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Mercury mobilization in urban stormwater runoff

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ABSTRACT

Urban stormwater runoff has been identified as a leading cause of waterway impairment for many pollutants, but there has been a lack of research that directly measures Hg in urban stormwater runoff. The objectives of this research were: to use high frequency sampling to characterize the Hg export dynamics from an urban micro-catchment (i.e. a parking lot) during individual rain events; determine the relationship between suspended sediments and Hg transport; assemble event-scale mass balances of atmospheric Hg inputs, surface storage, and Hg export in runoff to evaluate the relative importance of rainfall-derived Hg and surface-derived Hg in runoff; and finally, to compare the yield of Hg from the urban micro-catchment to that of a larger mixed land-use urban catchment to evaluate the feasibility of scaling the results. The results found that the highest Hg concentrations in runoff were observed during the rising limb of the hydrograph (first flush effect), which was dominated by particulate bound Hg (84±8%). There was a significant relationship between the Hg and total suspended solids (TSS) concentrations in runoff. For all events, the largest Hg flux occurred during the period of peak discharge, even though the Hg concentrations were substantially lower during this period. The catchment surface Hg load (i.e. street dust) varied over the course of the study, but the changes were not clearly linked to the rain events. The mass balance of the Hg inputs and outputs from the catchment showed that it could act as a Hg sink or a source depending on the rainfall characteristics. The export of Hg from the larger mixed land-cover catchment were all within the range of the values from the parking-lot catchment, though tended to be lower as a result of increased retention and fewer sources/disturbance within the catchment.

1. Introduction

Mercury (Hg) is a pollutant of concern due to its high toxicity at low concentrations (Ratcliffe et al., 1996). The main route of human exposure to Hg is through the consumption of contaminated fish (Myers et al., 2000). Several studies have found that rivers flowing through urban areas have higher Hg concentrations compared to rural areas (Hurley et al., 1995; Mason and Sullivan, 1998; Lawson et al., 2001). The elevated Hg concentrations found in these studies occurred during stormflow periods (baseflow Hg concentrations were similar or lower to non-urban rivers) and were predominantly a result of increases in Hg associated with suspended particles (HgP).

The elevated HgP in urban rivers could originate from internal sources such as bank erosion or bed re-suspension and/or from external sources, which could include inputs from rain and/or mobilization of surface Hg. Surface-derived Hg could have manifold origins including dry atmospheric deposition, near surface automobile inputs, anthropogenic and/or plant litter, and abrasion of surface materials. The relative importance of internal versus external sources of Hg to rivers will vary depending on the surrounding land-use,

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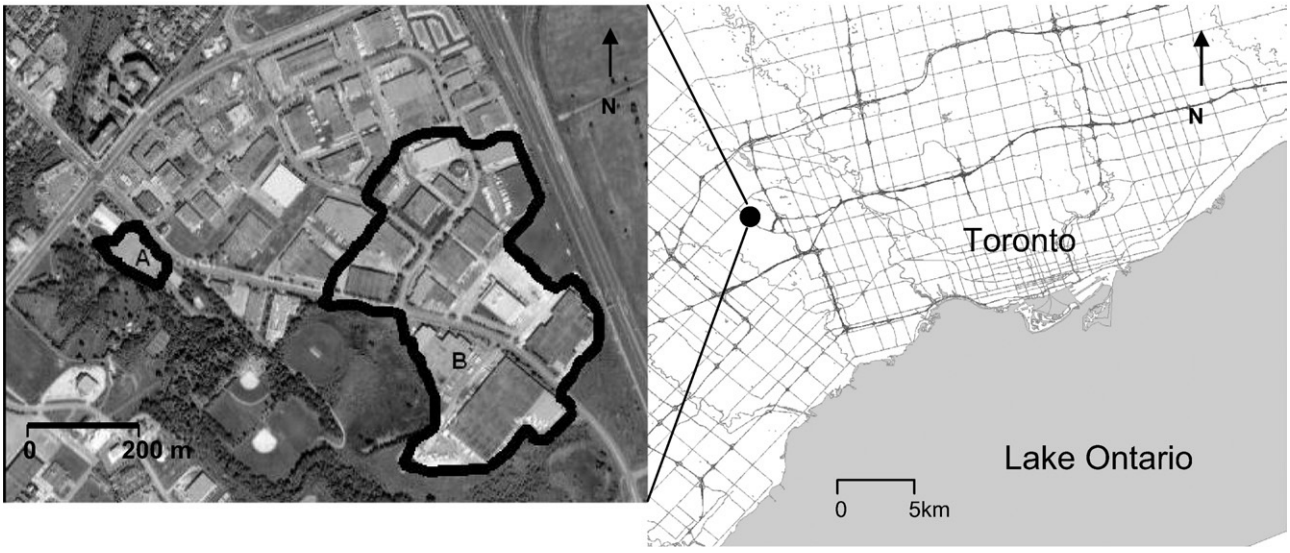

