; Moore et al., 1989). Relative to larger particles such as sand, fine particles of silt and clay have a higher surface to volume ratio and therefore adsorb greater amounts of contaminants per unit weight. Metals can also be associated with organic matter, and this association could be due to metal accumulation and deposition by phytoplankton (Sigg et al., 1987). Since, in general, littoral sediments had low amounts of fine inorganic and organic particles, metal concentrations are therefore expected to be low, unless the sediments were directly impacted by a local point source (e.g., site D14 discussed below). Further, the higher metal concentrations observed at a few sites, for example those exceeding the lowest effect level of the sediment quality guidelines (except Hg at site D14 and possibly Zn at D2), are most likely a result of the higher amounts of fine particulate materials in the sediments of those sites. That sediment composition significantly explains part of the variation in metal concentrations, however, does not exclude the possibility for the influence of local sources, since the residual variation in metal concentrations could potentially be related to the proximity of a local source.

The much higher Hg concentration at study site D14, however, can not be adequately explained by the composition of the sediments. Sediments at that site had only a moderate amount of organic matter (mean of 5.9%) and a low proportion of silt and clay (mean of 17%). Yet, the concentration of Hg (mean of 1.12 μg/g) was found to be 10 times higher than the value expected from the regression models of Hg concentrations on the amount of fine inorganic particles and of organic matter. The occurrence of high Hg levels in that area was also observed by Kauss et al. (1988), Anderson (1990) and Richman (1994). For example, a mean Hg concentration of 3.26 μg/g in the sediments of a nearby shallow site was found by Richman (1994). The high Hg enrichment at site D14 therefore strongly suggests an impact from a local point source (present or historical). Study site D14 is in fact situated 1 km downstream of Domtar Fine Papers and ICI Forest Products, two known historical sources of Hg (St. Lawrence RAP Team, 1992).

The low metal concentrations in littoral sediments contrast with levels reported by Poulton (1994) and Richman (1994) for deep open-water (up to 13 m) and relatively deep near-shore (1.5 - 4 m) sites in the same area. On average, littoral sediments were found to have lower metal concentrations than the sediments of those sites (Table 7). Cd concentrations, however, were similar to levels reported by Poulton (1994). In particular, concentrations at many of the sites assessed by Richman (1994) exceeded, for most metals, the lowest effect level of the sediment quality guidelines. However, these differences in metal levels between littoral sediments and deeper sites were, except for Hg, mainly attributable to differences in sediment composition, as the proportion of silt and clay significantly explained most of the variation in metal concentrations among the three studies (Table 8 and Fig. 9). In fact, littoral sediments had much lower amounts of fine inorganic particles, median of 17%, compared to medians of 42% and 68% at sites assessed by Poulton (1994) and Richman (1994), respectively. The smaller proportion of fine particles in littoral sediments is consistent with the fact that this section of the St. Lawrence River is fast flowing and, as a consequence, the littoral zone is considered to be a high energy area rather than a zone of accumulation of fine-grained material (Sloterdijk, 1991). The spatial distribution of metals in surficial sediments of the Cornwall area, therefore, mainly reflected the sorting of fine particles toward the deeper parts of the river (Poulton et al., 1988).

However, even after correction for the effect of the amount of silt and clay on metal concentrations, small significant differences in metal levels among the three studies were generally detected (Table 8), with littoral sediments having usually lower metal concentrations (Fig. 9). Except for Hg, these differences could be due to natural variability or to variations in analytical techniques and performance (e.g. the use of a "coarser" method for the determination of particle size distribution in our study). In the case of Hg, however, sediment composition is not the most important factor in explaining the difference in concentrations among studies (Table 8). Hg concentrations (corrected for the proportion of silt and clay; Fig. 9) at sites assessed by Richman (1994) were on average higher by about one order of magnitude compared to sites assessed by Poulton (1994) and to littoral sediments (this study). This Hg enrichment was highest at sites close to known local sources of Hg (also true for site D14 in the present study). Therefore, this suggests that local sources contributed to the Hg contamination of sediments over a substantial area.

Metal concentrations in littoral sediments were also compared to levels reported for several contaminated areas of the Great Lakes (Jaagumagi and Persaud, 1992) (Table 9). Median concentrations for most metals were toward the lower end of the ranges of values for those areas. Only Hg concentrations (median of 0.08 μg/g corresponding to the 36th percentile of the Great Lakes values), and to a smaller extent Ni (8.9 μg/g, 21%) and Zn (44 μg/g, 22%), approached the median concentration values for the Great Lakes areas. This comparison shows that littoral sediments in the Cornwall region were less contaminated than the sediments of industrialized and urbanized areas in the Great Lakes, consistent with the finer nature of those sediments (based on a subset of the data). The relatively higher mercury concentrations in littoral sediments, however, suggest a problem of Hg contamination in the Cornwall region.

Metal levels in littoral macroinvertebrates

Littoral macroinvertebrates showed metal-specific differences in bioaccumulation among taxonomic groups. These differences were evident among 3 groups of taxa: chironomids/oligochaetes, amphipods/decapods, and gastropods. These differences in metal concentrations could reflect variation in feeding habits and, with that, variation in the degree of association with the sediments, but they could also reflect the influence of gut content and variation in physiological processes (Smock, 1983; Amiard et al., 1987; Krantzberg and Stokes, 1989; Timmermans et al., 1989; Hare, 1992). Chironomids and oligochaetes bury themselves in sediments and are, therefore, very closely associated with sediment-bound metals. Oligochaetes ingest sediments along with detritus, while chironomids feed on particles on the surface of the sediments or trapped on their tube walls (Jackson, 1988; Bendell-Young et al., 1994). In contrast, gammarids and juvenile decapods are both detritivores and herbivores, while *B. tentaculata* is an organism that feeds on periphyton (Thorp and Covich, 1991; Amyot et al., 1994; Flessas, 1994). Since bottom sediments are considerably enriched in metals with respect to overlying waters, macroinvertebrates that are more closely associated with sediments are more likely to accumulate higher metal concentrations than others living in the same general habitat (Smock, 1983; Jackson, 1988). Indeed, chironomids and oligochaetes accumulated higher Cr, Fe, Ni, and Pb concentrations than gammarids and decapods. Cd, Hg, and Zn levels were, however, relatively similar among these four taxa, while Cu levels were higher in gammarids and decapods. Another possibility, then, is the influence of gut contents on metal concentrations in invertebrates. For example, Amyot et al. (1994) showed that Cr, Fe, Ni, and Pb concentrations in gut content affected metal concentrations in *G. fasciatus*, while Cu, Zn, and Cd had no significant effect (Hg not investigated). If this is also true for chironomids and oligochaetes, then the possibly greater proportion of sediments in the gut of these invertebrates could contribute to higher levels of Cr, Fe, Ni, and Pb, while no effect would be expected on Cd, Cu, and Zn concentrations.

Cu levels in gammarids and decapods reflect the presence of the copper-containing pigment haemocyanin in their haemolymph. Depledge (1987) estimated the copper bound in enzymes and haemocyanin to be around 80 µg/g dry weight of whole body of the marine decapod *Carcinus maenas*, approximately what we mesured in gammarids and decapods.

Chironomids and oligochaetes also accumulated higher Fe and Pb levels than the herbivore *B. tentaculata*; however, Cd, Cu, Hg, and Zn concentrations were found to be highest in this gastropod species, compared to any of the other taxa. This result could possibly indicate a greater metal bioavailability from periphyton, or marked differences in the physiological processes of metal uptake and regulation among these organisms. The higher Cu concentrations in gastropods are consistent with the fact these invertebrates also possess haemocyanin in their haemolymph. These differences in metal bioaccumulation among taxonomic groups indicate that the impact of sediment contamination on the littoral macroinvertebrate community (i.e. metal bioavailability) differs among taxa. This demonstrates the importance of assessing metal accumulation in organisms from different feeding habits, including the mode of association with the sediments, and physiology in order to adequately assess the impact of sediment contamination on macroinvertebrate communities (Smock, 1983; Moore et al., 1991; Clements and Kiffney, 1994).

The comparison of metal concentrations in organisms with levels in the underlying sediments showed that Cu and Zn were accumulated above sediment levels in all five taxa, whereas Cd (except in *B. tentaculata*), Cr, Fe, Ni and Pb concentrations were generally lower than levels in the sediments. Timmermans et al. (1989) also observed a similar pattern in a littoral foodweb, including the occurrence of higher Cd concentrations in a gastropod species. Cu and Zn are essential metals that can be regulated by aquatic macroinvertebrates (Amiard et al., 1987; Krantzberg, 1989; Krantzberg and Stokes, 1989). Therefore, this could explain their higher concentrations in all taxa compared to the underlying sediments. Hg concentrations in all taxa were found to be similar to levels in the sediments, except for higher concentrations in *B. tentaculata*. This suggests a higher bioavailability of Hg relative to the other metals, and this may possibly be related to the potential of this metal to biomagnify up the food chain (Jackson, 1988; Allard and Stokes, 1989; Bryan and Langston, 1992).

The high Hg concentrations in sediments at study site D14 were paralleled by elevated levels in all macroinvertebrate taxa, except *B. tentaculata*. Hg levels were surprisingly low in this organism; however, this observation is based on only one sample of several individuals. As with sediments, these high Hg levels in invertebrates at site D14 strongly suggest an impact from a local source. As discussed above, study site D14 is located close to two known historical sources of Hg (St. Lawrence RAP Team, 1992). The similar biological response of most taxa to Hg enrichment at that site suggests that, when organisms are exposed to high levels in sediments, bioaccumulation is a community-level response (Luoma, 1989; Moore et al., 1991; Cain et al., 1992).

Metal concentrations in littoral macroinvertebrates were generally similar or lower than levels described for the same or related taxa from low to moderately contaminated environments (unless indicated otherwise, organisms in those studies were not depurated). Concentrations of Cd, Cr, Cu, Fe, Ni, Pb and Zn in *G. fasciatus* were comparable to or lower than levels reported for the same species (Table 10; Hg not investigated) from moderately contaminated littoral areas of Lake St. Louis and Lake St. Pierre, two fluvial lakes on the St. Lawrence River impacted by point sources of metals (Amyot et al., 1994). In their study, Amyot et al. (1994) concluded that the highest observed metal concentrations were similar to levels described for other gammarids from moderately contaminated aquatic systems. Cd, Cu, Pb and Zn levels in *G. fasciatus*, oligochaetes, and chironomids were generally similar to concentrations in *G. pulex*, *Tubifex* sp. (Oligochaeta), and *Chironomus* spp. (Chironomidae), respectively (gut content purged), from low contaminated littoral areas of two European lakes (van Hattum et al., 1991). In that study, metal levels in sediments were similar to concentrations in the present study. Also, Cd, Cu, Fe and Zn concentrations in chironomids were lower (Cd was one order of magnitude lower) than levels described for these organisms from Canadian Shield lakes impacted by acid rain (Bendell Young and Harvey, 1991). Concentrations of Cu, Fe and Pb in *B. tentaculata*, however, were greater than values reported for this gastropod species from Lake St. Louis, comparable for Zn and lower for Cd (Flessas, 1994; Table 10). In that study, organisms were sampled from the same sites established for the study of Amyot et al. (1994). Finally, Cd, Cu, Ni and Zn levels in *Orconectes* sp. were similar to or lower (Cd was one order of magnitude lower) than reported values in the crayfish *Cambarus bartoni* (without gut content) from a moderately contaminated lake in Northeastern Ontario (Alikhan et al., 1990). Fe levels were much higher in *Orconectes* sp., however, possibly due in part to the influence of gut content.

Metal concentrations in Oligochaeta and Chironomidae were also compared to reported levels in benthic macroinvertebrates (samples mostly composed of oligochaetes and chironomids) from the same areas of the Great Lakes discussed above (Jaagumagi and Persaud, 1992; Table 9). Pooled Fe concentrations in Oligochaeta and Chironomidae (median of 5035 µg/g corresponding to the 89th percentile of the Great Lakes values) were higher than the median level for the Great Lakes areas, similar for Ni (4.8 µg/g (chironomids only), 54th percentile) and Cr (8.5 µg/g (chironomids only), 53th percentile), and lower for the other five metals. Except for Fe, this comparison shows that oligochaetes and chironomids from littoral areas (this study) were as contaminated (Cr, Ni) or less contaminated (Cd, Cu, Hg, Pb, Zn) than benthic macroinvertebrates from contaminated areas of the Great Lakes. Jaagumagi and Persaud (1992) also assessed the bioaccumulation of metals in macroinvertebrates (samples also mostly composed of oligochaetes and chironomids) from deep open-water and relatively deep near-shore sites of the Cornwall area. Metal levels in the sediments of those sites, which were also sampled by Richman (1994), were similar to those reported by that author (see above). Comparison with these data (Table 11) shows that oligochaetes and chironomids from littoral areas accumulated higher or similar Cr, Fe, Ni and Zn levels, and lower Cd, Cu, Hg and Pb concentrations than organisms of deeper sites. Fe levels were particularly much higher in invertebrates from the littoral zone. Richman (1994) also assessed Hg concentrations in chironomids from deeper sites and reported mean Hg levels per site ranging from 0.05 to 0.43 µg/g (median of 0.15 µg/g, values transformed from on a wet to dry weight basis using the formula of Table 9, gut content purged). Concentrations in littoral chironomids (this study; median of 0.10 µg/g) were only slightly lower than these levels, and this despite the fact that Hg concentrations in littoral sediments were much lower.

Results of these comparisons with contaminated areas of the Great Lakes and deeper areas in the Cornwall region are not consistent with the relatively much lower metal concentrations found in littoral sediments compared to the sediments of those areas. These conflicting results suggest that metal bioavailability (at least relative to chironomids and oligochaetes) differs between sandy littoral sediments and areas of finer sediments. Apart from variations in geochemistry which can affect the availability of metals from sediments (Tessier and Campbell, 1987; Luoma, 1989), the feeding habit of those organisms may play a role. Since chironomids and oligochaetes selectively feed on small particles (Jackson, 1988; Bendell-Young et al., 1994), and because metals are predominantly associated with these particles (see above), ingestion of fine particulate material from the littoral sediments could possibly result in exposure to metals approaching that of finer and more contaminated sediments. However, since Fe and Ni levels are higher in organisms from littoral areas, variation in gut content could affect those levels, if the proportion of sediments in their digestive tract was higher than in organisms of those areas.

Conversion of metal concentrations reported in these studies from wet weight to dry weight in order to perform the comparisons, and differences in analytical techniques and performance may also account for part of the observed differences in macroinvertebrate metal concentrations among the littoral zone and those areas.

Influence of local point sources

Comparisons of metal concentrations among reference, most impacted, and least impacted sites, as defined relative to their position to Domtar Fine Papers/ICI Forest Products and Courtaulds Fibers/Courtaulds Films, have shown that sediments and some macroinvertebrate taxa had, on average, significantly higher Hg and Zn concentrations at the most impacted sites. As well, comparison of metal concentrations among upstream (in Lake St. Lawrence) and downstream littoral sites showed that downstream sediments had significantly higher Hg concentrations. With the high Hg levels in sediments and in most invertebrate taxa at study site D14, the results of these comparisons suggest that local sources of Hg and Zn have contributed to the contamination of littoral sediments and macroinvertebrates in the Cornwall area. The results of these analyses also suggest, for the remaining metals, that concentrations in littoral sediments and macroinvertebrates reflected the influence of upstream sources (Lake Ontario) or natural levels.

