



Application of fingerprint-based multivariate statistical analyses in source characterization and tracking of contaminated sediment migration in surface water



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ABSTRACT

This study investigates the suitability of multivariate techniques, including principal component analysis and discriminant function analysis, for analysing polycyclic aromatic hydrocarbon and heavy metal-contaminated aquatic sediment data. We show that multivariate “fingerprint” analysis of relative abundances of contaminants can characterize a contamination source and distinguish contaminated sediments of interest from background contamination. Thereafter, analysis of the unstandardized concentrations among samples contaminated from the same source can identify migration pathways within a study area that is hydraulically complex and has a long contamination history, without reliance on complex hydrodynamic data and modelling techniques. Together, these methods provide an effective tool for drinking water source monitoring and protection.

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1. Introduction

The co-existence of polycyclic aromatic hydrocarbons (PAHs) with heavy metals in industrial sites has been widely reported due to their common sources such as smelting, motor vehicle emissions, and gasification wastes (Gocht et al., 2001; Wang et al., 2004; Morillo et al., 2008; Thavamani et al., 2011; Chen et al., 2013). The presence of heavy metals may inhibit PAH biodegradation by naturally-occurring microorganisms, therefore amplifying the potency of the contamination and increasing the difficulty of contamination mitigation (Wild et al., 1991; Wang et al., 2004).

PAHs in aquatic environments tend to bind to particles and ultimately deposit with sediments due to their low aqueous solubility and high hydrophobicity (Chiou et al., 1998). Sediments in rivers, lakes, and oceans have been widely reported as important sinks for PAHs (Yang et al., 2008; Orecchio et al., 2010). Similarly, when heavy metals are introduced into the aquatic environment, they can be bound to different compartments within sediments through physicochemical adsorption, clogging in amorphous materials, complexation with organic matter, and bioaccumulation in benthic organisms (Jain and Sharma, 2001; Singh et al., 2005). Therefore

heavy metals tend to be retained and enriched to different extents in various sediment fractions (Salomons and Stigliani, 1995; Peng et al., 2009) and sediments can behave either as carriers or sinks for heavy metals (Singh et al., 2005). As important environmental indicators and major sinks for heavy metals and PAHs in aquatic environments, sediments are widely used to monitor and assess their contamination level (Soares et al., 1999; Stout and Graan, 2010). The high capacity of aquatic sediments to accumulate and integrate the low concentrations of trace contaminants in water can help to identify upstream contamination sources and characterize trace contaminants even when their levels in water are extremely low (Soares et al., 1999).

When PAHs and heavy metals co-occur in aquatic sediments, the fate of both is not only dependent on their physicochemical characteristics and biological interactions, but also the interactions among the contaminants, e.g., inhibition of PAH biodegradation by heavy metals (Thavamani et al., 2011). However, the co-deposition and co-migration of PAHs and metals in aquatic sediments have rarely been studied (Gocht et al., 2001; Wang et al., 2004). Gocht et al. (2001) reported that both types of contaminants from a common source will be deposited syngenetically in floodplain sediments and exhibit generally similar historical trends over most of the vertical distribution profiles. Total PAHs and total metals from a common source were also found to be significantly correlated in river sediments (Wang et al., 2004).

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Multivariate statistical techniques have been widely used to apportion the contributions of contaminants derived from different sources and investigate the distribution pattern and association of contaminants in soil and aquatic sediment (Scrimshaw and Lester, 2001; Sofowote et al., 2008; Stout and Graan, 2010; Thavamani et al., 2012). The most popular techniques include principal component analysis (PCA), discriminant function analysis (DFA), positive matrix factorization (PMF), and cluster analysis. Most applications of these multivariate techniques are based on contaminant concentrations in aquatic sediments. However, McCready et al. (2000) found that the relative abundances of PAH compounds from a common source in harbour sediments were strikingly consistent among sites. This strongly suggests that impacts of post-depositional processes such as biodegradation, desorption, and volatilization on PAH composition are, for the most part, insignificant even though geochemical characteristics vary greatly among the sites and among the compounds (McCready et al., 2000). A similar phenomenon has been reported by Plumb (2004) for anthropogenic contaminants (e.g., dioxin-furan, PAHs) and geochemical parameters (e.g., alkalinity, chloride, magnesium, and sulphate). Therefore, the relative abundance of contaminants, as opposed to absolute concentrations, can be considered as a chemical signature specific to a point source or contaminant plume. This chemical signature/fingerprint can help to characterize the contaminated sediments and track their migration away from the original point source.

The major objectives of this study were to investigate the suitability of using fingerprint-based multivariate statistical techniques to analyse the contamination of aquatic sediments with PAHs and heavy metals and evaluate their potential impacts on a local

drinking water intake. Fingerprint- and concentration-based multivariate analyses were employed to characterize the contaminated sediments, investigate the distribution pattern of contaminants within the study area, and identify the migration pathway of the contaminated sediments. The work reported herein builds upon a previous investigation by three of the authors (Chen et al., 2013) that utilized a fingerprint-based approach, but without multivariate statistical techniques. Regarding contaminated sediment mixing and migration, the additional insights and interpretations provided by using the multivariate techniques are substantial.

