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Contaminant Levels and Loading Estimates: the first step in
identifying restoration options for a Great Lakes Area of
Concern

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This publication reviews how two non-point sources of contamination were polluting the Dofasco boatslip. The run-off from the coal piles of Dofasco Inc. was badly contaminated with PAHs presumably at least partly from the oil sprayed onto the coal to suppress dust emissions. Dofasco cooperated with this study and after this study, they released a contract to trap and treat the runoff from these piles. Much of the work was done as part of Dr. Irvine's sabbatical from SUNY. His earlier work in Buffalo was partly on the efficiency of their coal pile drainage treatment. Although coal pile management in Hamilton Harbour is behind that in Buffalo, the other aspect of this study was the same in both cities; runoff from these industrial sites was badly contaminated with metals. In Hamilton, the combined storm sewer overflow of greatest metal concentrations did not drain Dofasco but this sewershed included recycling yards. Both types of non-point source contaminants make the effectiveness of dredging in the Dofasco Boatslip short-lived.

Summary

Annexes 1 and 2 of the 1987 Protocol to the U.S.-Canada Great Lakes Water Quality Agreement have emphasized the development of ecosystem objectives, maintenance, and restoration of the physical, chemical, and biological integrity of each of the Great Lakes. Contaminated sediment is one source of environmental impairment in the Great Lakes and sediment remediation often is considered as a restoration option. However, evaluation of current contaminant loadings and fate is an essential first step in assessing the potential for long term remediation success. The development of planning level loading estimates for selected metals and PAHs from various contaminant sources discharging to an area of Hamilton Harbour is illustrated. Contaminant sources included combined sewer overflows (CSOs), industrial cooling water discharge, coal pile runoff, industrial stormwater discharge, and groundwater discharge. Levels and loadings of total metals (Pb, Cr, Zn, Fe) and PAHs (fluoranthene, phenanthrene, benzo(a)pyrene, pyrene, chrysene) were determined for the period April 1 through October 31, 1996, using a combination of sampling, mathematical modeling, and literature review. The mean levels of all metals in the CSOs and coal pile runoff exceeded Canadian Water Quality Guidelines. Guideline exceedances of mean metals levels for the three cooling water discharge sites were: Pb, all sites; Fe, two of three sites; Zn, one of three sites; and Cr, no sites. Although the metals levels in the cooling water discharges were lower than other sampled sources, the large volume of discharged cooling water resulted in metals loadings that were greater than all other sources combined. Metals loadings for the study period from all sources combined were: 788 kg of Pb; 220 kg of Cr; 4,708 kg of Zn; and 12,715 kg of Fe. PAH levels were not determined for the cooling water discharges, but mean levels of fluoranthene, phenanthrene, pyrene, and chrysene in the particulate phase of the coal pile runoff exceeded the provincial "Severe Effect Level" for sediment. PAH levels and loadings in the coal pile runoff were greater than those measured for the CSOs. It appears that the current levels and loadings of selected

metals and PAHs are sufficient to negatively impact the long term viability of restoration efforts in this area of the harbour. Source control measures (e.g. stormwater best management practices) should be evaluated and implemented prior to initiation of costly sediment and habitat remediation.

Contaminant levels and loading estimates: The first step in identifying restoration options for a Great Lakes Area of Concern

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Introduction

Impairment of Great Lakes water quality has been documented since the 1800's and there have been efforts for almost as long to remediate and restore water quality (International Joint Commission (IJC), 1987; Rossi, 1995). Initially, the greatest concerns were associated with pathogenic contamination due to sewage discharges, but with a growing population and industrial activity, the focus of remediation shifted to cultural eutrophication, and subsequently to persistent contaminants (organics and metals). Although there is a long history of environmental impairment, there also have been success stories in water quality and habitat restoration throughout the Great Lakes. For example, point source control of phosphorus discharge and the efforts of the Pollution from Land Use Activities Reference Group (PLUARG) have helped to reverse cultural eutrophication problems in the lower Great Lakes (Yaksich and Rumer, 1980; IJC, 1987; Charlton, 1997). More recently, Annexes 1 and 2 of the 1987 Protocol to the U.S.-Canada Great Lakes Water Quality Agreement (IJC, 1987b) have emphasized the development of ecosystem objectives, maintenance, and restoration of the physical, chemical, and biological integrity of each lake (Kelso and Hartig, 1995). Management and remediation initiatives for the lakes have been focused through programs such as Remedial Action Plan (RAP) development for the 42 Areas of Concern (AOCs) and Lakewide Management Plans (LaMPs). Areas of Concern are designated by the IJC because they exhibit some type of environmental impairment. The IJC (1997) concluded that:

"Contaminated sediment is a major cause of environmental problems and a key factor in many impairments to beneficial uses of the Great Lakes. Based on application of chemical guidelines, all 42 Great Lakes Areas of Concern have contaminated sediment... A variety of sediment management options is available, ranging from source control and natural recovery to full-scale remediation depending on the severity of the problem... In recognition of the scope of this problem and the limited progress in addressing it, IJC identified contaminated sediment as a priority for the 1995-1997 biennial cycle."

Various *in situ* treatment and removal technologies are becoming available to remediate contaminated sediment (e.g. Randall, 1992; U.S. EPA, 1994; Murphy *et al.*, 1995; Zeman and Patterson, 1997), but thorough evaluation of current contaminant inputs and fate should be done before any remediation is considered, to help ensure that the remediation will produce the desired benefits. Sediment remediation is expensive and it is essential that a contamination problem does not recur some years after remediation because of a lack of source control.

Mass balance modeling increasingly has been applied in the Great Lakes as a management tool to help guide policies regarding contaminant loading reductions, exposure risks, and remediation options (e.g. DePinto, 1994; Diamond and Ling-Lamprecht, 1996; Diamond *et al.*, 1996; IJC, 1997). Mass balance modeling accounts for chemical inputs to, outputs from, and transformations within a defined body of water. An important component of mass balance modeling is the determination of contaminant inputs or loadings from the various possible sources. Although the loading estimates do not provide information on the transport and fate of the contaminant within the water body (DePinto, 1994), the estimates do provide important planning level insights to the potential magnitude of the contaminant problem and the relative importance of current contaminant sources. These planning level loading estimates can be derived with considerably fewer resources than a full mass balance, while at the same time acting as an initial step should a mass balance study be required.

The objective of this paper is to illustrate the development of planning level loading estimates for selected metals and PAHs from various contaminant sources discharging to an area of the Hamilton Harbour, as an essential first step in making aquatic restoration decisions. The harbour is an AOC and there is considerable interest in furthering the habitat and water quality improvements that have been made over the last 15 years (e.g. Charlton and Le Sage, 1996; McNeill, 1997).