The spatial distribution of Zn levels in sediments and aquatic macroinvertebrates is consistent with the results of previous studies that had identified Courtaulds Fibers, before its closure in 1992, as a major source of Zn to the St. Lawrence River (Kauss et al., 1988; Anderson, 1990; St. Lawrence RAP Team, 1992; Richman, 1994). In the present study, concentrations in sediments and in two taxa (*B. tentaculata* and *G. fasciatus*) were found to be significantly higher in the most impacted zone of Courtaulds. However, Zn levels in *B. tentaculata* may not differ significantly, on average, from concentrations in the reference zone because of higher concentrations at one ($n = 1$) out of the three reference sites. Still, levels at the other two sites were lower than levels in the least impacted zone. Zn concentrations in oligochaetes, although not significant, were also found to be higher at a few sites downstream of Courtaulds. An exception to this is the higher than expected Zn tissue levels in chironomids at the two downstream sites closest to Domtar Fine Papers/ICI Forest Products (levels similar to concentrations downstream of Courtaulds). However, neither of these two industries, nor Cornwall Chemicals which discharges its effluents through the same outfall, are major contributors of Zn to the area (St. Lawrence RAP Team, 1992).

The spatial distribution of Hg concentrations in sediments and macroinvertebrates is also consistent with previous studies that have indicated Domtar Fine Papers/ICI Forest Products and Courtaulds Fibers/Courtaulds Films as major sources of Hg in this region (Kauss et al., 1988; Anderson, 1990; St. Lawrence RAP Team, 1992; Richman, 1994). As discussed above, sediments and most taxa at site D14, which is situated 1 km downstream of Domtar Fine Papers/ICI Forest Products, showed high Hg concentrations relative to any other sites. Hg concentrations in sediments and in oligochaetes were also significantly higher in the most impacted zone downstream of Domtar Fine Papers/ICI Forest Products (zone also impacted by Courtaulds Fibers/Courtaulds Films) and in *B. tentaculata* in the most impacted zone downstream of Courtaulds Fibers/Courtaulds Films. Although not significant, Hg concentrations in the other taxa were generally higher at the most impacted sites. Finally, sediments downstream of these local sources were found to have significantly higher Hg levels than upstream sediments (in Lake St. Lawrence).

Although I can not exclude an impact of the sewage treatment plant on metal levels in sediments and macroinvertebrates strictly based on my study design (since no study sites were located between that plant and Courtaulds, and since most of the sites in the most impacted zone are situated below it), an important effect is highly unlikely because the sewage treatment plant was never identified as a major source of Hg and Zn to the St. Lawrence River. Moreover, the area of highly contaminated sediments downstream of Courtaulds is situated upstream of the sewage treatment plant.

In this study, I did not find any evidence that local sources had influenced Pb and Cu levels in sediments, or in macroinvertebrates, as was suggested by Richman (1994), although Pb concentrations seemed to be slightly higher in sediments and in a few taxa at study site D16, located 2 km downstream of Courtaulds Fibers/Courtaulds Films. It is important to note, however, that Richman (1994) only found higher than expected Pb and Cu concentrations in sediments at a site just below the shore based outfall of Courtaulds Fibers. Since none of my study sites were situated in that area, my results do not contradict the possibility of a very localized impact on environmental Pb and Cu levels. That no evidence was found that local sources of metals had significantly contributed to Cd, Cr, Fe and Ni contamination of littoral sediments and macroinvertebrates is consistent with previous studies which concluded that there were no important local sources of these metals in the area, and that consequently levels in the Cornwall region reflected upstream inputs (anthropogenic or natural) from Lake Ontario (Anderson, 1990; Richman, 1994).

Conclusions

This study showed that metal concentrations in littoral sediments were generally low. These low concentrations were most likely attributable to the small amount of fine inorganic and organic particles in the predominantly sandy sediments, as metals are mainly associated with these fine particulate materials. Sediment composition explained in part the higher metal levels observed at some study sites, except for the high Hg concentration at site D14 which suggested an impact from local sources. Metal concentrations in littoral sediments were lower than levels reported for contaminated areas of the Great Lakes. However, the relatively elevated Hg levels in littoral sediments suggested a problem of Hg contamination in the Cornwall area. Once corrected for the proportion of silt and clay, metal concentrations in littoral sediments were still slightly lower than in finer sediments of deeper sites in the area assessed in previous studies, possibly due to natural variability and to variation in analytical techniques and performance. For Hg, however, the important difference in concentrations among studies suggested that local sources had contributed to the Hg contamination of sediments over a substantial area. Nevertheless, the spatial distribution of metals in surficial sediments of the Cornwall area mainly reflected (except for Hg) the sorting of fine particles toward the deeper parts of the river.

Consistent with the low concentrations in sediments, metal concentrations in littoral macroinvertebrates were lower or similar to levels described for the same or related taxa from low to moderately contaminated aquatic environments, including two fluvial lakes on the St. Lawrence River. However, the high Hg concentration in the sediments of site D14 was reflected in elevated concentrations in most taxa, therefore also suggesting an impact from local sources. Littoral macroinvertebrates showed metal-specific differences in bioaccumulation among taxonomic groups, and these differences were likely related to variation in feeding habit (including the degree of association with the sediments), to the influence of gut content, and to variation in physiological processes. Cu and Zn were accumulated by all taxa at concentrations higher than in the underlying sediments, likely due to the ability of aquatic macroinvertebrates to regulate these nutritionally essential metals. For Hg, concentrations in invertebrates were similar, and even slightly higher in one taxon, to levels in sediments. This result could reflect the potential of this metal to biomagnify in food webs. When compared to contaminated areas of the Great Lakes and deeper areas in the Cornwall region, concentrations of Cr, Fe, Ni, and Zn in chironomids and oligochaetes were similar or higher than levels in benthic organisms from those areas, and this despite the relatively much lower metal concentrations in littoral sediments. These conflicting results could be due to the feeding habit of these invertebrates and to variation in gut content.

This study showed that local sources of Hg and Zn contributed to the contamination of littoral sediments and macroinvertebrates in the Cornwall area. However, the low concentrations in sediments and organisms suggest that these local sources had only a small impact, except for elevated Hg levels in sediments and invertebrates immediately downstream of Domtar Fine Papers/ICI Forest Products. A similar important impact immediately below Courtaulds can not be excluded, however, since we did not measure metal concentrations in sediments and invertebrates in that area. Results of these analyses also suggested that concentrations of other metals in littoral sediments and macroinvertebrates reflected the influence of upstream sources (Lake Ontario) or natural levels, although a highly localized impact of Pb and Cu downstream of Courtaulds could not be excluded for the same reason just mentioned. With the closures of Courtaulds Films in 1989 and Courtaulds Fibers in 1992, a major source of Zn and Hg has been eliminated in the Cornwall area. As well, with the abandonment of the use of mercury-based slimicides by Domtar Fine Papers in 1970 and of the use of a mercury cell process at ICI Forest Products in 1994, these industries are no longer significant sources of Hg in the area. Therefore, deposition of new sediments in the littoral zone should gradually restore Hg and Zn concentrations to levels reflecting upstream inputs. However, the highly contaminated area described in previous studies just below Courtauld's effluents could still represent a potential source of Hg and Zn to the littoral zone, through resuspension and deposition of contaminated sediments. Finally, further study of the area below Domtar Fine Papers/ICI Forest Products should be initiated to characterize the full extent of the Hg contamination I and other researchers have observed in that area and to investigate Hg transfer to aquatic macroinvertebrates.

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Table 1. Classification of study sites into zones of potential impact according to their position relative to local sources of metals. Within each zone, sites are ordered from upstream to downstream.

Domtar Fine Papers and ICI Forest Products			Courtaulds Fibers and Courtaulds Films		
Reference	Most impacted[1]	Least impacted	Reference	Most impacted	Least impacted
D12	D14	D6	D12	D16	D6
D13	D15	D9	D13	D5	D9
D11	D16	D3	D11	D2	D3
	D5	D4	D14	D8	D4
	D2	D7	D15	D1	D7
	D8			D10	
	D1				
	D10				

[1] Except for D14 and D15, these sites are also impacted by Courtaulds Fibers/Courtaulds Films

TABLE 2. Particle size distribution and organic matter content of littoral sediments. Values are mean % (dry weight) per site; standard errors (SE) are given in parentheses. All study sites: $n = 3$, except D1 and D2: $n = 1$, D13 (for particle size only) and D16: $n = 2$. Study sites are ordered from upstream to downstream within the reference, most impacted and least impacted zones (see Table 1).

Study site	Particle size: Gravel (>2mm)		Particle size: Sand (63µm-2mm)		Particle size: Silt and Clay (<63 µm)		Organic matter	
D12	6.2	(6.0)	82.6	(6.0)	11.2	(0.5)	1.4	(0.3)
D13	0.0	(0.0)	87.0	(3.6)	13.1	(3.6)	1.4	(0.5)
D11	0.3	(0.2)	57.5	(6.4)	42.2	(6.5)	6.2	(1.0)
D14	5.8	(2.1)	77.2	(3.3)	17.0	(5.5)	5.9	(1.1)
D15	1.8	(0.6)	37.8	(5.4)	60.3	(5.3)	13.1	(0.2)
D16	4.5	(2.7)	82.0	(1.3)	13.6	(1.4)	1.4	(0.1)
D5	2.2	(1.0)	82.0	(3.5)	15.8	(4.4)	1.4	(0.4)
D2	7.7		48.8		43.5		4.1	
D8	2.1	(0.7)	81.7	(5.4)	16.3	(5.1)	3.3	(0.2)
D1	5.7		79.0		15.3		1.3	
D10	0.5	(0.2)	83.1	(2.7)	16.3	(2.5)	5.6	(1.0)
D6	0.4	(0.2)	74.2	(5.6)	25.4	(5.8)	2.4	(0.4)
D9	1.7	(0.7)	71.4	(4.7)	26.8	(5.2)	4.1	(0.7)
D3	1.4	(0.7)	60.7	(15.0)	37.9	(15.2)	18.0	(6.8)
D4	7.0	(4.2)	78.3	(3.4)	14.6	(1.0)	2.2	(0.1)
D7	0.9	(0.1)	78.0	(6.3)	21.0	(6.3)	2.6	(0.8)

TABLE 3. Matrix of Spearman's rank correlation coefficients for mean metal concentrations in littoral sediments. The critical values for statistical significance are 0.50 at $p \leq 0.05$, 0.64 at $p \leq 0.01$, and 0.76 at $p \leq 0.001$ ($n = 16$).

	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
Cd	1.00							
Cr	0.58	1.00						
Cu	0.81	0.79	1.00					
Fe	0.49	0.87	0.79	1.00				
Hg	0.71	0.63	0.83	0.69	1.00			
Ni	0.37	0.79	0.57	0.71	0.55	1.00		
Pb	0.68	0.67	0.76	0.64	0.79	0.44	1.00	
Zn	0.55	0.85	0.79	0.83	0.80	0.74	0.68	1.00

TABLE 4. Whole-body metal concentrations (µg/g dry weight) and body weight (mg dry weight) of littoral macroinvertebrates. Values are means per site; standard errors (SE) are given in parentheses. *n* is the number of samples per site. No data are available at sites with no values. Study sites are ordered from upstream to downstream within the reference, most impacted and least impacted zones (see Table 1).

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D12	0.26 (0.036)	2.7 (0.13)	53 (2.0)	582 (62)	0.06 (0.005)	1.7 (0.30)	1.3 (0.16)	51 (0.8)	1.8 (0.37)	7
D13	0.13 (0.006)	3.3 (0.61)	68 (1.4)	1200 (63)	0.08 (0.004)	2.1 (0.21)	0.6 (0.04)	66 (0.8)	2.3 (0.22)	18
D11	0.14 (0.009)	3.4 (0.15)	69 (1.3)	1725 (51)	0.07 (0.005)	1.5 (0.10)	1.1 (0.05)	62 (1.6)	1.7 (0.26)	6
D14	0.07 (0.006)	1.7 (0.08)	52 (0.7)	651 (44)	0.61 (0.016)	0.8 (0.07)	1.3 (0.08)	59 (0.7)	1.7 (0.14)	12
D15	0.14 (0.037)	2.2 (0.14)	46 (0.9)	755 (67)	0.12 (0.003)	1.0 (0.35)	1.6 (0.47)	53 (1.5)	1.7 (0.17)	11
D16	0.15 (0.019)	2.4 (0.07)	51 (1.0)	1141 (36)	0.11 (0.007)	1.3 (0.13)	1.8 (0.21)	70 (2.1)	1.5 (0.36)	5
D5	0.45 (0.057)	2.4 (0.10)	69 (1.9)	927 (61)	0.06 (0.003)	1.4 (0.10)	0.8 (0.04)	64 (1.5)	3.0 (0.47)	7
D2	0.11 (0.006)	3.8 (0.39)	63 (9.3)	1738 (173)	0.07 (0.006)	2.2 (0.26)	1.6 (0.18)	85 (5.3)	2.4 (0.80)	3
D8	0.38 (0.020)	2.3 (0.07)	60 (0.8)	790 (42)	0.09 (0.004)	1.4 (0.09)	0.9 (0.04)	67 (0.8)	2.3 (0.25)	19
D1	0.07	2.1	96	409	0.13	1.3	0.6	81	1.7	1
D10	0.06 (0.010)	2.3 (0.21)	60 (2.2)	945 (71)	0.05 (0.002)	1.3 (0.06)	0.9 (0.08)	66 (1.2)	1.9 (0.38)	5
D6	0.21 (0.077)	2.0 (0.05)	51 (2.0)	971 (21)	0.06 (0.002)	1.1 (0.08)	0.8 (0.05)	58 (1.0)	1.2 (0.24)	3
D9	0.17 (0.011)	1.8 (0.03)	61 (1.9)	721 (32)	0.05 (0.001)	0.9 (0.09)	0.8 (0.03)	62 (2.1)	2.0 (0.32)	6
D4	0.28 (0.024)	1.8 (0.12)	66 (4.6)	532 (55)	0.07 (0.007)	1.3 (0.10)	0.7 (0.17)	71 (2.6)	4.0 (1.09)	3
D7	0.46 (0.055)	2.6 (0.12)	60 (1.9)	790 (51)	0.07 (0.004)	1.3 (0.10)	0.9 (0.09)	67 (2.5)	2.6 (0.67)	4

TABLE 4. Continued.

(b) *Orconectes* sp.

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D12	0.15 (0.010)	1.5 (0.30)	58 (2.5)	887 (206)	0.08 (0.004)	1.3 (0.37)	0.6 (0.20)	70 (1.0)	23 (6.5)	2
D13										
D11	0.11 (0.010)	1.9 (0.16)	90 (2.0)	1268 (119)	0.09 (0.013)	1.3 (0.42)	0.6 (0.03)	72 (3.3)	29 (4.1)	3
D14	0.04 (0.008)	1.1 (0.08)	58 (4.9)	482 (97)	0.87 (0.031)	0.5 (0.10)	0.7 (0.20)	67 (1.0)	41 (8.4)	2
D15	0.03	1.4	64	815	0.18	0.3	0.6	69	46	1
D16	0.06	2.8	53	1473	0.17	1.3	2.2	81	25	1
D5	0.14	1.6	81	870	0.07	1.3	0.7	73	6	1
D2										
D8	0.12 (0.010)	1.8 (0.12)	48 (1.8)	906 (58)	0.10 (0.004)	1.5 (0.16)	0.8 (0.05)	63 (1.3)	46 (4.6)	13
D1										
D10	0.06	1.9	52	811	0.06	0.6	0.8	65	26	1
D6										
D9										
D4	0.11	1.3	56	540	0.07	1.1	0.5	71	9	1
D7	0.17 (0.017)	1.7 (0.07)	75 (3.9)	799 (57)	0.08 (0.007)	1.3 (0.07)	2.4 (1.47)	76 (2.3)	7 (2.1)	2

Table 4. Continued.

