



## Assessment of decadal changes in sediment contamination in a large connecting channel (Detroit River, North America)



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### HIGHLIGHTS

- A stratified random design was used to track temporal changes in sediment contamination.
- Geospatial clustering provided a refined picture of local contaminated and clean zones.
- Sediment contamination in the Detroit River remained stable over the past decade.

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### ABSTRACT

Concentrations of selected heavy metals (Cd, Cu, Hg, Pb, Zn) and organic contaminants (PCBs, PAHs) were investigated in samples from the Detroit River (Great Lakes, North America) in 1999 and 2008/09 collected using a stratified random sampling design. Getis–Ord geospatial analysis was used to further establish locations of areas demonstrating significantly high and low contaminant concentrations in the river. Based on the stratified random sampling design, a majority of the examined metals and organic contaminants demonstrated little or no trends with respect to regional sediment concentrations and river-wide mass balances over the investigated time interval. The Getis–Ord analysis revealed local scales of contaminated and clean areas which did not conform to the original strata used in the geostatistical sampling design. It is suggested that geospatial analyses such as Getis–Ord be used in the design of future sediment quality surveys to refine locations of strata that can simultaneously address sediment recovery over system-wide, regional and local spatial scales.

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### 1. Introduction

Contaminated sediments are a priority issue in many riverine systems and connecting channels impacted by urban development and heavy anthropogenic use (Taylor and Owens, 2009). Evaluation of temporal changes of sediment contamination is crucial to answer questions about the state of the system, whether it is improving in time or to determine if previously completed remediation activities have benefited the system. However, tracking temporal changes of sediment contamination in large riverine systems and connecting channels presents many logistic and technical challenges. Such systems exhibit complex hydrological properties involving altered flow in channels and deposition zones that contribute to heterogeneity in particle size, composition and degree of sediment consolidation. In addition, flow interruptions, storm events and other perturbations such as ice scouring or shipping can result in stochastic resorting of particles and downstream flux

of historically deposited sediments and associated contaminants (Reitsma, et al., 2002). The latter can easily displace contaminant deposits previously identified at micro- and macro scales.

Across sediment contamination studies there are two main categories of sampling designs used: judgmental and probability-based designs (EPA, 2002). The judgmental design involves selection of sampling locations on the basis of expert knowledge or results of previous studies which target known contaminated deposits or areas in proximity to point sources. Probability-based designs apply sampling theory and randomization of sampling locations. Judgmental sampling is often performed to delineate areas of sediment remediation or is associated with post-monitoring applications related to clean-up actions (e.g. Besser et al., 1996). Probability based sampling designs are used address questions about the state of the system (e.g. mass balance) and can identify spatial structure across multiple scales of analysis (Drouillard et al., 2006; Szalinska et al., 2006; others). While judgmental designs maximize sampling intensity and resolution at specific areas of interest, they contribute to biased interpretation about the overall state of the system (i.e. overemphasize contaminated

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areas) and can be confounded by sediment deposit movements which occur during stochastic events (Reistma et al., 2002). Similarly, probabilistic designs suffer from dilution of sampling effort at locations of specific interest but are better at compensating for shifts in spatial locations of major sediment deposits.

Tracking temporal trends of contaminated sediments is rarely addressed in sampling guidelines. For instance, recommendations included in the sediment guidance to the Water Framework Directive (EU, 2010) focus mostly on trend analysis of time series data obtained in routine monitoring programs. Therefore, multiple published studies present diverse approaches for assessing temporal changes, from collating data from different monitoring studies (i.e. weight of evidence approaches; e.g. Zwolsman et al., 1996; Marvin et al., 2004; Timoney and Lee, 2011) to periodic sampling of pre-determined (judgmental) sites (e.g. Bojakowska and Gliwicz, 2008; Choi et al., 2011).

The Detroit River (North America) provides a good case study for examining spatial/temporal trends of contaminated sediments representative of a large, complex connecting channel. Being part of one of the largest freshwater systems (connecting channel within the Laurentian Great Lakes) gives the study a supra-local perspective. As an international heritage river and International Joint Commission designated Great Lakes Area of Concern, understanding sediment contamination in the Detroit River has high importance from the point of view of bilateral, Canadian–American, relations. Finally, substantial effort has been put into sediment remediation actions including dredging of contaminated sites, dredging of shipping channels and source removal through improvement of sewage handling during the past 20 years (Zarull et al., 2001). Yet, post-monitoring clean-up activities were typically localized to the site of clean-up action and rarely placed remediation into the context of river improvements as a whole (Hartig et al., 2009). It is therefore important to be able to assess temporal changes of the system-wide contamination in this river. Although the Detroit River can boast one of the longest records of environmental quality monitoring in the Great Lakes, simple questions as to the state of the river and whether the river is improving or not could not be answered owing to changes in sampling designs and assessment techniques across surveys conducted in different years (Hartig et al., 2009).