Methods

Field site

The southern shore of Hamilton Harbour (Figure 1) is heavily industrialized, with steel production and steel-related industries being the predominant activities. The level of contamination in the bottom sediment varies greatly throughout the harbour. For example, PAH and PCB levels range from below detection to greater than 800 and 2,000 $\mu\text{g g}^{-1}$, respectively, in some "hot spots" near the industrial areas (Murphy *et al.*, 1991; RAP, 1992). Clearly, anthropogenic activity along the southern shore has nega-

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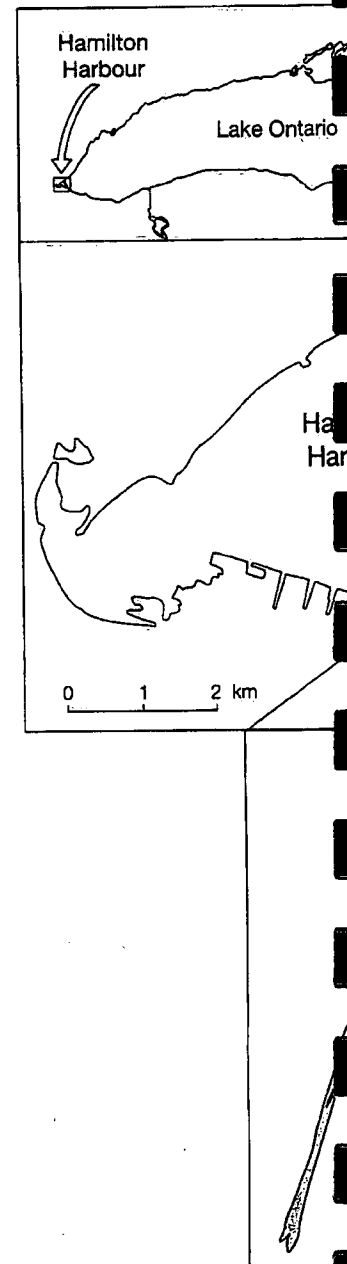


Fig. 1. Hamilton Harbour and local binned sewer outfall point; CP - co water discharge points.

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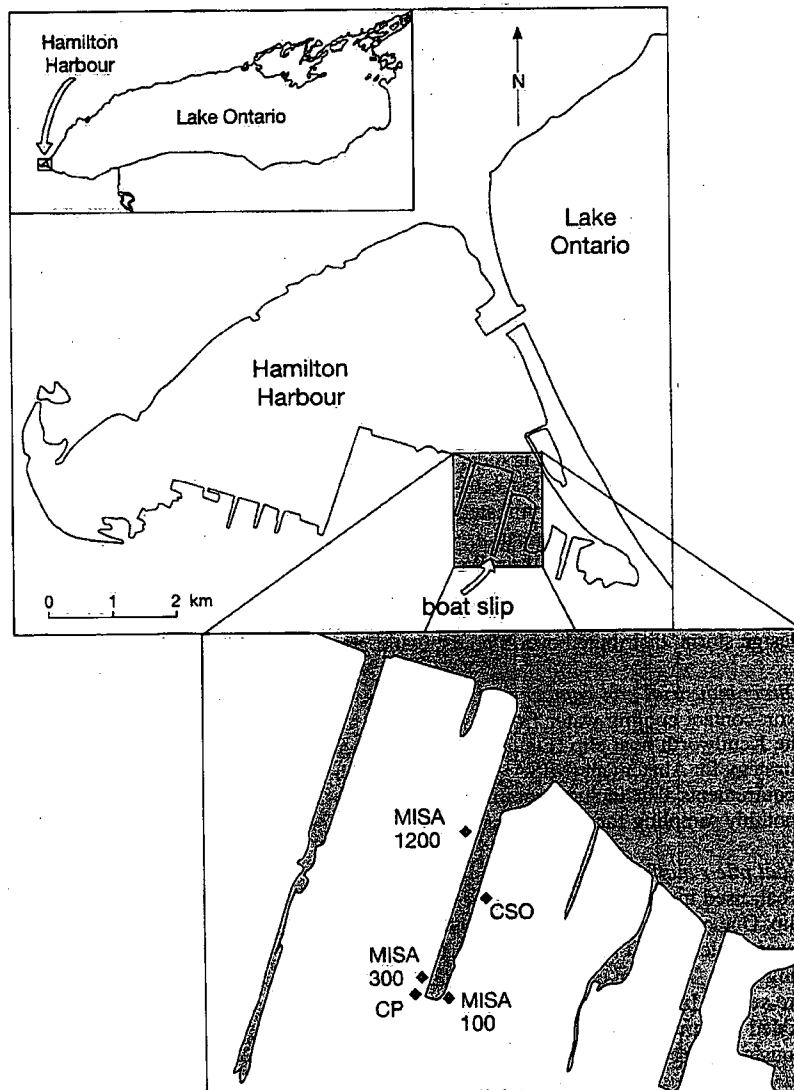


Fig. 1. Hamilton Harbour and location of sample sites within the Kenilworth boat slip. CSO – combined sewer outfall point; CP – coal pile tile drainage discharge point; MISA – permitted cooling water discharge points.

tively impacted aquatic habitat and biota and Holmes (1988) suggested that although a warm water fisheries rehabilitation might be done successfully in some parts of the harbour, the southern shore had no potential for rehabilitation. Despite Holmes' (1988) view of the potential for rehabilitation, and perhaps because of the more recent RAP efforts (RAP, 1992), there has been an increasing interest in remediating hot spot areas along the southern shore (e.g. Murphy *et al.*, 1995; McNeill, 1997).

One of these hot spot areas of interest is known as the Kenilworth boat slip, located at the south eastern end of the harbour (Figure 1). Our study focused on quantifying loadings of selected metals and PAHs to the boat slip from multiple sources that included combined sewer overflows (CSOs), direct discharge of industrial non-contact cooling water, untreated runoff from coal piles, stormwater runoff from adjacent industrial properties, and groundwater discharge.

Combined sewer overflows

The Kenilworth sewershed (Figure 2) has a contributing area of 265.5 ha that is serviced principally by a combined sewer system. The CSO discharge point is located in the north eastern section of the boat slip (Figure 1). Much of the land use in the upper part of the sewershed is older residential, single family dwellings, with commercial ribbons along major streets. The lower sewershed (approximately 9% of the total contributing area) is dominated by a mix of industry, including steel mills, rail-car manufacture, metal fabrication, construction material recycling, and lead recycling facilities. The Hamilton-Wentworth Pollution Control Plan (Paul Theil Associates and Beak Consultants, 1991) indicated that the Kenilworth sewershed is one of the largest CSO discharge points (by volume) to the harbour and that a 5 mm design storm, distributed over 2 hours, could generate an overflow event.