2. Material and methods

2.1. Site description and data acquisition

2.1.1. Site history and description

Port Stanley harbour is located at the outlet of Kettle Creek to Lake Erie in southern Ontario, Canada (Fig. 1). Intake Protection Zone 1 (IPZ-1) is a 1 km radius around the drinking water intake for the Elgin Area Water Treatment Plant which provides water to the City of London, Ontario. IPZ-2 is a secondary protective zone that extends “upstream” from IPZ-1 and is delineated in Fig. 1 as a curved line in the lake that extends onto dry land. Port Stanley is frequently exposed to westerly and south-westerly winds and occasionally exposed to strong winds from the south through east (Stantec Consulting Ltd., 2009b). In general, the wave distributions are reported to be consistent with wind distributions.

An oil/coal gasification complex was situated at Port Stanley harbour adjacent to Kettle Creek between 1920 and 1950. Oil/coal tar (hereafter referred to as “oil tar”) wastes from the gasification processes were stored in disposal lagoons, which were eventually abandoned in 1987 (Griffiths and Smith, 2010). At that time, the Ontario Ministry of Environment (MOE) determined that the historical gasification complex lands, as well as Kettle Creek sediments, were contaminated with PAHs and heavy metals. By the mid-1990s, the site was remediated by Ontario MOE and the discharges of contaminants to Kettle Creek ceased. However, the oil tar contaminated

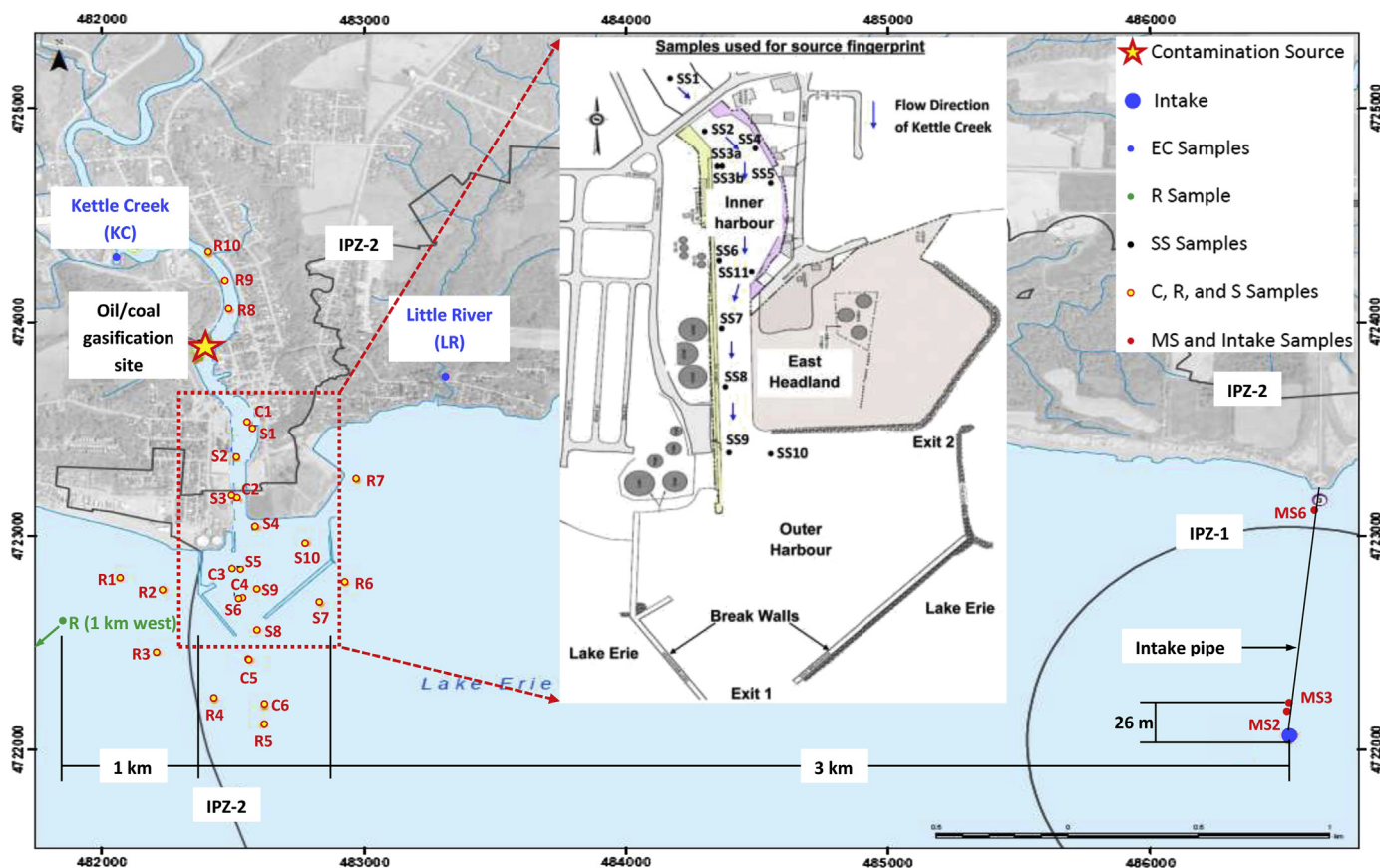


Fig. 1. Site layout and distribution of sediment sampling sites. Adapted from Stantec Consulting Ltd. (2009a) and CH2MHILL, 2009.

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