(c) *Bythinia tentaculata*

Site	Cd	Cu	Fe	Hg	Pb	Zn	Body weight	*n*
D12	0.95	565	4091	0.17	2.7	309	5.0	1
D13	0.35 (0.014)	183 (5)	3060 (364)	0.09 (0.009)	1.4 (0.21)	155 (9)	7.4 (0.23)	3
D11								
D14	0.55	212	2581	0.10	1.5	175	7.2	1
D15								
D16	0.44 (0.002)	569 (45)	4516 (138)	0.39 (0.046)	6.0 (0.08)	299 (8)	5.4 (0.36)	2
D5								
D2	0.25 (0.013)	304 (25)	2886 (92)	0.20 (0.002)	2.8 (0.10)	323 (2)	4.3 (0.14)	2
D8	0.89 (0.031)	652 (47)	2540 (294)	0.27 (0.015)	3.1 (0.36)	439 (3)	4.1 (0.10)	3
D1	0.40	656	3504	0.36	5.0	538	4.0	1
D10	0.21	270	3590	0.15	2.7	300	6.6	1
D6	0.46 (0.007)	218 (3)	4202 (688)	0.13 (0.000)	2.5 (0.29)	202 (23)	5.0 (0.75)	2
D9	0.89 (0.112)	280 (16)	2687 (432)	0.14 (0.011)	2.9 (0.44)	237 (19)	7.8 (0.53)	2
D4	0.30 (0.013)	154 (12)	2545 (16)	0.11 (0.002)	1.8 (0.02)	140 (2)	4.5 (0.25)	2
D7	0.80 (0.080)	391 (25)	3823 (84)	0.16 (0.012)	3.6 (0.03)	243 (11)	3.5 (0.72)	3

TABLE 4. Continued.

(d) Chironomidae

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D12	0.26 (0.010)	11.7 (0.4)	16 (0.4)	7707 (239)	0.11 (0.003)	7.2 (0.2)	5.7 (0.2)	72 (1.5)	1.1 (0.10)	6
D13	0.19 (0.025)	11.5 (1.1)	18 (0.7)	7631 (824)	0.07 (0.007)	6.9 (0.6)	3.8 (0.3)	67 (3.5)	1.2 (0.13)	6
D11										
D14	0.14 (0.024)	7.8 (1.6)	19 (0.8)	4151 (405)	1.14 (0.443)	4.2 (0.5)	7.4 (0.8)	79 (4.5)	1.3 (0.27)	3
D15	0.07 (0.005)	2.8 (0.8)	15 (3.5)	1743 (293)	0.06 (0.020)	1.4 (0.7)	1.4 (0.7)	90 (18.3)	1.1 (0.24)	2
D16	0.12 (0.068)	7.1 (1.4)	15 (1.1)	4521 (937)	0.17 (0.008)	4.2 (1.1)	5.7 (1.5)	87 (0.6)	1.5 (0.77)	2
D5										
D2	0.03	5.0	17	2643	0.10	3.1	3.4	102	2.8	1
D8	0.22 (0.028)	10.3 (1.7)	18 (0.6)	5543 (690)	0.12 (0.008)	5.8 (0.7)	6.0 (0.7)	77 (2.1)	0.9 (0.09)	8
D1										
D10										
D6	0.14 (0.004)	8.6 (0.7)	15 (0.2)	5096 (377)	0.05 (0.004)	5.3 (0.5)	3.7 (0.3)	69 (2.6)	1.5 (0.32)	5
D9	0.21 (0.009)	8.5 (0.6)	18 (0.8)	3901 (320)	0.07 (0.002)	4.4 (0.1)	4.2 (0.2)	72 (2.0)	0.8 (0.14)	7
D4										
D7	0.32	14.8	18	6218	0.10	6.4	7.0	69	1.0	1

TABLE 4. Continued.

(e) Oligochaeta

Site	Cd	Cu	Fe	Hg	Pb	Zn	Body weight	*n*
D12								
D13								
D11	0.26	16	8717	0.07	6.5	73	42	1
D14	0.06 (0.026)	9 (2.8)	2415 (430)	0.21 (0.045)	2.7 (0.63)	87 (11)	57 (12)	3
D15	0.16 (0.041)	7 (0.6)	4037 (686)	0.43 (0.366)	4.4 (0.60)	63 (1)	81 (7)	4
D16								
D5	0.58 (0.044)	14 (1.6)	5112 (258)	0.09 (0.004)	3.2 (0.29)	124 (16)	26 (6)	2
D2								
D8	0.21 (0.021)	16 (0.9)	5053 (182)	0.08 (0.001)	4.7 (0.01)	119 (14)	37 (24)	2
D1	0.15 (0.027)	13 (1.3)	5035 (1200)	0.07 (0.001)	5.0 (0.91)	130 (15)	21 (4)	2
D10	0.09 (0.024)	16 (3.9)	4458 (971)	0.07 (0.009)	4.5 (0.30)	124 (14)	27 (4)	2
D6	0.10 (0.007)	12 (2.1)	5179 (515)	0.03 (0.005)	3.2 (0.21)	84 (4)	49 (14)	2
D9	0.19	17	5828	0.04	4.1	100	48	1
D4	0.13 (0.003)	14 (3.5)	3883 (430)	0.04 (0.006)	2.4 (0.06)	100 (13)	64 (29)	2
D7								

TABLE 5. Coefficients and summary statistics of linear regression models of metal concentrations (µg/g dry weight) on body weight (mg dry weight) for selected taxa and sites. Non significant relationships ($p > 0.05$) are not presented. Models computed with log-transformed data.

Metal	Taxon	Site	Intercept	(SE)	Slope	(SE)	n	R^2	p	RMS
Cu	*Orconectes* sp.	D8	2.10	(0.12)	-0.26	(0.07)	13	0.53	0.005	0.0019
Zn	Chironomidae	D8	1.87	(0.01)	-0.19	(0.07)	8	0.52	0.042	0.0006
Cd	*G. fasciatus*	D8	-0.30	(0.03)	-0.44	(0.07)	19	0.72	<0.0001	0.0036
Cu	*G. fasciatus*	D8	1.80	(0.01)	-0.07	(0.02)	19	0.32	0.011	0.0005
Fe	*G. fasciatus*	D8	2.99	(0.04)	-0.33	(0.10)	19	0.37	0.006	0.0089
Hg	*G. fasciatus*	D8	-1.17	(0.02)	0.32	(0.06)	19	0.61	<0.0001	0.0032
Ni	*G. fasciatus*	D8	0.26	(0.04)	-0.42	(0.09)	19	0.54	0.0004	0.0072
Pb	*G. fasciatus*	D8	0.04	(0.03)	-0.30	(0.08)	19	0.44	0.002	0.0055
Zn	*G. fasciatus*	D8	1.80	(0.01)	0.08	(0.01)	19	0.67	<0.0001	0.0002
Zn	*G. fasciatus*	D13	1.79	(0.01)	0.08	(0.02)	18	0.41	0.004	0.0003
Zn	*G. fasciatus*	D14	1.75	(0.01)	0.09	(0.04)	12	0.41	0.026	0.0002
Zn	*G. fasciatus*	D15	1.69	(0.02)	0.18	(0.08)	11	0.37	0.049	0.0012

TABLE 6. Significant differences (ANCOVA, $p < 0.05$) in metal concentrations in macroinvertebrates among reference, most impacted and least impacted zones, as defined relative to (a) Domtar Fine Papers/ICI Forest Products and (b) Courtaulds Fibers/Courtaulds Films. Metals followed by a star showed higher levels in organisms from the most impacted zone.

(a) Domtar Fine Papers/ICI Forest Products

Metal	Taxon	*p*
Hg*	Oligochaeta	0.0002[a]
Zn*	Chironomidae	0.028
Cr	*G. fasciatus*	0.047

(b) Courtaulds Fibers/Courtaulds Films

Metal	Taxon	*p*
Hg*	*B. tentaculata*	0.041
Zn*[b]	*B. tentaculata*	0.033
Zn*	*G. fasciatus*	0.030

[a] No reference sites could be included in this comparison.
[b] Concentrations in the most impacted zone may not differ significantly from average levels in the reference zone.

TABLE 7. Comparison of metal concentrations in littoral sediments with levels reported in the studies of Poulton (1994) and Richman (1994) for deeper sites in the Cornwall area. Values are the median of mean concentrations (µg/g dry weight) per site; ranges are given in parentheses. This study: *n* = 16, Poulton (1994): *n* = 7, Richman (1994): *n* = 14.

	This study	Poulton (1994)[1]	Richman (1994)[1]
Cd	0.23 (0.11-0.88)	0.19 (0.05-1.1)	1.53 (0.48-2.25)
Cr	10 (7-25)	24 (10-41)	34 (12-54)
Cu	8 (2-30)	22 (5-44)	42 (9-106)
Fe	9145 (4952-22238)	15000 (8450-20000)	15417 (6667-21000)
Hg	0.08 (0.01-1.12)	0.23 (0.01-0.52)	1.13 (0.06-3.29)
Ni	9 (5-21)	17 (6-27)	25 (11-36)
Pb	7 (1-26)	15 (4-38)	36 (9-425)
Zn	44 (16-174)	81 (24-465)	380 (48-1233)

[1] Only the sites encompassed within my study area (downstream of the dam) were included in this comparison.

Table 8. Coefficients and summary statistics of ANCOVA models for the comparison of metal concentrations in littoral sediments with levels reported in the studies of Poulton (1994) and Richman (1994) for deeper sites in the Cornwall area. Models computed with log-transformed data.

	Effect of the proportion of silt and clay			Effect of main factor (study)	Effect[1] of Poulton		Effect[1] of Richman	
Metal	Coefficient	(SE)	*p*	*p*	Coefficient	*p*	Coefficient	*p*
Cd	1.01	(0.12)	<0.0001	<0.0001	-0.19	0.027	0.32	0.0002
Cr	0.62	(0.08)	<0.0001	0.0008	0.20	0.0009	0.19	0.001
Cu	0.91	(0.14)	<0.0001	0.013	0.25	0.018	0.27	0.007
Fe	0.37	(0.08)	<0.0001	0.20	0.09	0.11	0.00	0.99
Hg	0.85	(0.33)	0.014	0.003	0.01	0.97	0.74	0.002
Ni	0.57	(0.08)	<0.0001	0.022	0.11	0.06	0.15	0.008
Pb	0.91	(0.18)	<0.0001	0.027	0.18	0.17	0.35	0.008
Zn	0.96	(0.19)	<0.0001	0.044	0.16	0.26	0.34	0.013

[1] Test of the difference between levels reported by Poulton (1994) or Richman (1994) and concentrations in littoral sediments (this study) after correction for the proportion of silt and clay.

Table 9. Comparison of metal concentrations in littoral sediments and in chironomids and oligochaetes (values pooled) with levels reported for sediments and benthic macroinvertebrates of contaminated areas in the Great Lakes (Jaagumagi and Persaud, 1992). Values are the medians of mean concentrations (µg/g dry weight) per site; ranges are given in parentheses. Also given are the percentiles of the values reported for the Great Lakes areas corresponding to the median values in the present

	Littoral zone			Great Lakes[1]			
	Median	Range	*n*	Median	Range	*n*	Percentile
(a) Sediments							
Cd	0.23	(0.11-0.88)	16	0.83	(0.02-18.00)	138	11
Cr	10	(7-25)	16	34	(5-170)	66	5
Cu	8	(2-30)	16	38	(1-430)	138	8
Fe	9145	(4952-22238)	16	18000	(3900-110000)	138	14
Hg	0.08	(0.01-1.12)	16	0.12	(0.01-8.50)	132	36
Ni	9	(5-21)	16	15	(3-42)	61	21
Pb	7	(1-26)	16	50	(2-570)	138	9
Zn	44	(16-174)	16	105	(10-2800)	126	22
(b) Macroinvertebrates[2,3]							
Cd	0.15	(0.03-0.58)	15	0.63	(0.03-8.36)	138	3
Cr	8.5	(2.8-14.8)	10	8.2	(1-70)	66	53
Cu	16	(11-18)	15	23	(0-145)	138	28
Fe	5035	(2643-8717)	15	2770	(377-16069)	138	89
Hg	0.09	(0.04-0.68)	15	0.20	(0.01-2.48)	132	15
Ni	4.8	(1.4-7.2)	10	4.2	(0.1-18.9)	61	54
Pb	4.5	(2.4-7.0)	15	6.8	(0.6-112.5)	138	18
Zn	86	(67-130)	15	138	(12-635)	126	16

[1] Only the sites for which there were data for both sediments and macroinvertebrates were used in the comparisons.

[2] For Cr and Ni, only the concentration values for chironomids (this study) were used in the comparisons.

[3] Macroinvertebrate samples from the Great Lakes areas were mostly composed of oligochaetes and chironomids. Concentrations were converted from on a wet weight basis to on a dry weight basis using the formula:
value in µg/g dry weight = value in µg/g wet weight * 100/15.9 (Persaud et al.,

TABLE 10. Comparison of metal concentrations in *G. fasciatus* and *B. tentaculata* with values reported for the same species from two fluvial lakes of the St. Lawrence River in Amyot et al. (1994; *G. fasciatus*) and Flessas (1994; *B. tentaculata*). Values are the medians of mean metal concentrations (µg/g dry weight) per site; ranges are given in parentheses.

	This study	Lake St. Louis	Lake St. Pierre
(a) *G. fasciatus*			
Cd	0.15 (0.06-0.46)	0.44 (0.15-1.76)	0.40 (0.22-0.54)
Cr	2.3 (1.7-3.8)	2.2 (1.7-3.4)	- -
Cu	60 (46-96)	74 (62-91)	76 (64-102)
Fe	790 (409-1738)	520 (331-1297)	762 (382-1115)
Ni	1.3 (0.8-2.2)	1.3 (0.8-1.9)	2.1 (1.3-3.2)
Pb	0.9 (0.6-1.8)	0.8 (0.1-1.2)	2.7 (0.6-25.5)
Zn	66 (51-85)	75 (66-140)	80 (66-104)
	n = 15	*n* = 12	*n* = 10
(b) *B. tentaculata*			
Cd	0.45 (0.21-0.95)	1.09 (0.35-1.73)	
Cu	292 (154-656)	237 (140-486)	
Fe	3282 (2540-4516)	1335 (630-2269)	
Pb	2.7 (1.4-6.0)	1.6 (0.3-9.7)	
Zn	271 (140-538)	319 (184-506)	
	n = 12	*n* = 11	

TABLE 11. Comparison of metal concentrations in chironomids and oligochaetes (values pooled) with levels reported for benthic macroinvertebrates of deeper sites in the Cornwall area (Jaagumagi and Persaud, 1992). Values are the medians of mean concentrations (µg/g dry weight) per site; ranges are given in parentheses. This study: *n* = 15, except for Cr and Ni, *n* = 10; Jaagumagi and Persaud (1992): *n* = 8.