Direct industrial non-contact cooling water discharge

Non-contact cooling water from a major steel mill is discharged at three points to the Kenilworth boat slip (Figure 1). Under the provincial Municipal and Industrial Strategy for Abatement (MISA) program, these discharges must comply with permit requirements that include continuous monitoring of flow quantity and weekly to monthly sampling for various quality parameters.

Coal pile runoff

Coal, used in the production of steel, is stored in several locations around the boat slip. One of these storage fields, located to the south west of the boat slip, is drained by an underground tile system that discharges untreated runoff directly to the water (Figure 1). The total contributing area of this coal field is 1.9 ha and typically there are several large coal piles on site, although pile size continually changes as coal is added or removed. The flow volume from the tile drain system is monitored continuously as part of the MISA program.

Overland stormwater discharge and groundwater discharge

Overland runoff and groundwater from adjacent industrial property discharges directly to the boat slip. This runoff can originate from coal, iron ore, and limestone storage areas, oil bulking, scrap metal storage, and general industrial areas. The total contributing area for runoff is approximately 58.4 ha.



Fig. 2. The Kenilworth Sewershed

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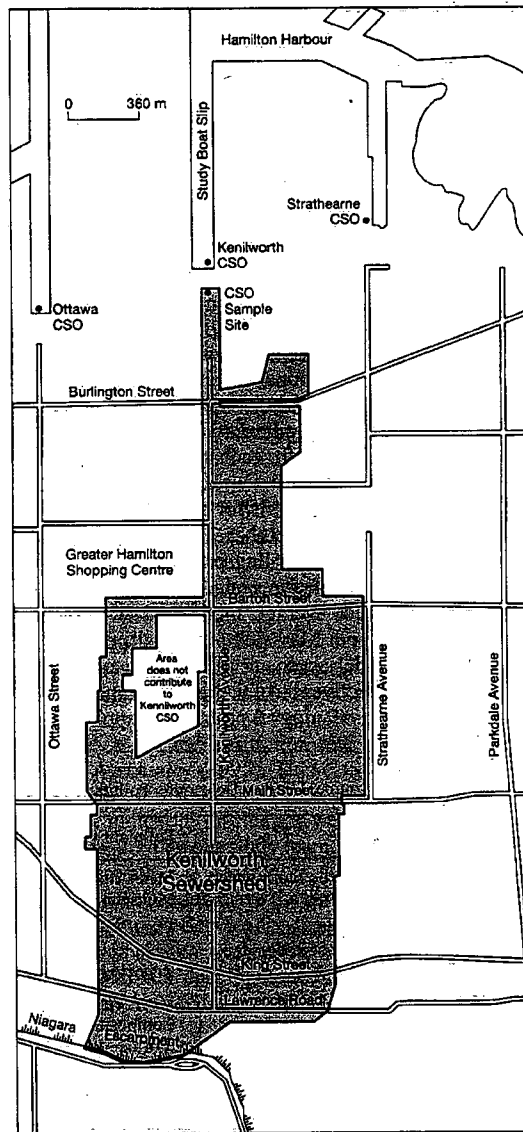


Fig. 2. The Kenilworth Sewershed and CSO sample site location.

Loading estimate approach

A simple, volumetric approach often is used to estimate contaminant loadings on an annual basis (e.g. Marsalek and Ng, 1989; Marsalek, 1990; Nix, 1990; Irvine et al., 1993; Pratt et al., 1995) according to the following equation:

$$L_j = \sum(Q_i \cdot C_{ji}) \quad (1)$$

where: L_j is the annual load of pollutant, j
 Q_i is the volume of each overflow event, i
 C_{ji} is the event mean concentration of pollutant, j

Clearly, there are two components in equation (1) that need to be determined; volume of flow and a *representative* pollutant concentration. The approaches taken in our study to determine these two components varied by source and are summarized in Table 1. Table 1 indicates, in fact, variations of equation (1) were used to calculate loadings, primarily due to sampling limitations. In general, a single arithmetic mean value calculated from all samples for each source was used to determine a *representative* pollutant concentration for that source. This representative concentration subsequently was multiplied by flow volume for the period of interest. Loading estimates were made for April 1 through October 31, 1996 since this was the general period covered under our first year sampling program. Marsalek (1990) reviewed several methods by which representative concentrations could be calculated for equation (1), including the direct average method, flow-weighted concentration method, and the regression method. Marsalek (1990) ultimately recommended use of a probability distribution model approach to define event mean concentrations. The probability approach is attractive, but for this preliminary study the data were not sufficient for a rigorous and meaningful probability application.

Table 1. Summary of Approaches, by Source, Used to Determine Pollutant Loadings.

Pollutant Source	Flow Estimate	Pollutant Concentration Estimate
CSO	Application of the Stormwater Management Model	Mean Value of Grab Samples over Multiple Storms
Direct Industrial Discharge	Continuously Measured by Industry and Reported under MISA	Mean Value of 24-hour Composite Samples Collected by Industry and Reported under MISA
Tile-Drained Coal Pile Runoff	Measured and Modeled	Mean Value of Flow-proportioned, Composite Samples for Multiple Storms
Overland Runoff	Application of the Stormwater Management Model	Literature Search, by Source-Area
Groundwater Discharge	Calculated in a Preliminary Hydrogeological Study of the Area	Literature Search, by Source-Area

Contaminant levels and load

Volume estimates

The CSO volumes to operate personal computer (PC) PCSWMM4) run in continuous 1986 (a typical rainfall year). PCSWMM is a dynamic, distributed route this runoff through a distributed RUNOFF computational block approach that considers infiltration surface runoff is routed through that considers both Manning and flow volume from the tiles within a discharge flow Loss Flowmeter. The instrumenterly in some cases and the multiple regression approach (r² = 0.74). The multiple (submitted).

Overland runoff to the basin using the RUNOFF block and overland runoff were determined for one of the local industrial land contributing areas were age areas discussed previously.

Groundwater discharge rate (Conestoga-Rovers & Associates area. These discharge rates within water flow system is in equilibrium the discharge volume. A study Conestoga-Rovers & Associates the relatively permeable fill).

Data for the direct industrial gram and were not measured mean values for each month.

The hourly rainfall data at the Atmospheric Environment ed approximately 10.5 km station gauge also were used in the Control Plan (Paul Theil Associates).