The main goal of this study was to draw a conclusion about the decadal changes for the Detroit River, and to achieve this goal a set of river-wide sediment surveys were performed in 1999 and 2008/09. A stratified random sampling design was implemented during both surveys. Six strata encompassing the entire river were developed in order to track broad spatial and temporal patterns of sediment contamination along the river. Randomized sampling enabled use a variety of statistical tools to accomplish secondary objectives of this study i.e. to establish river-wide chemical mass balance estimates of priority contaminants, compare changes in contamination on a strata by strata basis, and to delineate locations of contaminated and clean zones within the river.

## 2. Study area

The Detroit River is a 51 km connecting channel linking Lakes Huron and Erie via Lake St. Clair. Most of the water flow in the river drains from Lake St. Clair, and in less than 21 h is discharged into Lake Erie. The average flow of the river is  $5240 \text{ m}^3 \text{ s}^{-1}$ , and five right-bank tributaries (USA, Michigan) and three left-bank tributaries (Ontario, Canada) account for <5% of flow. The long-term average flow displays a seasonal variation from  $4400 \text{ m}^3 \text{ s}^{-1}$  in winter to  $5700 \text{ m}^3 \text{ s}^{-1}$  in summer (USACE, 2001; DRCCC, 2009). The upper half of the Detroit River has steep banks, a width of less

than 1 km, depths reaching 15 m, and two islands in its head, Belle Isle and Peche Island (Fig. 1). The lower part has gently sloping banks, a width of 6 km at the mouth and an average depth of 10 m. Reaching Lake Erie, the Detroit River averages 3 m depth, except for the dredged navigation channels which are maintained between 10 and 15 m (Herdendorf, 1993).

The Detroit River was designated an Area of Concern (AOC) under the Great Lakes Water Quality Agreement in 1987 (IJC, 1987) and has been identified as the largest source of contamination to Lake Erie (e.g. Fallon and Horvath, 1985; Carter and Hites, 1992). Sediment contamination in the Detroit River has been linked to the degradation of benthos and exceedance of water quality standards (e.g. Hudson and Ciborowski, 1996a,b). Indirectly, contaminated sediments provide an exposure vector to biota that result in restrictions on fish and wildlife consumption, degradation of fish and wildlife populations and fish tumors and other deformities (Metcalf et al., 2000). The extent and severity of sediment contamination was a main reason for cleanup efforts completed under Remedial Action Programs for the Detroit River (Zarull et al., 2001; Hartig et al., 2009) including the upper and lower US reaches of the river.

## 3. Materials and methods

### 3.1. Sample collection

Two sediment surveys of the Detroit River were completed in 1999 and in 2008/09. The primary study (1999) involved 150 sampling sites while this number had been decreased down to 65 in the following survey (2008/09) due to the budgetary and logistic restrictions. Sampling sites were selected to encompass the entire boundary of the river using a stratified random design which involved sampling sediments in six strata of the river (Fig. 1). The initial strata selections were based on large-scale features that included international boundary, hydraulic considerations and point source locations suspected as being different in each reach (Drouillard et al., 2006). Strata consisted of upstream, midstream and downstream reaches each divided by width into US and Canadian waters. Samples were randomly assigned within each reach in proportion to the surface area. The sampling strategy deemphasized dredged shipping channels since these areas are less susceptible to sediment accumulation, and emphasized near shore zones (<5.5 m depth).

Surface sediment sampling was performed with a petite Ponar sampler (150 × 150 mm) until 2 L of sediment was collected at each site.

After collection, samples were mixed, sieved (2 mm sieve) and split for grain size, organic matter, trace metals, and organic contaminant analyses. Sediments were stored in acid washed plastic jars (for trace metal and mercury analysis) or hexane rinsed glass jars (organic contaminant analysis) at  $-20 \text{ }^\circ\text{C}$  until chemical analyses.

### 3.2. Laboratory analyses

Grain size distribution was performed by dry sieving (dried overnight at  $110 \text{ }^\circ\text{C}$ ) in an automatic sieve shaker (CSC Scientific, USA) with a series of graded sieves (>8, 8–2, 2–1, 1–0.5, 0.5–0.25, 0.25–0.150, 0.150–0.075, and <0.075 mm).

Organic matter content was determined using loss on ignition (LOI) technique, involving combustion of preweighed dried samples at  $450 \text{ }^\circ\text{C}$  for 24 h. This LOI technique has been demonstrated previously to be in a good agreement with TOM analyser (Carlo Erba Elemental Analyser; Carlo Erba) results ( $r=0.82$ ,  $n=147$ ; Drouillard et al., 2006).

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