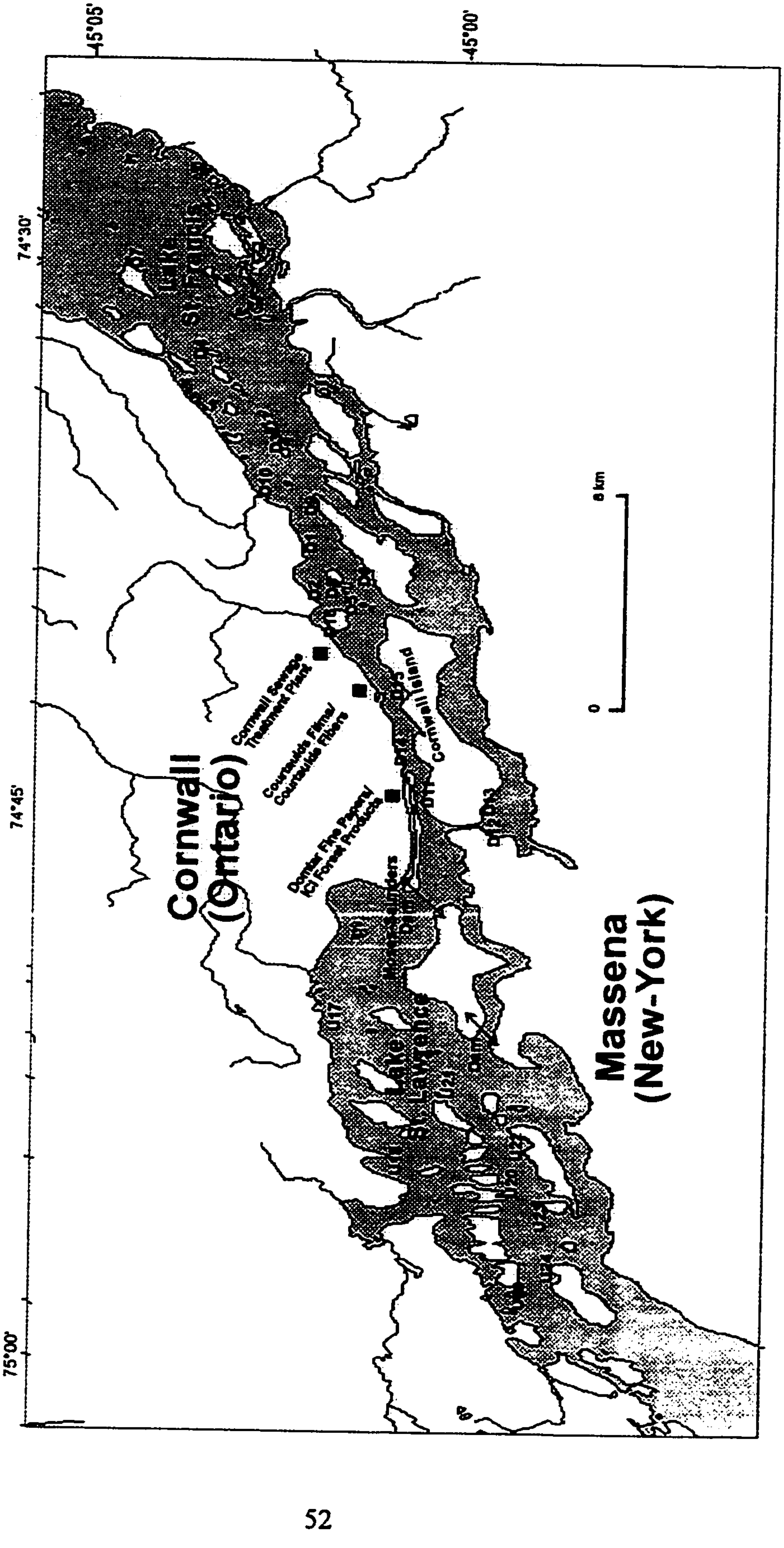
	This study[1]	Jaagumagi and Persaud (1992)[2,3]
Cd	0.15 (0.03-0.58)	0.35 (0.13-1.01)
Cr	8.5 (2.8-14.8)	5.7 (0.6-15.1)
Cu	16 (11-18)	47 (10-75)
Fe	5035 (2643-8717)	1075 (635-2893)
Hg	0.09 (0.04-0.68)	0.18 (0.03-0.63)
Ni	4.8 (1.4-7.2)	3.1 (2.5-5.7)
Pb	4.5 (2.4-7.0)	8.2 (3.8-26.4)
Zn	86 (67-130)	94 (69-220)

[1] For Cr and Ni, only the concentration values for chironomids were used in the comparisons.

[2] Only the sites encompassed within my study area were included in this comparison.

[3] Macroinvertebrate samples were mostly composed of oligochaetes and chironomids. Concentrations were converted from on a wet weight basis to on a dry weight basis using the formula of Table 9.

Fig. 1. Study area and location of sampling sites.



45°05'
45°00'
74°30'
74°45'
75°00'
Lake St. Francis
Cornwall (Ontario)
Cornwall Sewage Treatment Plant
Courtaulds Films/ Courtaulds Fibers
Domtar Fine Papers/ ICI Forest Products
Cornwall Island
Lake St. Lawrence
Dam
Massena (New-York)
0
8 km

Fig. 2. Correlation between the proportion of fine inorganic particles (silt and clay) and the organic matter content of littoral sediments. Values are mean % (dry weight) per site; *n* = 16.

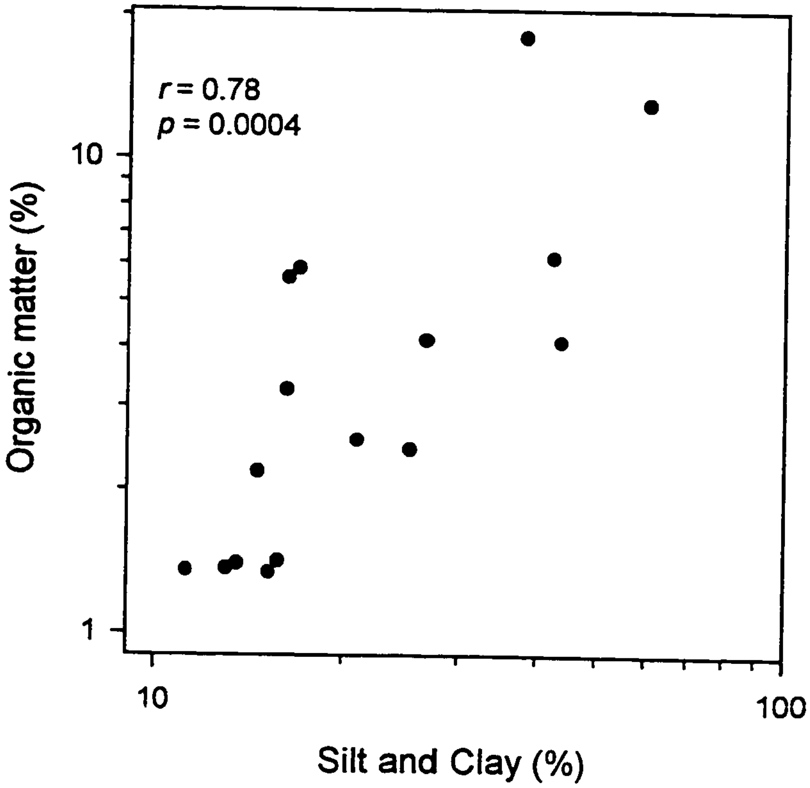

r = 0.78
p = 0.0004
Organic matter (%)
10
1
10
100
Silt and Clay (%)

Fig. 3. Mean metal concentrations (µg/g dry weight ± 1 SE) per site in littoral sediments. The dotted line indicates the lowest effect level (LEL) of the Ontario Provincial Sediment Quality Guidelines (Persaud et al., 1993). The severe effect level (SEL) is also given for comparison. The mean concentration of Hg at site D2 attains the LEL when the rounding procedure of the guidelines is applied (Persauld et al., 1993). All study sites: *n* = 3, except D1 and D2: *n* = 1, D16: *n* = 2, and (for Pb only) D3 and D12: *n* = 2. Study sites are ordered from upstream to downstream within the reference, most impacted and least impacted zones (see Table 1).

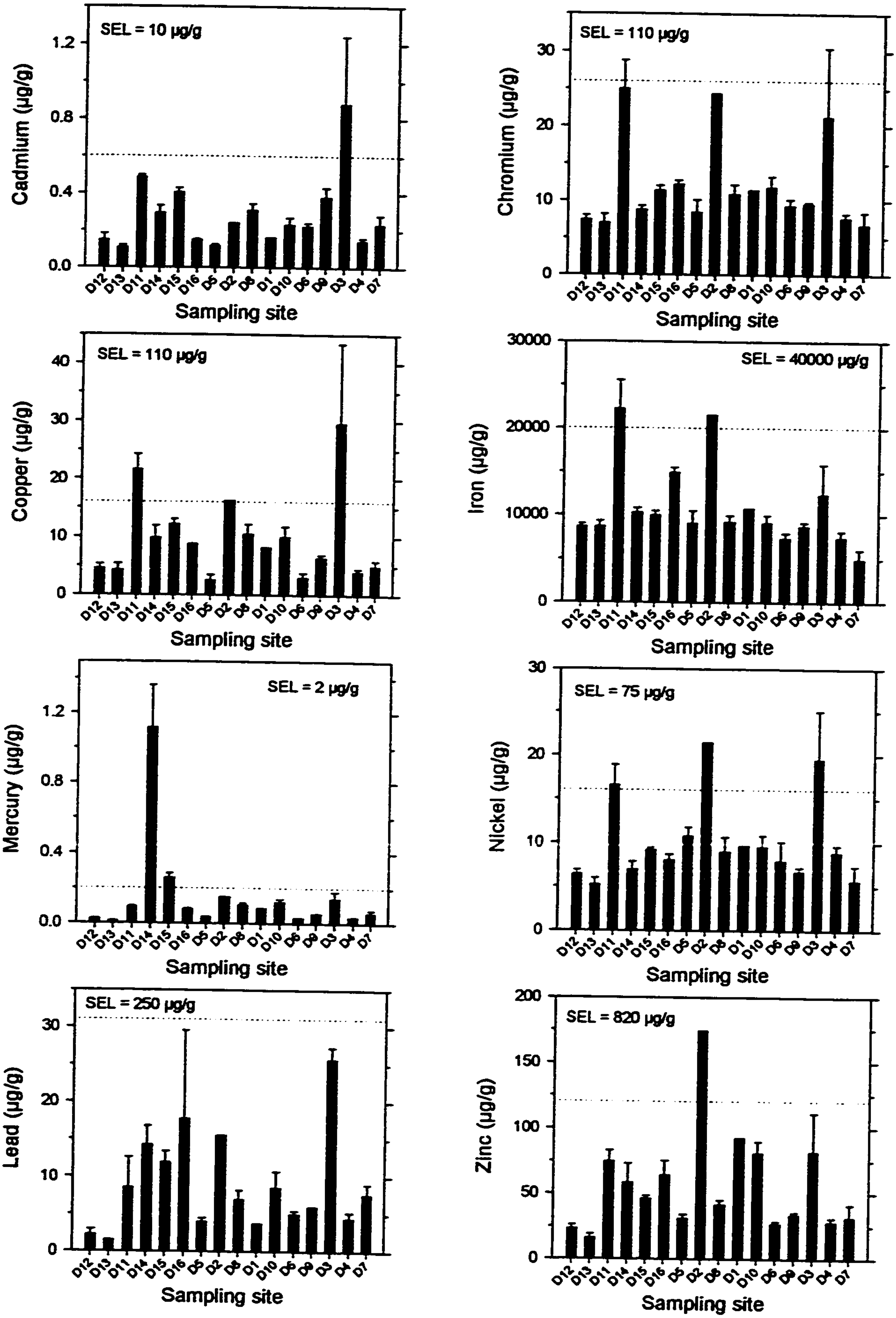
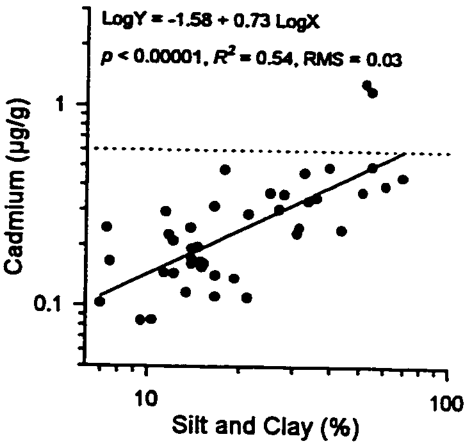
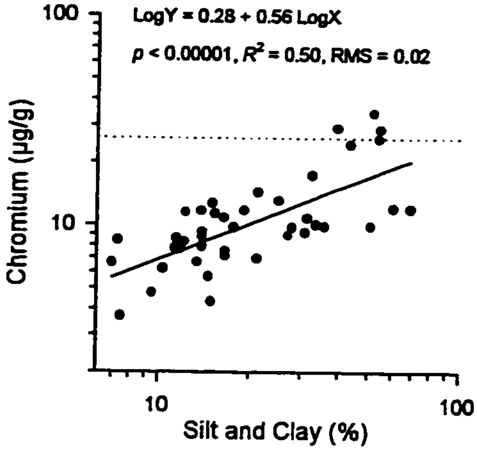
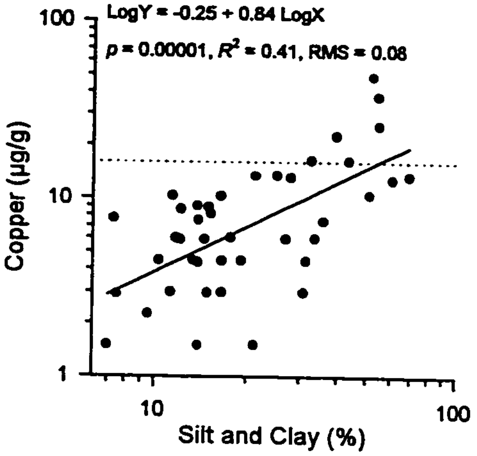
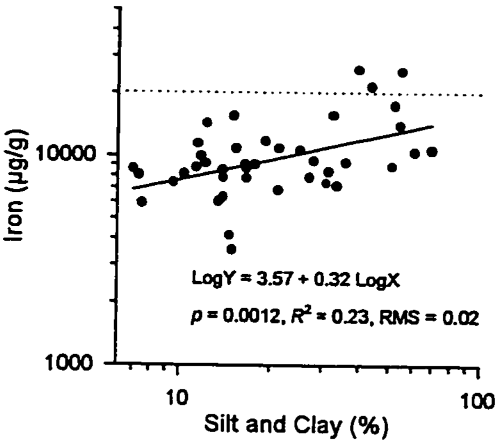

SEL = 10 µg/g
Cadmium (µg/g)
0.0
0.4
0.8
1.2
SEL = 110 µg/g
Chromium (µg/g)
0
10
20
30
SEL = 110 µg/g
Copper (µg/g)
0
10
20
30
40
SEL = 40000 µg/g
Iron (µg/g)
0
10000
20000
30000
SEL = 2 µg/g
Mercury (µg/g)
0.0
0.4
0.8
1.2
SEL = 75 µg/g
Nickel (µg/g)
0
10
20
30
SEL = 250 µg/g
Lead (µg/g)
0
10
20
30
SEL = 820 µg/g
Zinc (µg/g)
0
50
100
150
200
D12 D13 D11 D14 D15 D16 D5 D2 D8 D1 D10 D6 D9 D3 D4 D7
Sampling site

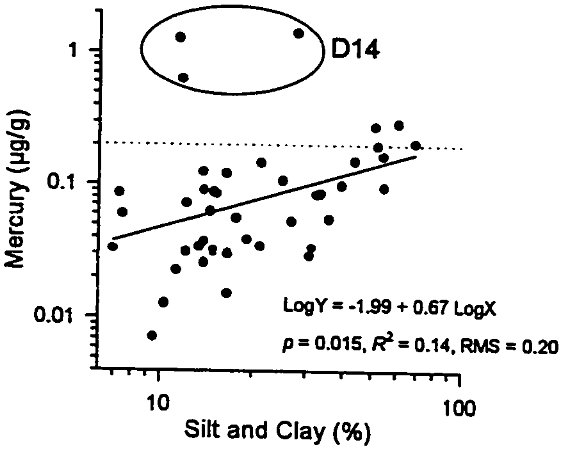
Fig. 4. Relationships between metal concentrations (µg/g dry weight) and the proportion of silt and clay (% dry weight) in littoral sediment samples (*n* = 42, except Pb: *n* = 40). The solid line represents the regression line and the dotted line indicates the lowest effect level of the sediment quality guidelines.