Water quality determination. Grab samples of the CSOs at pipe of the outfall mouth (Flow scoping sample pole that was analysis and 1,000 ml clean twinbox at the CSO sampling was done at the mid-point of

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Pollutant Loadings.

Pollutant Concentration Estimate

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Volume estimates

The CSO volumes to operationalize equation (1) were calculated using a calibrated personal computer (PC) Stormwater Management Model, version 4 (i.e. PCSWMM4) run in continuous mode. The general CSO modeling approach, using 1986 (a typical rainfall year) data, is described in detail by Irvine *et al.*, (1998). PCSWMM is a dynamic, deterministic model that can calculate surface runoff and route this runoff through a defined sewer network. Surface runoff is calculated in the RUNOFF computational block using Manning's equation and a non-linear reservoir approach that considers infiltration, surface storage, and evapotranspiration. The surface runoff is routed through the sewer network using a kinematic wave approach that considers both Manning's equation and a continuity equation.

Flow volume from the tile-drained coal pile site was measured at 15 minute intervals within a discharge flume using a recording Badger 2000 Obstructionless No-Loss Flowmeter. The instrumentation to measure flow volume did not operate properly in some cases and the flow volume for these events was estimated using a multiple regression approach that considered event rainfall depth and coal pile volume ($r^2 = 0.74$). The multiple regression analysis is discussed in detail by Curran *et al.* (submitted).

Overland runoff to the boat slip from adjacent industrial property was estimated using the RUNOFF block of PCSWMM. The contributing areas and direction of overland runoff were determined from a map produced by a private consulting firm for one of the local industries. For modeling and load estimate purposes, the overland contributing areas were divided according to surface type (i.e. the various storage areas discussed previously).

Groundwater discharge rates were taken from a consultant's preliminary report (Conestoga-Rovers & Associates, 1995) commissioned by one of the industries in the area. These discharge rates were calculated based on the assumption that the groundwater flow system is in equilibrium and the infiltrating volume therefore is equal to the discharge volume. A surface infiltration rate of 0.305 m yr^{-1} was assumed by Conestoga-Rovers & Associates (1995). This estimate was derived as a function of the relatively permeable fill material that makes up the water table unit of the area.

Data for the direct industrial discharges were obtained through the MISA program and were not measured by our study team. The flow data were reported as daily mean values for each month.

The hourly rainfall data used to drive the models in this study were obtained from the Atmospheric Environment Service for the gauge at Mount Hope Airport, located approximately 10.5 km south west of the boat slip. Data from the Mount Hope gauge also were used in the development of the Hamilton-Wentworth Pollution Control Plan (Paul Theil Associates and Beak Consultants, 1991).

Water quality determinations

Grab samples of the CSOs were collected at a manhole, approximately 110 m up-pipe of the outfall mouth (Figure 2). The grab samples were obtained using a telescoping sample pole that was fit with clean 500 ml polyethylene bottles for metals analysis and 1,000 ml clean amber glass bottles for PAH analysis. The sewer is a twinbox at the CSO sampling site, with each side being 2.13 m x 1.75 m. Sampling was done at the mid-point of the eastern box and it was assumed that both sides were

equally mixed. To the extent possible, multiple samples were collected at evenly spaced time periods through each CSO event. Sampling for metals began in May, 1996 and was completed at the end of October, 1996. Sampling for PAHs was conducted during October and November, 1996.

The Badger flowmeter in the coal pile discharge flume was connected to an American Sigma Streamline 700 pump sampler for the period June 1 through October 31, 1996. When flow in the flume reached a depth greater than 10 cm, the flowmeter triggered the pump sampler to begin collecting 200 ml samples at 10 m³ intervals. The pump sampler composited the 200 ml samples in a clean 10 l glass bottle for each event. These flow-proportioned composite samples were removed from the field as soon as possible (normally within 5 hours) after the end of each runoff event.

The MISA permit requires that weekly samples for most analytes of interest be collected by the steel manufacturer that discharges to the boat slip. These samples are 24-hour composites of the permitted direct discharge and the 1996 analytical results were obtained from the permitted industry. More recently, the MISA program also has required permitted industrial facilities to conduct a stormwater sampling program and develop stormwater management plans. The stormwater management report is archived by each industrial facility and is made available to the Ontario Ministry of Energy and Environment (MOEE) upon request. Although a sampling plan for stormwater runoff had been developed in 1996, results of the sampling were not available for our study. Therefore, *representative* contaminant concentrations of stormwater runoff and groundwater flow were determined using our CSO and coal pile runoff data as a guideline and also by considering the range of concentrations reported in the literature for different land uses.

Laboratory methods

Total metals levels in the CSO and coal pile runoff samples were determined by the National Laboratory for Environmental Testing, Burlington, Ontario, using inductively coupled argon plasma (ICAP) following Environment Canada Method 02-2001 (Environment Canada, 1994). Samples were fixed with 1 ml (1:1) Seastar nitric acid without filtration, upon retrieval from the field. All samples were concentrated 10 times and digested using nitric acid. Quantification was done using an Applied Research Laboratories (ARL) 3580 ICP optical emission spectrometer with a gas regulator suitable for argon and a Gilson 222 autosampler. The QA/QC procedures are described in detail by Curran *et al.* (submitted).

A 1 l sub-sample of the coal pile runoff composite was collected in the laboratory and was filtered through Whatman Grade #1 filter paper to separate the particulates from the water. The filtered particulates were analysed directly and henceforth the results are referred to as the particulate phase PAHs, while the PAHs in the filtrate are referred to as the dissolved phase PAHs. Analysis was performed by Wastewater Technology International (WTI), Burlington, Ontario, using Gas Chromatography-Mass Spectrometry (GC-MS) and adapted from U.S. EPA Method 624 (U.S. EPA, 1984) for the dissolved phase and U.S. EPA Method 8270 (U.S. EPA, 1994b) for the particulate phase.

For the dissolved phase analysis, the 1 l sub-sample was spiked with a surrogate mixture and serially extracted three times with dichloromethane under basic condi-

Contaminant levels and loadings

tions. The combined extracts evaporated from the extract combined with an internal standard the sediment was spiked with a toluene-hexane (59:41) mix. The carbonate solution. The aqueous organic fractions combined. Total sulphate and concentrated

For both the dissolved and particulate phases, a Packard 5890 operated with a 6 degrees/min. to 285 °C, hold rowbore column with a 0.25 mm ID. MS was either a Hewlett Packard source temperature of 200 °C, interface temperature of 280 °C, and the analytical methodology and

The 1 l samples from the coal pile therefore reflect "total" concentrations. The analytical methodology followed the coal pile samples.