Fig. 1 – Map showing study location. Arial photo of the two catchments studied (A: parking-lot; B: mixed land-cover).

activity, and historic contamination. Identifying the source of Hg contamination in rivers is imperative to developing effective management strategies that could reduce concentrations. If external sources of HgP are significant, then traditional stormwater detention ponds may be effective at reducing the input load to rivers (though may have the adverse effect of generating methylmercury — Rumbold and Fink, 2006).

Quantifying the input of Hg into urban waterways is important because urban stormwater runoff has become a regulatory priority because it has been identified as a leading cause of waterway impairment (US EPA, 2002). Hg contamination of urban waterways can impact urban fisheries, which are often used for recreational and subsistence purposes (Kraft, 2000; Murkin et al., 2003). Elevated blood Hg concentrations have been found in anglers who frequently consume fish from urban areas around Lake Ontario (Cole et al., 2004). The mobilization of Hg stored in watersheds can be the dominant source of Hg to adjoining aquatic systems (Harris et al., 2007). Research comparing Hg in stormwater runoff from Precambrian Shield bedrock and a forested catchment showed that the relatively impervious bedrock surface transported much higher amounts of Hg in runoff (Allan et al., 2001). Urban areas are dominated by impervious surfaces and may behave similarly with higher Hg runoff loads compared to rural areas.

Numerous studies have been performed on the transport of other heavy metals, typically those from vehicular sources (cadmium, copper, lead, and zinc) in urban stormwater runoff (Charlesworth and Lees, 1999; Sansalone and Glenn, 2000; Yuan et al., 2001). While the sources, physical and chemical properties of Hg are distinct; the results from these studies can inform research on Hg in urban runoff. The cycling of street dust, partitioning between dissolved and particulate phases, and rainfall characteristics have all been shown to influence metal transport in urban runoff (Davis and Burns, 1999; Yuan et al., 2001). Metal concentrations have also been found to be significantly higher at the beginning of a hydrograph, referred to as the first flush effect and change rapidly throughout the hydrograph (Sansalone and Glenn, 2000; Shinya et al., 2000). This underscores the necessity to use high-resolution sampling techniques that can capture the entire hydrograph, with particular attention to the beginning of the event.

Based on the study of Hg in watercourses influenced by urbanization and evidence of solute dynamics from other studies of urban hydrochemistry, we hypothesize that runoff from urban catchments dominated by impervious surfaces will convey larger Hg loads than natural/undisturbed terrain, and the majority of this load will be in the form of surface-derived particulate Hg delivered early in the 'first flush' of the runoff event.

Given the minimal information on Hg fluxes from urbanized catchments, the confounding effects of mixed land-use, and the potential for extremely high temporal variability in mass transport, the objectives of this research were to: 1) Use high frequency sampling to characterize the water and Hg export dynamics from a homogeneous urban micro-catchment during individual rain events under a range of conditions; 2) Determine the relationship between suspended sediments and Hg transport; 3) Assemble event-scale mass balances of atmospheric Hg inputs, surface Hg associated with street dust, and Hg export in runoff for the micro-catchment in order to evaluate the relative importance of rainfall-derived Hg and surface-derived Hg in urban runoff; and 4) Compare the yield of Hg from the urban micro-catchment to that of a much larger mixed land-use urban sub-catchment to evaluate the feasibility of scaling the results of the small scale study.

2. Methods

2.1. *Site description*

The sample site chosen for this study was a catchment in the greater Toronto metropolitan area in Ontario Canada (population: 5,600,000 people) that drains into Mimico Creek, which flows directly into Lake Ontario approximately 15 km