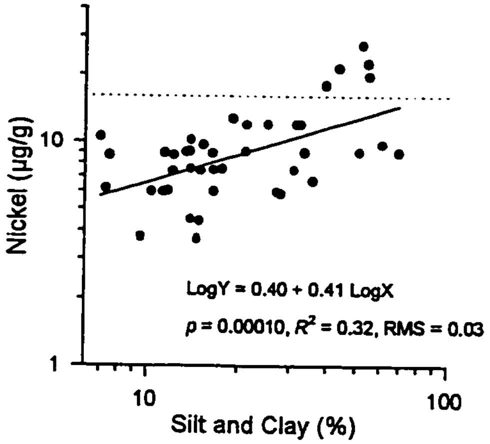

LogY = -1.58 + 0.73 LogX
$p < 0.00001$, $R^2 = 0.54$, RMS = 0.03
Cadmium (µg/g)
1
0.1
10
100
Silt and Clay (%)

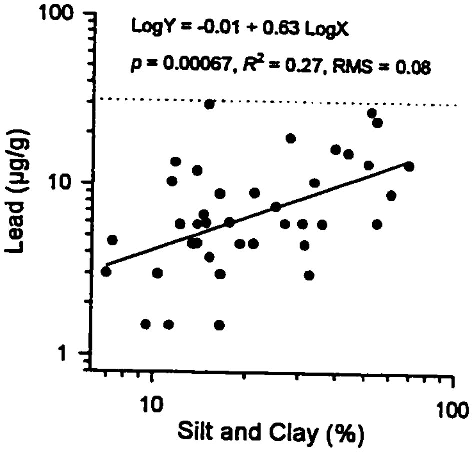

LogY = 0.28 + 0.56 LogX
$p < 0.00001$, $R^2 = 0.50$, RMS = 0.02
Chromium (µg/g)
100
10
10
100
Silt and Clay (%)


LogY = -0.25 + 0.84 LogX
$p = 0.00001$, $R^2 = 0.41$, RMS = 0.08
Copper (µg/g)
100
10
1
10
100
Silt and Clay (%)


Iron (µg/g)
10000
1000
LogY = 3.57 + 0.32 LogX
$p = 0.0012$, $R^2 = 0.23$, RMS = 0.02
10
100
Silt and Clay (%)


D14
Mercury (µg/g)
1
0.1
0.01
LogY = -1.99 + 0.67 LogX
$p = 0.015$, $R^2 = 0.14$, RMS = 0.20
10
100
Silt and Clay (%)


Nickel (µg/g)
10
1
LogY = 0.40 + 0.41 LogX
$p = 0.00010$, $R^2 = 0.32$, RMS = 0.03
10
100
Silt and Clay (%)


LogY = -0.01 + 0.63 LogX
$p = 0.00067$, $R^2 = 0.27$, RMS = 0.08
Lead (µg/g)
100
10
1
10
100
Silt and Clay (%)

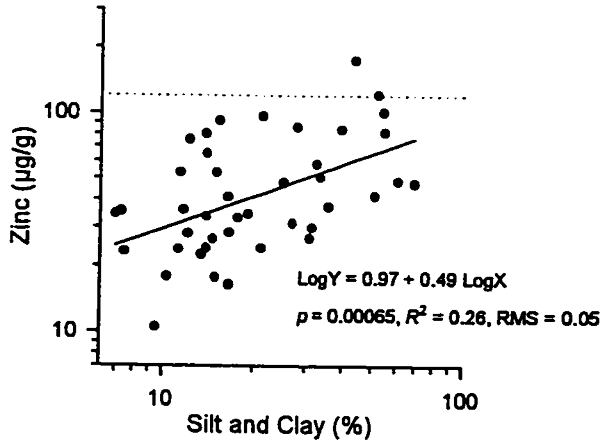

Zinc (µg/g)
100
10
LogY = 0.97 + 0.49 LogX
$p = 0.00065$, $R^2 = 0.26$, RMS = 0.05
10
100
Silt and Clay (%)

Fig. 5. Relationships between metal concentrations (µg/g dry weight) and the quantity of organic matter (% dry weight) in littoral sediment samples (*n* = 43, except Pb: *n* = 41). The solid line represents the regression line and the dotted line indicates the lowest effect level of the sediment quality guidelines.

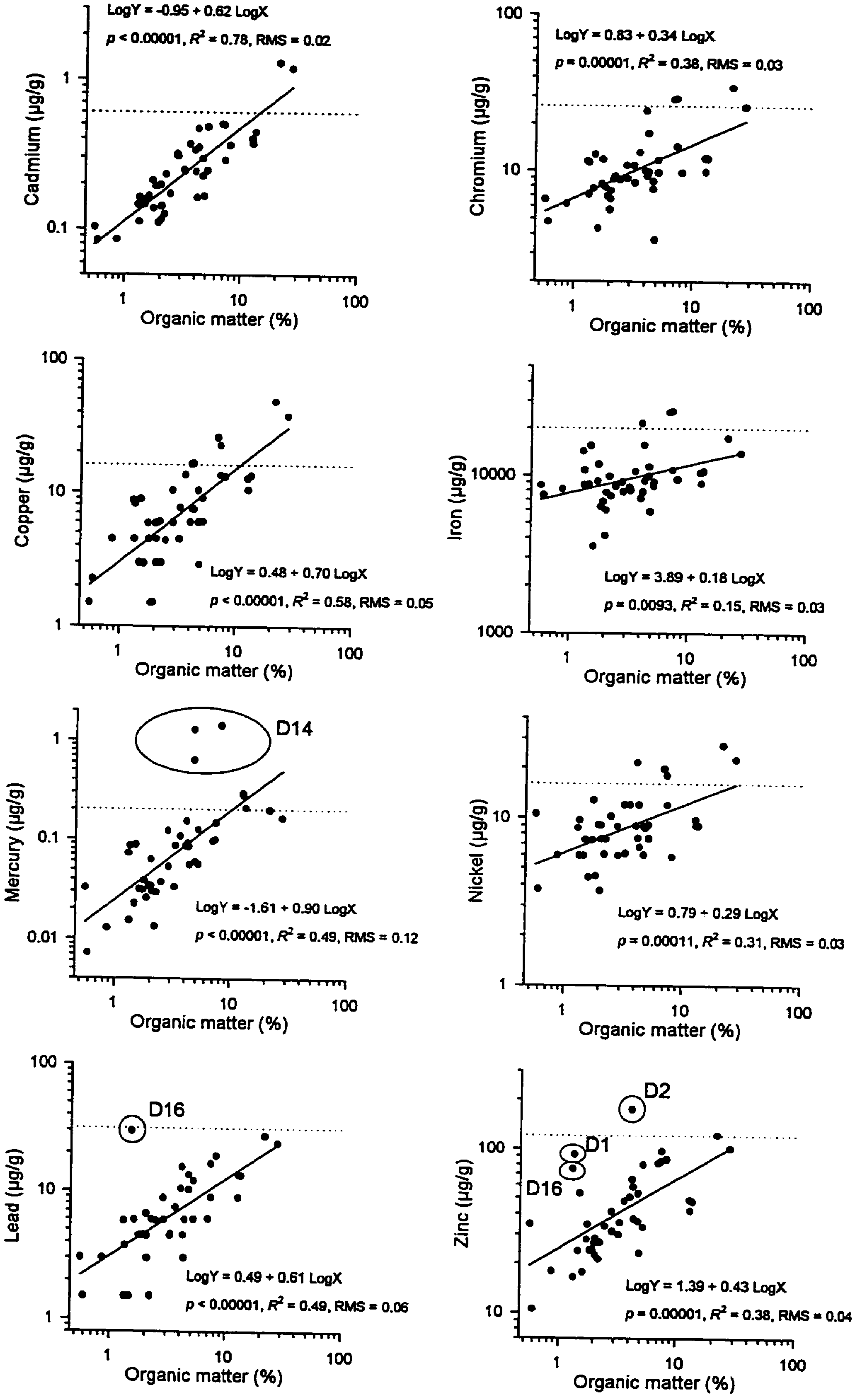

LogY = -0.95 + 0.62 LogX
p < 0.00001, R² = 0.78, RMS = 0.02
Cadmium (µg/g)
1
0.1
1
10
100
Organic matter (%)
LogY = 0.83 + 0.34 LogX
p = 0.00001, R² = 0.38, RMS = 0.03
Chromium (µg/g)
100
10
1
10
100
Organic matter (%)
Copper (µg/g)
100
10
1
LogY = 0.48 + 0.70 LogX
p < 0.00001, R² = 0.58, RMS = 0.05
1
10
100
Organic matter (%)
Iron (µg/g)
10000
1000
LogY = 3.89 + 0.18 LogX
p = 0.0093, R² = 0.15, RMS = 0.03
1
10
100
Organic matter (%)
D14
Mercury (µg/g)
1
0.1
0.01
LogY = -1.61 + 0.90 LogX
p < 0.00001, R² = 0.49, RMS = 0.12
1
10
100
Organic matter (%)
Nickel (µg/g)
10
1
LogY = 0.79 + 0.29 LogX
p = 0.00011, R² = 0.31, RMS = 0.03
1
10
100
Organic matter (%)
D16
Lead (µg/g)
100
10
1
LogY = 0.49 + 0.61 LogX
p < 0.00001, R² = 0.49, RMS = 0.06
1
10
100
Organic matter (%)
D2
D1
D16
Zinc (µg/g)
100
10
LogY = 1.39 + 0.43 LogX
p = 0.00001, R² = 0.38, RMS = 0.04
1
10
100
Organic matter (%)

Fig. 6. Comparison of whole-body metal concentrations in macroinvertebrates among taxa and with levels in sediments. Box plots represent the distribution of mean concentration values (µg/g dry weight) per site. The dotted line represents the median of mean sediment metal concentrations (µg/g dry weight) per site (calculated without site D3). *Gammarus fasciatus*: *n* = 15 sites, *Orconectes* sp.: *n* = 10, *Bythinia tentaculata*: *n* = 12, Chironomidae: *n* = 10, and Oligochaeta: *n* = 10.

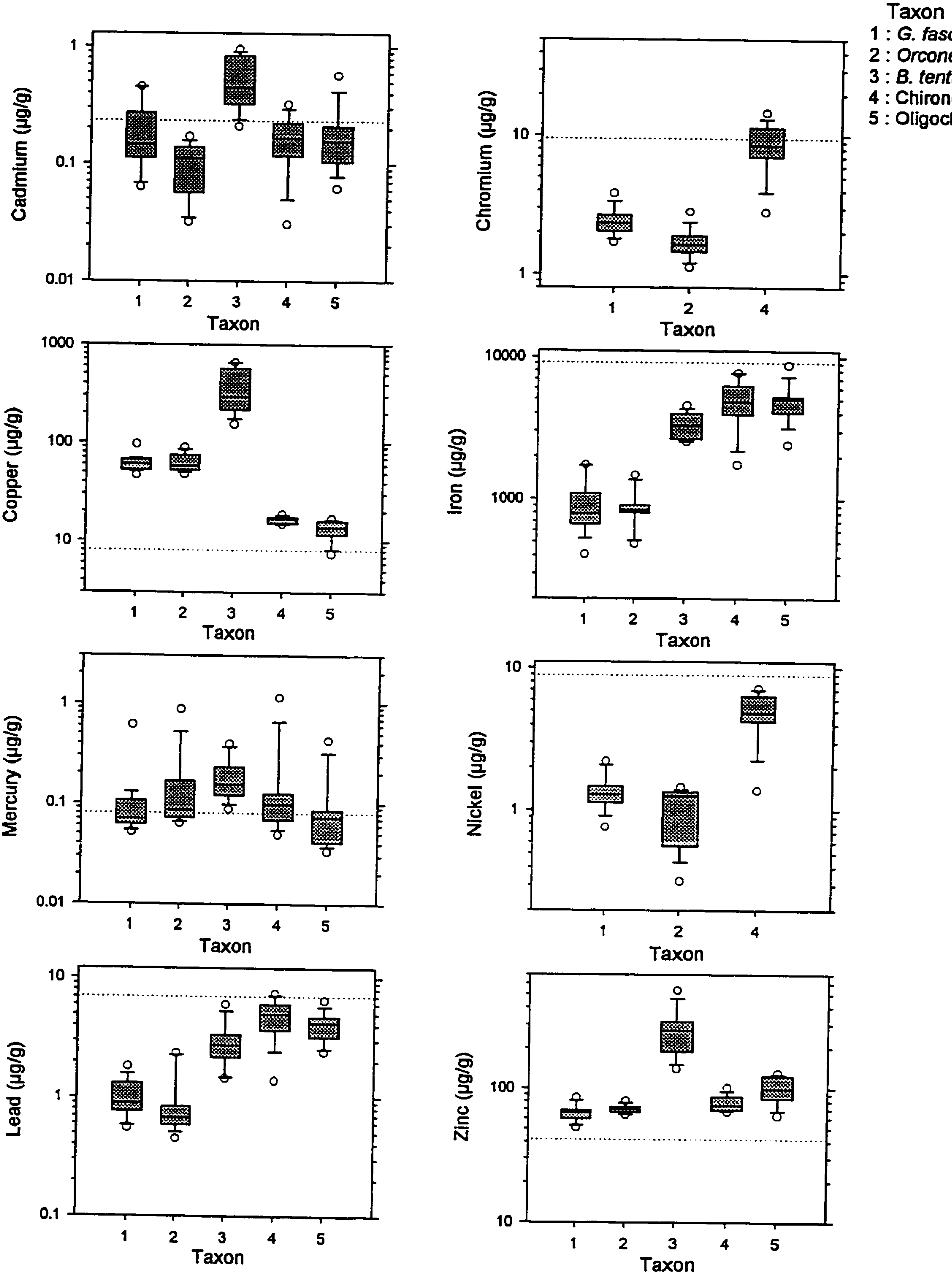

Taxon
1 : *G. fasc*
2 : *Orcone*
3 : *B. tent*
4 : Chiron
5 : Oligocl
Cadmium (µg/g)
1
0.1
0.01
Chromium (µg/g)
10
1
Copper (µg/g)
1000
100
10
Iron (µg/g)
10000
1000
Mercury (µg/g)
1
0.1
0.01
Nickel (µg/g)
10
1
Lead (µg/g)
10
1
0.1
Zinc (µg/g)
100
10
1
2
3
4
5
Taxon

Fig. 7. Comparison of mean sediment Hg and Zn concentrations (µg/g dry weight) per site among reference, most impacted and least impacted zones, as defined relative to (a) Domtar Fine Papers/ICI Forest Products (Hg) and (b) Courtaulds Fibers/Courtaulds Films (Zn). Concentrations are plotted against the mean amount of silt and clay and of organic matter (% dry weight). Lines through data points represent regression lines and the horizontal dotted line indicates the lowest effect level of the sediment quality guidelines.