Results

Flow volume

The estimated and measured flow volumes from the various sources for the year 1986 are summarized in Table 2. The CSO flow for the entire year, 1986 totaled 775 m³. The 1986 was representative of the flow for the 1996 study period totaled 775 m³.

The PCSWMM modeling indicated that the principal source-area for the CSO striking the pervious industrial site was the preliminary infiltration from Rovers & Associates (1995).

Table 2. Measured/Estimated Flow Volume, m³, October 31, 1996.

	MISA Site 100	MISA Site 300
Flow Volume, m ³	5,780,132	1,858,634

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tions. The combined extracts were dried over sodium sulphate. The solvent was evaporated from the extract to a final volume of 3 ml. The extract was then combined with an internal standard and analysed by GC-MS. For the particulate phase, the sediment was spiked with a surrogate mixture and Soxhlet-extracted with an acetone-hexane (59:41) mix. The extract was base-partitioned with 2% potassium bicarbonate solution. The aqueous medium was then back-extracted with hexane and the organic fractions combined. The combined organic extract was dried through sodium sulphate and concentrated for analysis by GC-MS.

For both the dissolved and particulate phase analyses, the GC was a Hewlett Packard 5890 operated with an initial temperature of 30 °C, hold for 1 min., ramp at 6 degrees/min. to 285 °C, hold for 16.5 min. The GC column was a DB-5, 30 m narrowbore column with a 0.25 mm internal diameter and a 0.25 µm film thickness. The MS was either a Hewlett Packard 5988 operated in electron impact mode with an ion source temperature of 200 °C, or a Hewlett Packard 5971 operated with a GC/MS interface temperature of 280 °C. Curran *et al.* (submitted) provide greater detail on the analytical methodology and QA/QC for the PAH analysis.

The 11 samples from the CSOs were not filtered and the PAH concentrations therefore reflect "total" concentration in these samples. Apart from this difference, the analytical methodology followed that described above for the dissolved phase of the coal pile samples.

Results

Flow volume

The estimated and measured flow volumes discharging to the Kenilworth boat slip from the various sources for the period April 1, 1996 through October 31, 1996 are summarized in Table 2. The CSO volume is comparable to the overflow volume, estimated for the entire year, 1986 (311,000 m³), from Irvine *et al.* (1998). The rainfall for the year 1986 totaled 775 mm and the monthly distribution of rainfall through 1986 was representative of the 30-year norm for the Hamilton area. The rainfall for the 1996 study period totaled 713 mm.

The PCSWMM modeling indicated that an area of buildings and paved parking was the principal source-area for surface runoff and that nearly all of the rainfall striking the pervious industrial land would infiltrate. These results are consistent with the preliminary infiltration/groundwater study done for the area by Conestoga-Rovers & Associates (1995).

Table 2. Measured/Estimated Flow Volumes to the Kenilworth Boat Slip, by Source, April 1 through October 31, 1996.

	MISA Site 100	MISA Site 300	MISA Site 1200	Coal Pile Runoff	CSO	Surface Runoff	Ground- water
Flow Volume, m ³	5,780,132	1,858,634	39,427,471	9,690	318,000	37,870	277,289

Metals and PAH levels

The mean and standard deviation of total Pb, Cr, Zn, and Fe levels in samples collected during the study period are presented in Table 3. Although other metals were analysed in the CSO and coal pile runoff samples (see Irvine *et al.*, 1998; Curran *et al.*, submitted), we have focused on these four metals for illustrative purposes because they are the only metals that are reported under the MISA program. The metals levels shown in Table 3 for the MISA sites were adjusted by subtracting a concentration representative of ambient levels in the harbour. This downward adjustment accounts for the metals levels that exist in the harbour water used for cooling purposes and the data in Table 3 therefore reflect only the metals levels added by industrial activity. The representative ambient harbour levels were calculated using data from Poulton (1987); RAP (1992); and Diamond and Ling-Lemprecht (1996), and were as follows: Pb - 0.0033 mg l⁻¹; Cr - 0.0009 mg l⁻¹; Zn - 0.014 mg l⁻¹; Fe - 0.22 mg l⁻¹.

Canadian Water Quality Guidelines (Canadian Council of Resource and Environment Ministers, 1993) for the individual metals also are shown in Table 3. The mean levels of all metals in the CSO and coal pile runoff samples exceeded these guidelines. The mean levels of Pb for the three direct industrial discharges exceeded the guidelines, while the mean levels of Cr for the same industrial discharges did not exceed the guidelines. Mean levels of Fe exceeded guidelines at two of three industrial discharge points and mean levels of Zn exceeded guidelines at one of three industrial discharges.

Samples of surface and groundwater runoff from the industrial properties were not collected for quality analysis. A literature review was conducted to determine a range of possible metals levels for these sources and the results are presented in Table 4. Our data for the CSO and coal pile discharges also were considered in determining representative metals levels for the surface and groundwater runoff and the results of our analysis are shown in Table 5.

Table 3. Metals Levels (mg l⁻¹) for Samples Collected during the Study Period*.

	Pb, Mean	Pb, S.D.	Cr, Mean	Cr, S.D.	Zn, Mean	Zn, S.D.	Fe, Mean	Fe, S.D.
MISA 100	0.0158 (n=28)	0.00019	0.0075 (n=6)	0.0103	0.0240 (n=28)	0.0171	0.633 (n=6)	0.555
MISA 300	0.0158 (n=25)	0.00019	0.0025 (n=6)	0.0029	0.0176 (n=25)	0.0137	0.363 (n=6)	0.350
MISA 1200	0.0154 (n=27)	0.00087	0.00371 (n=6)	0.0040	0.1021 (n=27)	0.0914	0.0571 (n=6)	0.0499
CSO	0.0902 (n=20)	0.0777	0.0244 (n=20)	0.0330	0.676 (n=20)	0.887	8.7 (n=20)	12.42
Coal Pile	0.022 (n=15)	0.018	0.024 (n=15)	0.024	0.28 (n=15)	0.30	15.5 (n=15)	14.4

* Canadian Water Quality Guidelines (for Protection of Aquatic Life) are: Pb - 0.007; Cr - 0.02; Zn - 0.03; Fe - 0.3 mg l⁻¹
S.D. - Sample Standard Deviation

Contaminant levels and loc.

Table 4. Metals Levels (mg l⁻¹) in Land Uses).

	Pb
Typical Range	0.1-0.

Sources: Weeks, 1982; Reinertsen, 1982; M and Maunder, 1993; Bannerman et al., 199

Table 5. Representative Metals L Property Surface and Groundwater

Surface and Groundwater Runoff Industrial Property
Groundwater Runoff from Coal St
Groundwater Runoff from Iron Or

The mean of selected PAHs marized in Table 6. Although focus on five of the PAHs benzo(a)pyrene) for illustrati detailed discussion because t deviations were not calculate Data on PAH levels are not although benzo(a)pyrene and other discharge points from tl

Canadian Water Quality C but guidelines for PAHs in sec by Curran *et al.* (submitted), r chrysene in the particulate p MOEE "Severe Effect Level"

Table 6. Mean Levels of Selected P.

	Fluora
CSO (total sample, µg l ⁻¹)	0.1 (n=)
Coal Pile Runoff (particulate phase, µg g ⁻¹)	20+ (n=)
Coal Pile Runoff (dissolved phase, µg l ⁻¹)	4. (n=)

and Fe levels in samples collected. Although other metals were analyzed (Irvine et al., 1998; Curran et al., 1996), they are not presented here for illustrative purposes because of the high variability of the data. A program was developed to subtract a concentration from the total concentration to account for downward adjustment accounts for cooling purposes and the effect of industrial activity. The results using data from Poulton (1996), and were as follows: Zn = 0.17 mg l⁻¹; Fe = 0.22 mg l⁻¹. The results also are shown in Table 3. The runoff samples exceeded direct industrial discharges for the same industrial discharges. The runoff samples exceeded guidelines at two sites and exceeded guidelines at one site.

The industrial properties were analyzed to determine a baseline. The results are presented in Table 4. The runoff samples were also considered in determining groundwater runoff and the

Study Period*.

Zn, S.D.	Fe, Mean	Fe, S.D.
0.0171	0.633 (n=6)	0.555
0.0137	0.363 (n=6)	0.350
0.0914	0.0571 (n=6)	0.0499
0.887	8.7 (n=20)	12.42
0.30	15.5 (n=15)	14.4

*Cr = 0.02; Zn = 0.03;

Table 4. Metals Levels (mg l⁻¹) in Runoff from Urban Environments (with an Emphasis on Industrial Land Uses).

	Pb	Zn	Cr	Fe
Typical Range	0.1-0.6	0.2-0.7	0.001-0.58	4-7

Sources: Weeks, 1982; Reinertsen, 1982; Marsalek and Ng, 1989; Granier et al., 1990; Xanthopoulos and Hahn, 1993; D'Andrea and Maundier, 1993; Bannerman et al., 1993

Table 5. Representative Metals Levels (mg l⁻¹) used for Calculation of Loadings from Industrial Property Surface and Groundwater Runoff.

	Pb	Zn	Cr	Fe
Surface and Groundwater Runoff from General Industrial Property	0.1	0.5	0.02	9
Groundwater Runoff from Coal Storage Areas	0.022	0.28	0.024	16
Groundwater Runoff from Iron Ore Storage Areas	0.1	0.5	0.02	16

The mean of selected PAHs in samples collected during the study period are summarized in Table 6. Although we analysed for 16 different PAHs, it was decided to focus on five of the PAHs (fluoranthene, pyrene, chrysene, phenanthrene, and benzo(a)pyrene) for illustrative purposes. These five PAHs were selected for more detailed discussion because they frequently are reported in the literature. Standard deviations were not calculated for the PAH data because of the small sample size. Data on PAH levels are not collected at the MISA sites of interest to our study, although benzo(a)pyrene and naphthalene loading limits have been identified for other discharge points from the steel mill.

Canadian Water Quality Guidelines have not been developed for PAHs in water, but guidelines for PAHs in sediment have been established (MOEE, 1993). As noted by Curran et al. (submitted), mean levels of fluoranthene, phenanthrene, pyrene, and chrysene in the particulate phase of the coal pile runoff (Table 6) exceeded the MOEE "Severe Effect Level" for sediment samples.

Table 6. Mean Levels of Selected PAHs for Samples Collected During the Study Period.

	Fluoranthene	Phenanthrene	Benzo(a)pyrene	Pyrene	Chrysene
CSO (total sample, µg l ⁻¹)	0.99 (n=2)	0.52 (n=2)	0.35 (n=2)	0.90 (n=2)	0.64 (n=2)
Coal Pile Runoff (particulate phase, µg g ⁻¹)	208.2 (n=4)	284.3 (n=4)	71 (n=4)	139.9 (n=4)	46.6 (n=4)
Coal Pile Runoff (dissolved phase, µg l ⁻¹)	4.5 (n=6)	6.1 (n=6)	0.9 (n=6)	3.1 (n=6)	0.7 (n=6)

Metals and PAH loadings

Using the volumetric approach outlined by equation (1), the metals loadings (kg) for the study period are summarized, by source, in Table 7. The percentage contribution of each source also is summarized in Figure 3. Because of the limited PAH data, loadings were calculated only for the CSO and coal pile discharges. Again, using the volumetric approach outlined by equation (1), the PAH loadings for these two sources are summarized in Table 8. In the case of the coal pile discharge, PAH loadings are the sum of the particulate and dissolved phase loadings.

Conclusions and recommendations

Contaminant levels and loadings both must be considered in assessing the potential impact of current sources on the success of sediment and aquatic remediation in the Kenilworth boat slip. Sampled metals levels generally were lower in the cooling water discharge than in the CSOs or coal pile runoff. In fact, for some metals (e.g. Cr and Zn), the mean levels typically did not exceed Canadian Water Quality Guidelines in the cooling water discharge. On the other hand, mean levels of all metals in the CSOs and coal pile runoff exceeded Canadian Water Quality Guidelines. Metals levels exceeding the Canadian Water Quality Guidelines potentially could present some acute toxicity problems. The metals levels reported in our study represent total concentrations, as this is the standard used under the Canadian Water Quality Guidelines. However, it has been argued that because of bioavailability

Table 7. Calculated loadings (kg) to the Kenilworth Boat Slip, by Source, April 1 through October 31, 1996.

	Pb	Fe	Cr	Zn
CSO	28.7	2,767	7.76	215
Coal Pile Runoff	0.213	150	0.238	2.72
Cooling Water Discharge*	729	6,338	206	4,337
Surface Runoff, Industrial Property	3.79	341	0.757	18.9
Groundwater Runoff	26	3,119	5.63	134

* Sum of all three MISA sites

Table 8. Calculated Loadings (kg) of PAHs to the Kenilworth Boat Slip, by Source, April 1 through October 31, 1996.