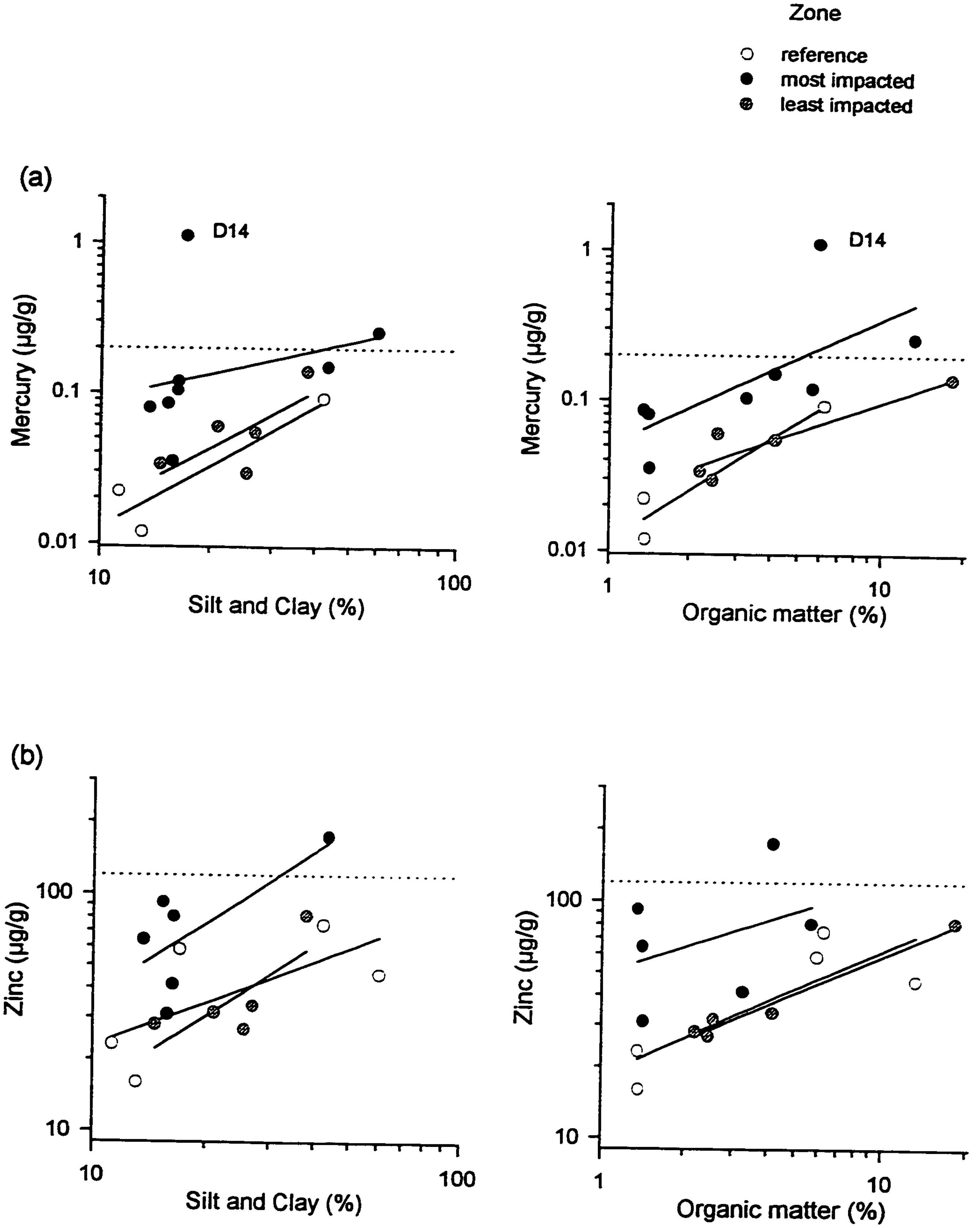
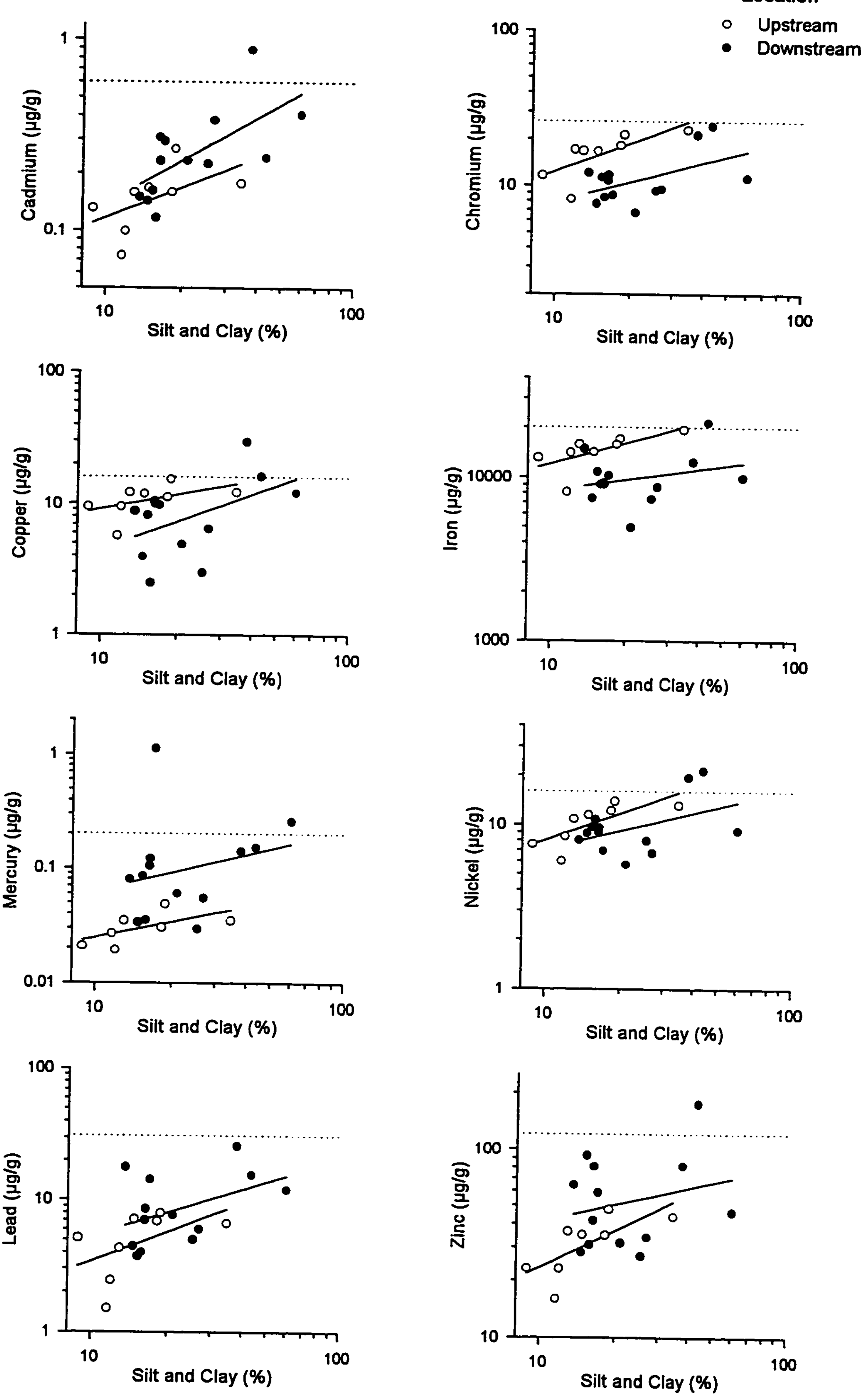
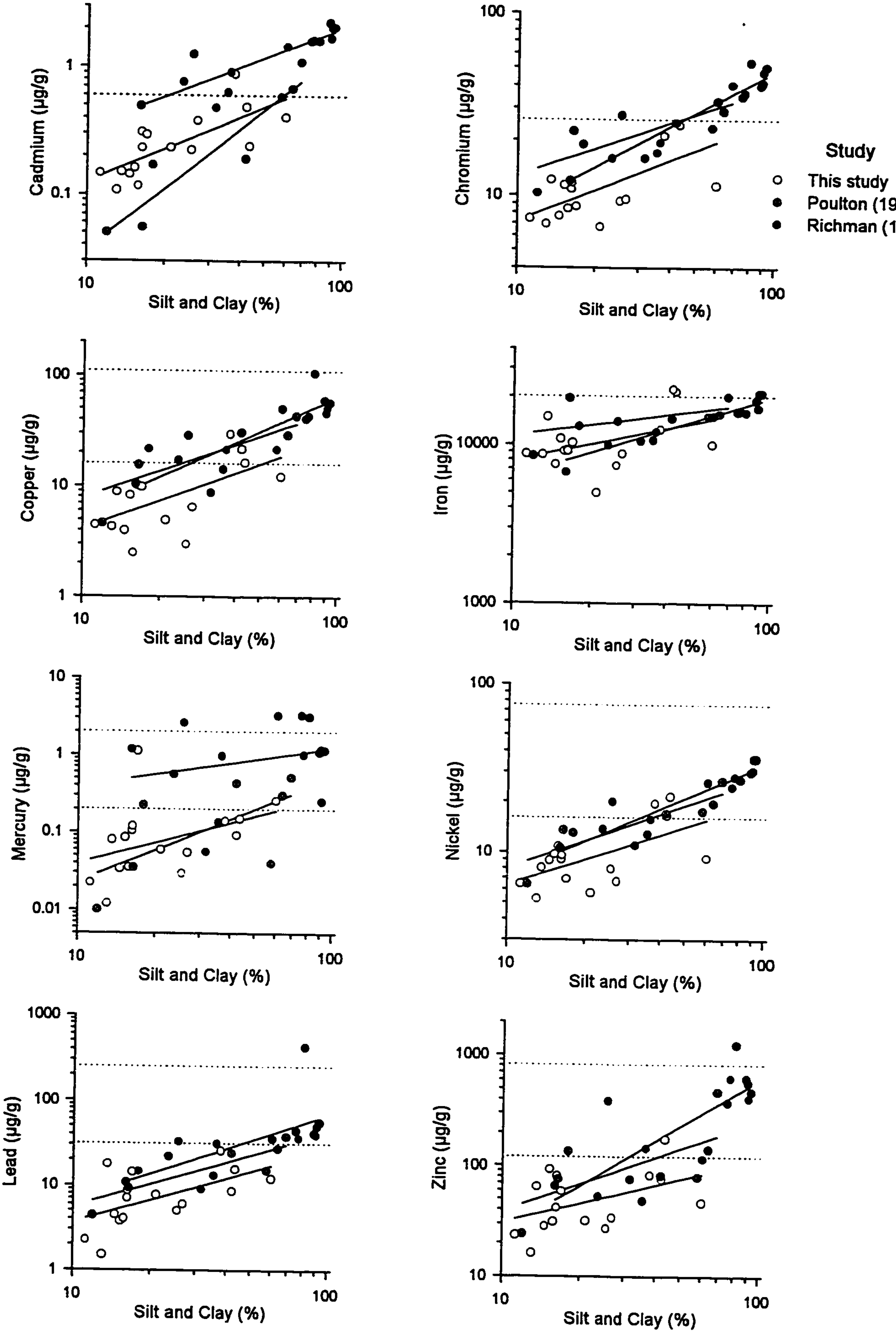

Zone
○ reference
● most impacted
⊛ least impacted
(a)
D14
Mercury (µg/g)
1
0.1
0.01
10
100
Silt and Clay (%)
D14
Mercury (µg/g)
1
0.1
0.01
1
10
Organic matter (%)
(b)
Zinc (µg/g)
100
10
10
100
Silt and Clay (%)
Zinc (µg/g)
100
10
1
10
Organic matter (%)

Fig. 8. Comparison of mean sediment metal concentrations (µg/g dry weight) per site between upstream (*n* = 8) and downstream (*n* = 13) littoral sites. Concentrations are plotted against the mean proportion of silt and clay (% dry weight). Solid lines represent regression lines and the dotted line indicates the lowest effect level of the sediment quality guidelines.



Upstream
Downstream
Cadmium (µg/g)
1
0.1
10
100
Silt and Clay (%)
Chromium (µg/g)
100
10
10
100
Silt and Clay (%)
Copper (µg/g)
100
10
1
10
100
Silt and Clay (%)
Iron (µg/g)
10000
1000
10
100
Silt and Clay (%)
Mercury (µg/g)
1
0.1
0.01
10
100
Silt and Clay (%)
Nickel (µg/g)
10
1
10
100
Silt and Clay (%)
Lead (µg/g)
100
10
1
10
100
Silt and Clay (%)
Zinc (µg/g)
100
10
10
100
Silt and Clay (%)

Fig. 9. Comparison of metal concentrations (mean µg/g dry weight per site) in littoral sediments with levels reported in the studies of Poulton (1994) and Richman (1994) for deeper sites in the Cornwall area. Concentrations are plotted against the mean amount of silt and clay (% dry weight). Lines through data points represent regression lines and the horizontal dotted lines indicate the lowest effect (LEL) and the severe effect (SEL) levels of the sediment quality guidelines. This study: $n = 16$, Poulton (1994): $n = 7$, Richman (1994): $n = 14$. Only the sites encompassed within my study area (downstream of the dam) were included in this comparison.


Cadmium (µg/g)
Silt and Clay (%)
Chromium (µg/g)
Silt and Clay (%)
Study
○ This study
● Poulton (19
● Richman (1
Copper (µg/g)
Silt and Clay (%)
Iron (µg/g)
Silt and Clay (%)
Mercury (µg/g)
Silt and Clay (%)
Nickel (µg/g)
Silt and Clay (%)
Lead (µg/g)
Silt and Clay (%)
Zinc (µg/g)
Silt and Clay (%)
0.1
1
10
100
1000
10000
0.01

APPENDICES

Appendix 1. Particle size distribution (% dry weight), organic matter content (% dry weight), and metal concentrations (μg/g dry weight) in littoral sediments (downstream of the Moses-Saunders Dam).