	Fluoranthene	Phenanthrene	Benzo(a)pyrene	Pyrene	Chrysene
CSO	0.315	0.165	0.111	0.286	0.204
Coal Pile Runoff	4.457	6.086	1.512	2.995	0.995

Contaminant levels and loadings

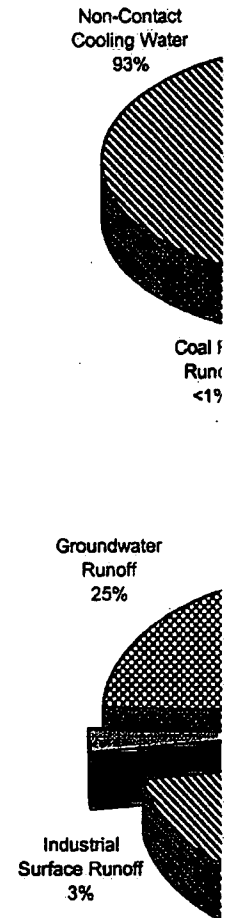


Fig. 3. Relative loading contribution of sources to the Kenilworth Boat Slip. Zn were similar to that of Pb.

effects, the use of total metals data is not recommended. In response to this concern, the use of dissolved metals to set and meet water quality objectives (MISA 1997). Although use of dissolved metals to set and meet water quality objectives is a bioavailability issue, there is

), the metals loadings (kg) for . The percentage contribution use of the limited PAH data, e discharges. Again, using the PAH loadings for these two coal pile discharge, PAH load- e loadings.

ered in assessing the potential and aquatic remediation in the ly were lower in the cooling In fact, for some metals (e.g. eed Canadian Water Quality r hand, mean levels of all met- an Water Quality Guidelines. Guidelines potentially could ls reported in our study repre- d under the Canadian Water at because of bioavailability

by Source, April 1 through October

Cr	Zn
7.76	215
0.238	2.72
206	4,337
0.757	18.9
5.63	134

oat Slip, by Source, April 1 through

(a)pyrene	Pyrene	Chrysene
1.111	0.286	0.204
1.512	2.995	0.995

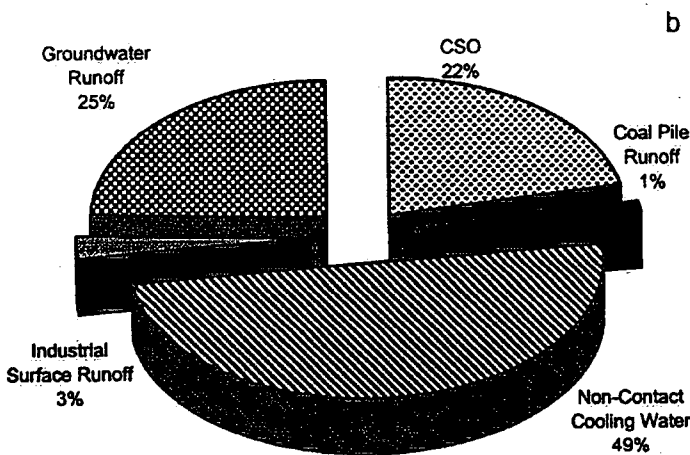
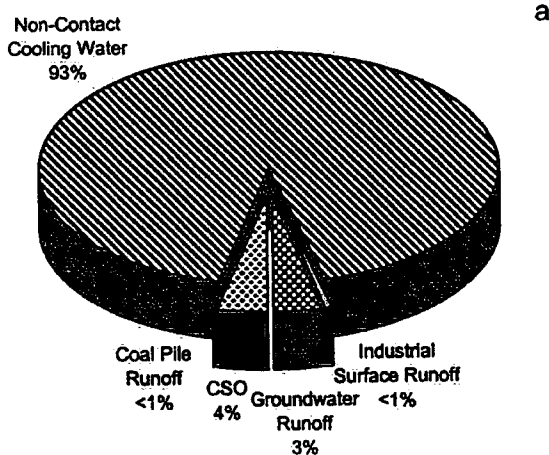


Fig. 3. Relative loading contributions, by source, for: (a) Pb; and (b) Fe. The contributions for Cr and Zn were similar to that of Pb.

effects, the use of total metals is overly conservative in developing water quality criteria. In response to this concern, the U.S. EPA recently has allowed the use of dissolved metals to set and measure compliance with water quality standards (Renner, 1997). Although use of dissolved phase metals addresses some aspects of the bioavailability issue, there are a host of natural factors that also can affect toxicity

(e.g. pH, temperature, dissolved oxygen, presence of organic and inorganic ligands). Furthermore, it has been argued that the free metal ions (e.g. Cu^{2+}) are most important in governing toxicity (e.g. Allen and Hansen, 1997; Renner, 1997). Given the relatively low levels of metals in the cooling water discharge, a study on the bioavailability of these metals should be conducted before major capital expenditures are undertaken to alleviate possible acute toxicity problems.

Because of the large volumes of water used for cooling purposes, the metals loads in the cooling water (even when adjusted for ambient metals levels in the harbour) were greater than all other sources combined. The large loading associated with the cooling water discharge potentially may represent a more chronic type of aquatic and sediment degradation, particularly if the metals settle and accumulate in the bed sediment of the boat slip. The large volume of cooling water containing relatively lower levels of metals may be difficult to treat with current engineering approaches. Other aspects of the large loadings associated with the cooling water also should be considered. For example, the high Fe loadings may contribute to the brown colour of the harbour water (in addition to humic matter from wetlands areas in tributaries). The Fe also may enhance precipitation of phosphorus and reduce algal growth.

Data on PAH levels were collected for two sources, CSOs and coal pile runoff. PAH levels in the coal pile runoff were greater than those measured for the CSOs. Although the mean levels of fluoranthene, phenanthrene, pyrene, and chrysene in the particulate phase of the coal pile runoff exceeded the provincial Severe Effect Level for sediment, there is some uncertainty as to the bioavailability of coal-associated PAHs (e.g. Bender *et al.*, 1987; Chapman *et al.*, 1996). It is possible that the levels of PAHs observed in the particulate phase of the coal pile runoff may have less impact upon the aquatic biota than the provincial guidelines suggest and detailed, site-specific bioavailability testing should be done. The PAH loadings from the coal pile runoff typically were an order of magnitude greater than for the CSOs, primarily because of the higher levels associated with the particulate phase. Given the smaller volume of runoff that would have to be treated and the hydrophobic nature of PAHs, construction of settling basins to contain the coal pile discharge could be an effective loading reduction tool.