Site	Gravel	Sand	Silt and Clay	Organic matter	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
D1	5.7	79.0	15.3	1.3	0.16	11	8	10799	0.085	10	4	92
D2	7.7	48.8	43.5	4.1	0.24	24	16	21511	0.152	21	16	174
D3-1	1.9	90.6	7.5	4.9	0.17	4	3	5892	0.059	9		23
D3-2	2.2	43.6	54.2	27.7	1.19	26	38	13966	0.164	23	24	101
D3-3	0.0	48.0	52.0	21.5	1.29	34	49	17400	0.198	27	27	121
D4-1	15.1	71.5	13.4	2.0	0.12	7	4	6017	0.034	9	4	22
D4-2	0.9	82.5	16.6	2.1	0.14	8	3	7764	0.030	7	3	28
D4-3	5.1	81.0	13.9	2.5	0.17	9	4	8512	0.037	10	6	34
D5-1	4.1	88.9	7.0	0.6	0.10	7	1	8582	0.033	10	3	34
D5-2	0.9	79.9	19.2	1.8	0.14	12	4	11742	0.038	13	4	34
D5-3	1.5	77.3	21.2	1.9	0.11	7	2	6829	0.034	9	5	24
D6-1	0.4	68.2	31.4	3.2	0.25	11	4	8364	0.033	12	4	30
D6-2	0.7	85.4	13.9	1.9	0.19	8	1	6324	0.026	4	4	24
D6-3	0.0	69.1	30.9	2.2	0.23	9	3	7405	0.029	7	6	27
D7-1	1.0	65.4	33.6	4.0	0.33	10	6	7160	0.086	9	10	51
D7-2	1.0	84.4	14.6	2.0	0.20	6	6	4157	0.062	4	7	26
D7-3	0.8	84.3	14.9	1.6	0.17	4	3	3538	0.032	4	6	18
D8-1	3.1	71.8	25.1	3.6	0.37	13	13	10610	0.107	12	7	48
D8-2	0.7	82.9	16.4	2.8	0.31	11	10	9019	0.121	9	9	41
D8-3	2.4	90.3	7.3	3.3	0.24	8	8	8039	0.086	6	5	35

Appendix 1. Continued.

Site	Gravel	Sand	Silt and Clay	Organic matter	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
D9-1	0.8	72.2	27.0	2.9	0.31	9	6	7864	0.053	6	6	31
D9-2	1.3	62.9	35.8	4.4	0.35	10	7	9236	0.055	7	6	37
D9-3	3.1	79.2	17.7	5.2	0.48	10	6	9046	0.056	8	6	33
D10-1	0.4	85.8	13.8	5.2	0.24	12	9	8526	0.125	9	12	80
D10-2	0.2	85.9	13.9	4.2	0.16	9	8	7849	0.090	8	5	65
D10-3	1.0	77.7	21.3	7.4	0.29	14	13	10827	0.147	12	9	97
D11-1	0.5	60.1	39.4	7.4	0.49	29	22	25744	0.099	18	16	84
D11-2	0.0	45.3	54.7	7.0	0.50	29	26	25365	0.096	20	6	82
D11-3	0.4	67.0	32.6	4.3	0.46	17	16	15606	0.084	12	3	58
D12-1	0.2	87.7	12.1	1.7	0.21	8	6	9152	0.031	7		28
D12-2	18.1	70.6	11.3	1.5	0.15	8	3	8734	0.023	6	1	24
D12-3	0.2	89.5	10.3	0.9	0.08	6	4	8116	0.013	6	3	18
D13-1	0.0	90.5	9.5	0.6	0.08	5	2	7418	0.007	4	1	10
D13-2	0.0	83.4	16.6	1.3	0.11	7	4	8645	0.015	6	1	16
D13-3				2.2	0.13	9	6	9820	0.013	6	2	21
D14-1	1.6	70.5	27.9	8.2	0.36	10	13	9455	1.429	6	19	86
D14-2	7.3	81.0	11.7	4.7	0.23	8	6	9932	0.631	6	13	36
D14-3	8.6	80.0	11.4	4.7	0.29	9	10	11373	1.290	9	10	53
D15-1	2.2	46.8	51.0	12.9	0.37	10	10	8888	0.279	9	13	42
D15-2	0.6	38.7	60.7	12.8	0.40	12	13	10412	0.293	10	9	49
D15-3	2.7	28.0	69.3	13.6	0.44	12	13	10622	0.208	9	13	48
D16-1	7.1	80.7	12.2	1.3	0.14	12	9	14264	0.072	9	6	75
D16-2	1.8	83.2	15.0	1.5	0.16	13	9	15451	0.087	7	30	53

Appendix 2. Whole-body metal concentrations (µg/g dry weight) and body weight (mg dry weight) of littoral macroinvertebrates. *n* is the number of individuals composing a sample. No data are available at sites with no values.

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D1	0.07	2.1	96	409	0.13	1.3	0.6	81	1.7	29
D2	0.11	4.3	60	2071	0.07	2.1	1.8	87	2.2	25
D2	0.09	3.1	48	1489	0.06	1.8	1.7	75	1.1	52
D2	0.11	4.1	80	1652	0.08	2.7	1.2	93	3.8	7
D4	0.29	2.0	70	448	0.08	1.1	1.1	74	6.0	10
D4	0.23	1.9	70	512	0.08	1.4	0.5	73	4.0	12
D4	0.31	1.6	56	635	0.06	1.3	0.6	66	2.2	29
D5	0.70	2.5	77	1107	0.05	1.6	0.9	61	1.2	50
D5	0.38	1.9	67	719	0.06	1.3	0.7	61	3.2	19
D5	0.52	2.7	74	1110	0.07	1.7	0.9	64	2.6	19
D5	0.57	2.5	67	1056	0.06	1.4	0.9	62	2.0	31
D5	0.30	2.4	67	835	0.08	1.4	0.7	72	4.6	13
D5	0.29	2.1	67	785	0.07	1.3	0.8	66	4.5	12
D5	0.41	2.4	62	878	0.06	0.9	0.7	66	3.2	19
D6	0.14	1.9	49	933	0.06	1.1	0.8	56	0.9	68
D6	0.37	2.0	49	1005	0.06	1.2	0.9	58	1.0	58
D6	0.13	2.1	55	975	0.06	0.9	0.7	59	1.7	35
D7	0.58	2.5	58	881	0.06	1.3	1.1	61	1.2	51
D7	0.47	2.9	58	875	0.07	1.6	1.0	64	2.0	24
D7	0.46	2.6	65	720	0.08	1.2	0.8	73	3.1	16
D7	0.31	2.3	58	686	0.07	1.1	0.7	69	4.2	15

Appendix 2. Continued.

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D8-1	0.41	2.4	56	763	0.08	1.2	0.9	66	2.0	28
D8-1	0.33	2.5	61	755	0.10	1.4	0.8	68	2.6	14
D8-1	0.24	2.4	54	664	0.13	0.9	0.7	76	4.0	13
D8-1	0.34	2.3	56	819	0.10	1.2	0.9	67	2.5	23
D8-1	0.43	2.5	61	991	0.08	1.6	1.1	65	1.3	40
D8-1	0.49	2.6	62	999	0.07	1.7	1.2	62	0.9	69
D8-1	0.46	2.4	64	907	0.07	1.7	1.0	64	1.3	31
D8-2	0.31	2.4	57	760	0.09	1.0	0.8	65	2.1	30
D8-2	0.34	2.6	59	1025	0.09	1.1	1.1	67	2.6	24
D8-2	0.41	2.6	66	951	0.08	1.6	1.0	67	1.3	41
D8-2	0.21	2.4	61	727	0.13	1.2	0.8	75	4.3	12
D8-2	0.42	2.8	62	1103	0.08	1.7	1.1	64	1.2	46
D8-2	0.31	2.2	64	774	0.09	0.9	0.9	69	3.2	19
D8-3	0.27	2.0	55	564	0.12	0.8	0.7	69	4.4	14
D8-3	0.49	2.2	61	736	0.07	1.7	1.0	65	1.9	26
D8-3	0.39	1.4	58	338	0.08	0.9	0.5	67	3.1	20
D8-3	0.35	2.1	60	609	0.08	2.1	0.8	68	2.7	21
D8-3	0.52	2.1	60	739	0.06	1.6	0.8	65	1.4	39
D8-3	0.47	2.2	65	792	0.07	1.9	1.0	66	1.2	36
D9	0.15	1.9	58	755	0.05	0.8	0.8	62	1.9	30
D9	0.17	1.7	65	625	0.06	0.7	0.7	66	2.5	24
D9	0.19	1.7	69	623	0.06	1.3	0.7	70	3.3	13
D9	0.17	1.8	62	753	0.05	0.8	0.8	58	1.3	46
D9	0.20	1.8	59	813	0.05	1.0	0.8	56	1.2	48
D9	0.13	1.9	56	758	0.05	0.8	0.8	60	1.9	31

Appendix 2. Continued.

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D10	0.06	1.9	59	746	0.05	1.1	0.7	69	3.1	17
D10	0.07	2.0	54	857	0.05	1.3	0.8	65	2.2	26
D10	0.03	3.0	67	1148	0.05	1.2	1.2	67	1.0	54
D10	0.08	2.6	62	1058	0.04	1.5	1.1	63	1.1	51
D10	0.08	2.3	60	917	0.06	1.3	0.9	69	2.1	26
D11	0.11	3.2	67	1752	0.07	1.7	1.1	61	1.7	34
D11	0.16	3.6	75	1854	0.06	1.7	1.3	59	1.0	51
D11	0.16	3.3	70	1870	0.05	1.7	1.2	58	1.0	53
D11	0.13	3.9	67	1558	0.09	1.2	0.9	69	2.7	22
D11	0.15	3.1	67	1685	0.07	1.4	1.0	63	2.1	26
D11	0.11	2.9	66	1632	0.08	1.3	1.0	62	1.6	35
D12	0.45	3.4	52	422	0.09	3.3	2.1	55	3.7	8
D12	0.18	2.7	50	536	0.06	1.5	1.4	50	1.7	33
D12	0.27	2.6	50	564	0.06	1.4	1.2	51	1.6	33
D12	0.29	2.5	46	406	0.06	1.7	1.6	48	2.5	23
D12-1	0.19	2.4	60	570	0.05	1.2	0.9	50	1.0	59
D12-2	0.24	2.6	58	689	0.05	0.9	1.1	53	0.9	65
D12-3	0.19	2.7	58	884	0.06	1.5	0.9	51	1.1	27
D13	0.11	2.8	74	1289	0.09	1.9	0.6	74	4.8	9
D13-1	0.13	2.8	71	1190	0.05	2.0	0.6	65	1.1	47
D13-1	0.12	2.0	60	673	0.07	2.1	0.3	71	3.4	18
D13-1	0.14	2.1	58	779	0.07	2.0	0.3	65	2.6	25
D13-1	0.15	2.5	64	842	0.07	1.5	0.3	71	2.6	19
D13-1	0.15	2.6	59	1037	0.06	1.8	0.4	62	1.9	29
D13-1	0.15	2.3	66	892	0.06	1.6	0.3	65	1.8	29

Appendix 2. Continued.

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D13-2	0.10	3.4	69	1641	0.07	2.2	0.8	62	1.2	49
D13-2	0.10	3.1	64	1357	0.08	2.1	0.7	67	2.7	23
D13-2	0.18	2.6	71	1140	0.08	2.1	0.6	64	1.9	28
D13-2	0.13	3.1	64	1361	0.07	1.7	0.7	66	1.9	27
D13-3	0.13	2.9	67	1380	0.12	1.5	0.7	65	1.9	28
D13-3	0.13	3.1	71	1411	0.09	1.8	0.7	64	1.9	30
D13-3	0.16	3.4	80	1517	0.08	1.9	0.8	67	1.2	49
D13-3	0.13	2.5	74	1109	0.10	1.7	0.6	65	1.9	30
D13-3	0.08	2.8	63	1271	0.09	2.1	0.7	65	3.5	14
D13-3	0.11	2.8	71	1273	0.08	1.9	0.7	65	2.7	20
D13-3	0.13	13.5	73	1441	0.10	5.6	0.7	70	2.6	21
D14-1	0.09	1.6	53	701	0.58	0.7	1.3	62	1.8	27
D14-1	0.06	1.5	52	592	0.60	0.2	1.0	60	2.5	24
D14-1	0.07	1.3	48	503	0.51	0.7	0.9	57	1.2	49
D14-1	0.06	1.7	54	639	0.64	0.8	1.1	61	1.8	29
D14-2	0.11	2.1	53	925	0.55	1.1	1.8	58	1.1	47
D14-2	0.03	2.0	57	731	0.56	1.1	1.4	59	1.2	34
D14-2	0.06	2.0	49	893	0.58	1.0	1.7	60	2.3	26
D14-2	0.08	1.9	51	769	0.65	0.8	1.6	60	1.9	32
D14-3	0.09	1.9	55	504	0.70	0.9	1.3	61	1.7	26
D14-3	0.06	1.5	53	470	0.65	0.6	1.0	60	2.3	26
D14-3	0.08	1.4	52	555	0.62	0.5	1.2	54	1.0	58
D14-3	0.08	1.4	52	533	0.68	0.6	1.1	55	1.7	32

Appendix 2. Continued.

(a) *Gammarus fasciatus*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D15-1	0.10	2.0	49	904	0.10	0.7	1.0	53	1.2	50
D15-1	0.09	2.1	49	908	0.10	0.9	1.0	52	1.2	50
D15-1	0.08	1.6	51	572	0.12	0.3	0.6	58	2.7	22
D15-1	0.09	2.4	46	876	0.12	0.3	0.9	55	1.8	32
D15-2	0.08	2.1	40	838	0.13	0.3	0.8	56	2.5	20
D15-2	0.08	2.0	46	810	0.12	0.5	1.2	56	1.9	29
D15-2	0.09	1.8	48	845	0.13	0.3	0.9	54	1.3	38
D15-2	0.08	2.1	47	1047	0.13	0.9	0.9	54	1.2	31
D15-3	0.06	1.7	47	767	0.14	0.3	0.8	59	2.4	21
D15-3	0.28	3.0	43	395	0.11	2.1	3.9	42	1.1	54
D15-3	0.46	3.0	44	342	0.12	4.0	5.4	47	1.8	26
D16	0.10	2.6	53	1151	0.14	1.8	2.6	77	2.8	12
D16	0.21	2.4	53	1189	0.10	1.2	1.7	66	0.7	82
D16	0.12	2.4	49	1096	0.12	1.1	1.4	71	1.7	29
D16-1	0.14	2.2	49	1031	0.10	1.1	1.7	67	1.2	51
D16-2	0.15	2.5	53	1240	0.11	1.1	1.7	67	1.2	51

Appendix 2. Continued.

(b) *Orconectes sp.*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D4	0.11	1.3	56	540	0.07	1.1	0.5	71	8.8	5
D5-2	0.14	1.6	81	870	0.07	1.3	0.7	73	5.6	13
D7	0.19	1.6	71	741	0.08	1.4	3.8	78	9.5	6
D7	0.15	1.8	79	856	0.09	1.3	0.9	74	5.2	11
D8	0.11	1.7	60	970	0.12	1.3	1.0	70	25.1	2
D8-2	0.15	1.3	39	683	0.08	1.7	0.6	60	71.1	1
D8-2	0.14	2.4	54	1186	0.09	1.6	1.0	64	47.2	1
D8-2	0.14	2.2	51	938	0.08	1.2	0.7	59	32.6	1
D8-2	0.04	2.0	45	980	0.10	1.2	1.1	66	37.6	1
D8-2	0.14	1.7	45	818	0.08	2.1	0.8	73	66.2	1
D8-2	0.10	1.5	46	705	0.10	1.0	0.6	62	54.2	1
D8-2	0.11	1.2	53	646	0.10	0.5	0.6	66	21.8	3
D8-2	0.17	2.7	44	1405	0.10	1.9	1.2	57	39.9	1
D8-2	0.13	1.8	44	974	0.10	1.6	0.9	62	67.7	1
D8-2	0.14	1.6	36	817	0.12	2.4	0.8	56	57.4	1
D8-2	0.09	1.5	49	807	0.10	0.6	0.7	64	52.1	1
D8-2	0.11	2.0	52	853	0.09	1.8	0.8	63	31.4	2
D10-2	0.06	1.9	52	811	0.06	0.6	0.8	65	26.0	1
D11	0.09	2.2	86	1206	0.07	1.9	0.6	71	31.7	2
D11	0.12	1.6	91	1101	0.11	0.5	0.6	67	34.9	2
D11	0.12	1.9	92	1498	0.08	1.3	0.7	78	21.2	2
D12	0.16	1.2	60	681	0.08	0.9	0.4	69	16.6	3
D12	0.13	1.8	55	1093	0.09	1.6	0.8	71	29.5	2
D14-2	0.04	1.2	63	385	0.90	0.4	0.5	66	32.8	1
D14-3	0.03	1.0	53	579	0.84	0.7	0.9	68	49.6	1
D15	0.03	1.4	64	815	0.18	0.3	0.6	69	45.9	1
D16	0.06	2.8	53	1473	0.17	1.3	2.2	81	24.8	1

Appendix 2. Continued.

(c) *Bythinia tentaculata*

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D1	0.40	8.9	656	3504	0.36	3.8	5.0	538	4.0	14
D2	0.24	6.7	279	2794	0.20	4.5	2.7	322	4.4	16
D2	0.26	6.9	329	2979	0.19	4.3	2.9	325	4.2	16
D4	0.31	4.7	166	2562	0.11	2.6	1.8	138	4.7	20
D4	0.28	5.1	142	2529	0.11	2.3	1.7	141	4.2	18
D6	0.46	12.1	221	3514	0.13	7.8	2.2	225	4.3	23
D6	0.47	25.2	215	4890	0.13	17.1	2.8	180	5.8	16
D7	0.70	11.5	399	3676	0.16	6.8	3.5	237	3.7	18
D7	0.96	10.0	429	3966	0.18	3.3	3.6	264	4.7	15
D7	0.73	11.2	343	3825	0.14	6.5	3.5	229	2.2	28
D8-1	0.93	12.9	728	3093	0.29	10.1	3.8	436	4.0	17
D8-2	0.91	8.5	664	2442	0.27	6.5	3.1	444	4.1	17
D8-3	0.83	6.9	565	2087	0.24	5.7	2.5	435	4.3	16
D9	0.78	5.6	264	2255	0.13	3.9	2.4	218	8.3	12
D9	1.00	7.8	295	3119	0.15	5.2	3.3	255	7.2	12
D10	0.21	7.7	270	3590	0.15	4.6	2.7	300	6.6	15
D12	0.95	11.7	565	4091	0.17	8.5	2.7	309	5.0	14
D13-1	0.37	5.8	189	2499	0.07	4.5	1.1	160	7.7	12
D13-2	0.33	7.1	173	3743	0.09	5.1	1.8	137	7.6	12
D13-3	0.36	6.0	188	2940	0.10	4.6	1.5	168	6.9	12
D14	0.55	5.7	212	2581	0.10	5.1	1.5	175	7.2	14
D16	0.44	9.0	615	4654	0.44	5.7	5.9	307	5.1	15
D16	0.43	7.8	524	4378	0.35	5.4	6.1	292	5.8	15

Appendix 2. Continued.

(d) Chironomidae

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D2	0.03	5.0	17	2643	0.10	3.1	3.4	102	2.8	17
D6	0.14	11.0	15	6286	0.05	6.8	4.4	68	2.0	33
D6	0.15	7.6	14	4446	0.04	4.6	3.3	67	0.8	78
D6	0.14	7.0	15	4220	0.04	4.2	2.9	79	0.7	99
D6	0.15	9.6	15	5565	0.05	6.1	4.1	66	2.0	31
D6	0.13	7.8	15	4965	0.06	4.7	3.6	64	2.2	26
D7	0.32	14.8	18	6218	0.10	6.4	7.0	69	1.0	42
D8	0.28	8.3	18	5377	0.11	5.2	5.5	84	0.5	96
D8	0.21	10.8	18	6328	0.14	6.6	6.8	78	1.1	44
D8-1	0.12	5.4	20	3099	0.11	3.4	3.5	75	0.9	61
D8-1	0.15	6.3	16	3105	0.10	3.7	4.0	74	0.9	57
D8-1	0.14	6.5	17	3968	0.11	4.2	4.2	72	0.8	70
D8-2	0.24	11.6	16	7143	0.16	7.3	8.0	67	1.3	41
D8-3	0.36	13.2	19	7823	0.14	8.6	8.5	83	0.8	55
D8-3	0.27	20.0	20	7500	0.13	7.4	7.7	82	0.8	68
D9	0.20	10.5	14	4129	0.08	4.5	4.7	63	1.6	37
D9	0.22	7.2	19	3325	0.06	4.0	4.0	79	0.5	109
D9-1	0.24	7.9	18	4384	0.07	4.3	4.4	70	0.8	86
D9-2	0.19	7.6	16	2403	0.07	4.6	4.1	72	0.7	89
D9-2	0.24	7.0	18	4126	0.07	4.1	3.5	71	0.7	96
D9-3	0.18	10.4	18	5079	0.07	4.7	4.8	75	0.7	67
D9-3	0.21	8.9	20	3858	0.07	4.5	4.3	77	0.6	75

Appendix 2. Continued.

(d) Chironomidae

Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D12	0.25	12.0	16	8326	0.12	7.9	6.1	72	1.2	54
D12	0.24	11.8	16	7317	0.12	7.1	6.0	70	1.3	47
D12	0.25	10.9	18	6743	0.11	6.7	5.0	78	0.7	45
D12	0.29	10.8	15	8137	0.12	7.1	5.8	69	1.2	49
D12	0.30	11.4	15	7763	0.11	6.9	5.8	70	1.1	55
D12	0.25	13.6	15	7956	0.11	7.3	5.6	76	1.4	19
D13	0.12	6.5	18	3817	0.04	4.2	2.3	83	0.6	97
D13-1	0.25	12.5	21	8300	0.08	7.8	4.6	63	1.5	34
D13-1	0.27	12.2	18	7322	0.09	7.3	4.1	62	1.5	28
D13-2	0.15	14.3	16	9160	0.07	7.4	4.2	60	1.3	31
D13-2	0.16	11.0	16	7849	0.08	6.7	3.6	68	1.4	39
D13-3	0.18	12.3	17	9338	0.06	8.0	4.0	68	1.1	56
D14	0.14	10.3	19	4182	0.83	4.7	7.7	72	1.6	27
D14	0.19	8.3	17	4836	2.02	4.8	8.7	88	1.6	34
D14	0.10	4.8	20	3435	0.59	3.2	5.9	78	0.8	90
D15	0.06	1.9	12	1449	0.04	0.6	0.7	109	1.4	17
D15	0.07	3.6	19	2036	0.08	2.1	2.1	72	0.9	65
D16	0.19	8.5	14	5458	0.17	5.3	7.2	87	2.3	22
D16	0.05	5.6	16	3584	0.16	3.2	4.2	88	0.7	40

Appendix 2. Continued.

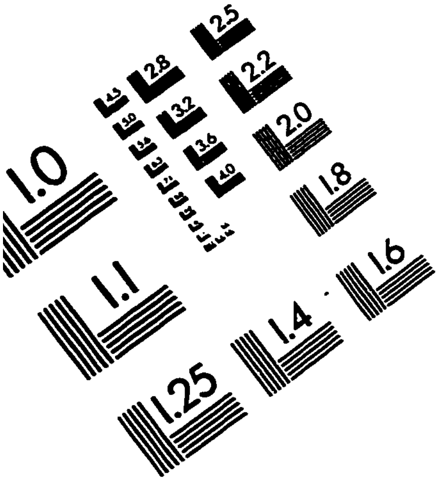
(e) Oligochaeta

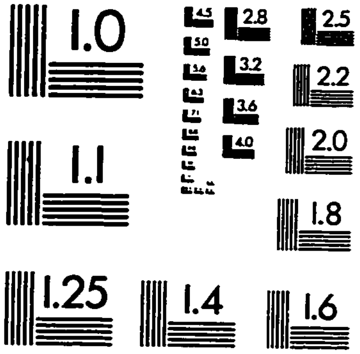
Site	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn	Body weight	*n*
D1	0.18	7.9	15	6236	0.07	3.7	5.9	145	24.1	3
D1	0.13	4.3	12	3835	0.08	2.2	4.1	115	17.1	4
D4	0.13	6.6	17	4313	0.05	3.5	2.5	113	92.7	1
D4	0.14	5.5	10	3452	0.04	3.2	2.3	86	34.5	3
D5	0.54	5.1	16	4854	0.09	2.6	3.5	140	19.5	4
D5	0.62	6.0	13	5370	0.08	3.0	2.9	108	32.3	3
D6	0.11	9.1	14	4664	0.04	5.4	3.0	88	35.0	2
D6	0.10	10.0	10	5694	0.03	6.3	3.4	80	63.3	1
D8	0.19	5.5	15	4870	0.08	2.7	4.7	104	12.5	6
D8	0.23	6.1	17	5235	0.08	3.3	4.7	133	61.1	1
D9	0.19	9.9	17	5828	0.04	5.6	4.1	100	48.3	2
D10	0.07	4.8	12	3487	0.06	3.4	4.2	110	30.9	2
D10	0.12	9.2	20	5430	0.08	5.6	4.8	138	22.6	3
D11	0.26	14.4	16	8717	0.07	8.4	6.5	73	42.0	2
D14-1	0.11	5.4	14	3097	0.29	3.7	3.4	95	65.7	1
D14-2	0.02	1.9	4	1620	0.14	1.6	1.4	67	71.4	1
D14-2	0.06	3.0	9	2527	0.18	2.0	3.2	101	32.7	2
D15-1	0.13	6.8	7	4073	0.07	4.8	3.5	64	96.5	1
D15-1	0.28	5.8	7	3309	0.06	4.3	4.1	61	64.2	1
D15-2	0.15	13.0	9	5944	1.53	8.6	6.1	61	87.5	1
D15-3	0.09	6.2	7	2822	0.05	4.4	3.8	64	75.9	1

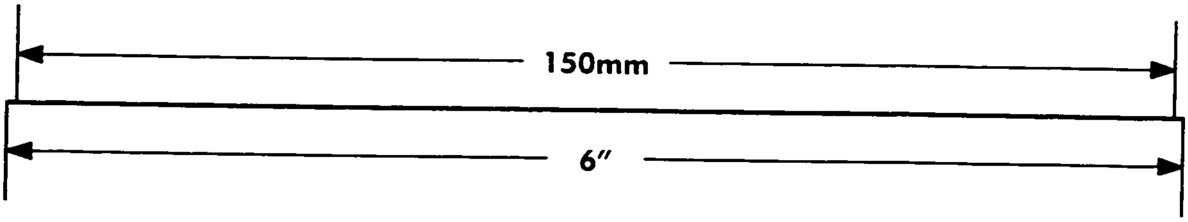
Appendix 3. Particle size distribution (% dry weight) and metal concentrations (µg/g dry weight) in upstream littoral sediments (in Lake St. Lawrence).

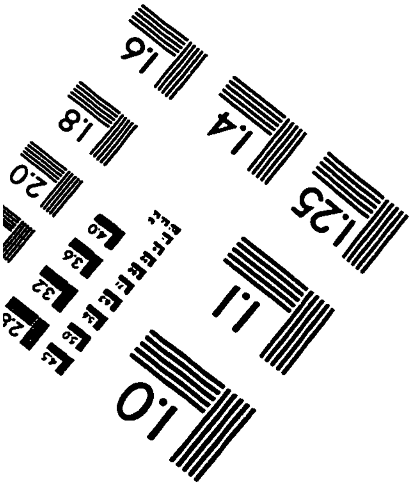
Site	Gravel	Sand	Silt and Clay	Cd	Cr	Cu	Fe	Hg	Ni	Pb	Zn
U17-1	1.4	86.7	11.9	0.16	18	16	15698	0.030	12	4.0	41
U17-2	0.5	80.3	19.2	0.17	19	13	18903	0.037	11	4.3	39
U17-3	3.6	88.7	7.7	0.15	14	8	12885	0.036	9	4.5	29
U18-1	1.2	56.2	42.6	0.19	23	12	19053	0.037	13	10.4	44
U18-2	2.1	74.2	23.7	0.14	19	10	16287	0.027	12	2.9	33
U18-3	2.7	60.0	37.3	0.20	27	15	23142	0.041	15	6.6	53
U19-1	0.3	94.8	4.9	0.05	5	5	6161	0.008	5	1.5	11
U19-2	5.4	74.5	20.1	0.11	12	8	11106	0.017	8	1.5	24
U19-3	1.5	88.8	9.7	0.06	7	4	7110	0.055	6	1.5	13
U20-1	1.0	88.6	10.4	0.09	17	8	14950	0.017	8	4.2	23
U20-2	1.6	87.6	10.8	0.08	12	9	10305	0.014	9	1.5	19
U20-3	4.8	80.6	14.6	0.12	22	11	16892	0.027	8	1.6	27
U21-1	1.9	89.8	8.3	0.13	11	10	11026	0.020	7	5.2	21
U21-2	5.1	88.3	6.6	0.13	11	9	10866	0.017	7	2.8	21
U21-3	12.3	76.0	11.7	0.14	13	10	17425	0.026	9	7.3	26
U22-1	0.8	77.9	21.3	0.28	21	13	15329	0.049	13	8.9	46
U22-2	0.5	80.4	19.1	0.32	27	24	22694	0.066	19	7.5	63
U22-3	2.3	81.7	16.0	0.21	16	9	13332	0.030	9	7.4	34
U23-1	0.8	80.8	18.4	0.18	17	12	14733	0.036	13	5.8	39
U23-2	0.7	85.7	13.6	0.19	18	12	14567	0.038	12	7.3	38
U23-3	1.7	86.0	12.3	0.13	15	12	13372	0.025	10	8.3	28
U24-1	0.2	82.1	17.7	0.16	19	10	15871	0.029	12	7.3	35
U24-2	0.0	83.1	16.9	0.14	18	11	15943	0.029	13	5.7	34
U24-3	1.7	78.2	20.1	0.17	18	12	15574	0.032	12	7.6	35

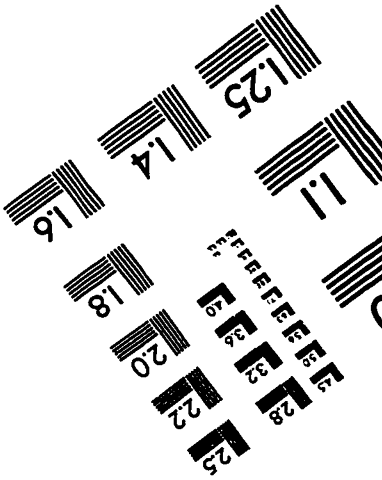
IMAGE EVALUATION TEST TARGET (QA-3)











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