This study provided a planning level evaluation of levels and loads associated with different sources that discharge to the Kenilworth boat slip. We were not able to quantify the impact of direct atmospheric deposition to the boat slip and harbour. In some areas of the Great Lakes, direct deposition has been considered an important source for certain contaminants (e.g. Kelly *et al.*, 1991; Zhang *et al.*, 1993). Atmospheric deposition on the urban surfaces would be integrated in the CSO discharges. In addition to impacting water quality, atmospheric emissions also may have a negative effect on human health (e.g. Chiras, 1991). Because of the planning level nature of this study, there clearly will be some uncertainty in the loading estimates for the sources that we examined. The greatest uncertainty is associated with those sources for which representative contaminant levels were obtained from a literature review. Nonetheless, we expect that our estimates are within the correct order of magnitude. It appears that the current levels and loadings of selected metals and PAHs are sufficient to negatively impact sediment or aquatic habitat remediation efforts in the boat slip area. Although the contaminated sediment could be removed

and treated, or treated *in situ* sources are not reduced.

Hamilton-Wentworth Regi should greatly reduce the nun (e.g. Stirrup, 1996). Likewise grams have been implemented encouraged as a means of However, given the volume o be expensive. Stormwater bes (including coal pile runoff) al future may be the most techni

The contaminant sources Lakes AOCs and the general this paper could be applied should be done at all sites prio iment remediation work.

Summary

Annexes 1 and 2 of the 1987 I Agreement have emphasized nance, and restoration of the the Great Lakes. Contaminat in the Great Lakes and sedin option. However, evaluation o first step in assessing the pote ment of planning level loadin; contaminant sources dischar; Contaminant sources included water discharge, coal pile run- discharge. Levels and loading thene, phenanthrene, benzo(a period April 1 through Octob; matical modeling, and literat and coal pile runoff exceed exceedances of mean metals l Pb, all sites; Fe, two of three the metals levels in the cool sources, the large volume of that were greater than all other od from all sources combined 12,715 kg of Fe. PAH levels but mean levels of fluoranth late phase of the coal pile run sediment. PAH levels and loa measured for the CSOs. It ap

nic and inorganic ligands). (e.g. Cu²⁺) are most important (Renner, 1997). Given the importance, a study on the bioavailability and capital expenditures are

ing purposes, the metals and nonmetals levels in the harbor are large loading associated with a more chronic type of loading settle and accumulate in the cooling water containing relationship with current engineering practices and the cooling water discharges may contribute to the matter from wetlands areas of phosphorus and reduce

CSOs and coal pile runoff. The levels measured for the CSOs, pyrene, and chrysene in the provincial Severe Effect Level (availability of coal-associated runoff, 1996). It is possible that the coal pile runoff may have less loading than suggested and detailed, PAH loadings from the coal pile runoff than for the CSOs, particulate phase. Given the hydrophobic nature of coal pile discharge could be

levels and loads associated with the boat slip. We were not able to measure the boat slip and harbour. It has been considered an important issue (Zhang *et al.*, 1993). The data integrated in the CSO discharge and emissions also may be important. Because of the planning uncertainty in the loading estimates, the data were obtained from a literature within the correct order of magnitude of selected metals and aquatic habitat remediation sediment could be removed

and treated, or treated *in situ*, the contamination problem will return if current sources are not reduced.

Hamilton-Wentworth Region's Pollution Control Plan, when fully implemented, should greatly reduce the number and volume of CSOs discharging to the boat slip (e.g. Stirrup, 1996). Likewise, industrial pretreatment and pollution prevention programs have been implemented in the past (RAP, 1992) and should continue to be encouraged as a means of reducing loadings from direct industrial discharges. However, given the volume of water involved, current approaches to treatment may be expensive. Stormwater best management practices for industrial property runoff (including coal pile runoff) also should be vigorously pursued and for the immediate future may be the most technically feasible and cost-effective source control option.

The contaminant sources evaluated in this study are common to many Great Lakes AOCs and the general planning level evaluation methodology discussed in this paper could be applied to any of these sites. Quantitative source evaluations should be done at all sites prior to the planning and implementation of habitat or sediment remediation work.

Summary

Annexes 1 and 2 of the 1987 Protocol to the U.S.-Canada Great Lakes Water Quality Agreement have emphasized the development of ecosystem objectives, maintenance, and restoration of the physical, chemical, and biological integrity of each of the Great Lakes. Contaminated sediment is one source of environmental impairment in the Great Lakes and sediment remediation often is considered as a restoration option. However, evaluation of current contaminant loadings and fate is an essential first step in assessing the potential for long term remediation success. The development of planning level loading estimates for selected metals and PAHs from various contaminant sources discharging to an area of Hamilton Harbour is illustrated. Contaminant sources included combined sewer overflows (CSOs), industrial cooling water discharge, coal pile runoff, industrial stormwater discharge, and groundwater discharge. Levels and loadings of total metals (Pb, Cr, Zn, Fe) and PAHs (fluoranthene, phenanthrene, benzo(a)pyrene, pyrene, chrysene) were determined for the period April 1 through October 31, 1996, using a combination of sampling, mathematical modeling, and literature review. The mean levels of all metals in the CSOs and coal pile runoff exceeded Canadian Water Quality Guidelines. Guideline exceedances of mean metals levels for the three cooling water discharge sites were: Pb, all sites; Fe, two of three sites; Zn, one of three sites; and Cr, no sites. Although the metals levels in the cooling water discharges were lower than other sampled sources, the large volume of discharged cooling water resulted in metals loadings that were greater than all other sources combined. Metals loadings for the study period from all sources combined were: 788 kg of Pb; 220 kg of Cr; 4,708 kg of Zn; and 12,715 kg of Fe. PAH levels were not determined for the cooling water discharges, but mean levels of fluoranthene, phenanthrene, pyrene, and chrysene in the particulate phase of the coal pile runoff exceeded the provincial "Severe Effect Level" for sediment. PAH levels and loadings in the coal pile runoff were greater than those measured for the CSOs. It appears that the current levels and loadings of selected

metals and PAHs are sufficient to negatively impact the long term viability of restoration efforts in this area of the harbour. Source control measures (e.g. stormwater best management practices) should be evaluated and implemented prior to initiation of costly sediment and habitat remediation.

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Bioremediation

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Introduction

Microbial degradation is a natural process that breaks down pollutants from the environment (Gibson and Subramanian, 1992). Biodegradation usually are used to degrade a wide range of organic pollutants (Bartha, 1992). Nevertheless, the biodegradation of Bunker C oil in 1970 from the time, a significant fraction of the oil was degraded (Prince et al., 1998). Bioremediation strategies have been used in Alaska, following the Prudhoe Bay oil spill (Atlas and Bartha, 1992; Prudhoe Bay Bioremediation Trials (Swannell et al., 1999). Bioremediation to be one of the most effective methods (Swannell and Head, 1994). The benefits and limitations relative to its application. This review discusses the principal technologies, highlights the principal factors to consider in

Development and evaluation

There are two main approaches to bioremediation (seeding) involves the addition of

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