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Exposure of caged mussels to metals in a primary-treated municipal wastewater plume

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Abstract

The biological availability of metals in municipal wastewater effluents is strongly influenced by the physical and chemical conditions of both the effluent and the receiving water. Aquatic organisms are exposed to both dissolved and particulate (food ingestion) forms of these metals. In the present study, the distribution of metals in specific tissues was used to distinguish between exposure routes (i.e. dissolved vs. particulate phase) and to examine metal bioavailability in mussels exposed to municipal effluents. Caged *Elliptio complanata* mussels were deployed at sites located between 1.5 km upstream and 12 km downstream of a major effluent outfall in the St. Lawrence River. Metals in surface water samples were fractionated by filtration techniques to determine their dissolved, truly-dissolved (<10 kDa), total-particulate and acid-reactive-particulate forms. At the end of the exposure period (90 days), pooled mussel soft tissues (digestive gland, gills, gonad, foot and mantle) were analyzed for several metals. The results showed that gills and digestive gland were generally the most important target tissues for metal bioaccumulation, while gill/digestive gland metal ratios suggest that both exposure routes should be considered for mussels exposed to municipal effluents. We also found that Ag and Cd in the dispersion plume nearest the outfall, in contrast to other metals such as Cu and Zn, are more closely associated with colloids and were generally less bioavailable than at the reference site in the St. Lawrence River.

Keywords: Metal speciation; Bioavailability; Colloids; Effluent

1. Introduction

Urban effluents represent a major source of chemical contaminants for the aquatic environment (Chambers et al., 1997). Because effluents from municipal wastewater treatment plants are derived from both domestic and industrial sources, they contain a wide variety of natural and industrial products. Surveys in Canada have shown that metals and organic chemicals are present in municipal effluents (OMOE, 1988; Rutherford et al., 1994; Ministère de l'Environnement du Québec and Environment Canada, 2001) and a number of relatively small industrial plants release metals directly into municipal sewer systems (Ministère de l'Environnement du Québec and Environment Canada, 2001). Contaminants released in urban wastewater discharges may be taken

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up by the aquatic biota, leading to toxic effects (Rutherford et al., 1994; Wong et al., 1995; Gagné et al., 2002).

Predicting the environmental impact of the metals contained in municipal effluent discharges requires an understanding of how their physical and chemical characteristics, and those of the receiving waters, affect metal uptake by aquatic organisms and the associated toxicity. Although the toxicity of a municipal effluent is dependent on factors related to the treatment processes it undergoes, the physical and chemical characteristics of the receiving waters can also influence the toxicity of contaminants once they are released to the environment (Lijklema et al., 1993). These same properties also affect the speciation of both dissolved and particulate metals (Lijklema et al., 1993; Gagnon and Saulnier, 2003). The biological availability of metals is also influenced by their speciation in both the dissolved and particulate phases (Luoma, 1983; Campbell et al., 1988; Wang et al., 1997; Gagnon and Fisher, 1997b).

Physico-chemical parameters such as pH, organic carbon, suspended particulate matter (SPM) and temperature have an effect on the fate of metals and their biological uptake by both dietary and waterborne vectors (Luoma, 1983; Roditi et al., 2000). Metal partitioning occurs between the dissolved and particulate phases in effluent discharged to the aquatic environment, with geochemical processes playing a major role (Gagnon and Saulnier, 2003; Karvelas et al., 2003). The distribution of particulate metals is considerably influenced by organic matter as well as by reactive particulate iron (Fe) and manganese (Mn) (Gagnon and Saulnier, 2003). Various metal associations with both particulate and dissolved phases are determined by the concentration and strength of specific ligands. Thus, aquatic organisms are exposed to a variety of chemical forms of a given metal (Campbell et al., 1988; Luoma et al., 1992; Fisher and Reinfelder, 1995; Wang et al., 1997). Metal bioaccumulation, toxicity and exposure routes are probably all influenced by the concentration of each form of the metal. It has been suggested that metal concentrations in the gill and digestive gland can be used to estimate waterborne and dietary exposures, respectively (Fisher et al., 1996). Such differences in metal tissue distribution were documented for mussels exposed to waterborne and dietary metals. For example, Long and Wang (2005) recently reported that metal concentrations in mussel gills were higher than those in the remaining tissues after exposure to waterborne metals and reversed patterns after dietary exposure.

We related the metal forms at different distances within the dispersion plume with those found in the tissues of mussels caged in the plume. The objectives of the study were to: (1) measure the distribution of various metals in mussel tissues; (2) estimate waterborne versus dietary exposure by comparing uptake in the gill versus the digestive gland; (3) compare the relative uptake from water and food by using gill/digestive gland ratios for various sites above and below the outfall; and (4) compare metal accumulation in tissue with metals in operationally dissolved (<0.45 μm), truly-dissolved (<10 kDa), particulate and reactive-particulate phases. Metal accumulation was assessed in mussels placed in the dispersion plume of an effluent whose dilution by receiving water can influence the bioavailability and distribution of metals among target tissues. In parallel, changes in the physico-chemical form of metals were studied in the effluent plume by measuring the dissolved, truly-dissolved, total-particulate, and reactive-particulate fractions of metals. Moreover, the biomarkers of vitellogenin and metallothionein were used to confirm mussel exposure to the effluent and its associated estrogenicity and metal exposure, respectively.

2. Methods

2.1. Water sampling

The effluent dispersion plume was previously characterized by the determination of total and fecal coliforms or chemical contaminants as markers for municipal effluents (Gagné et al., 2001a; Gagnon and Saulnier, 2003) (Fig. 1). Surface water and suspended matter were sampled at several sites ranging from 0.5 to 15 km downstream of a major municipal outfall in the St. Lawrence River (Fig. 1). Samples were also collected at a reference station 1.5 km upstream of the effluent outfall, in order to document the initial river water conditions. Surface water samples were collected using a Teflon pump following clean sampling procedures for trace metals, as described in Cossa et al. (1996).

2.2. Mussel exposure experiment

Elliptio complanata mussels were collected during the first week of May 2001 at Lake de L'Achigan in the Laurentians, Quebec, Canada, where mussels have been collected for previous studies. The animals were placed in experimental cages according to standard protocols (ASTM, 2001). Briefly, mussels were sorted by size (weight and shell length) and placed in cylindrical nets, which were attached to a PVC frame (1 m^2 surface area). The frames were attached to 20 kg blocks and marked with a submerged buoy. Two cages were deployed at each of four sites: 1.5 km upstream (reference station) and 5, 8 and 12 km downstream of the municipal effluent outfall (Fig. 1). The cages could have not been deployed any closer to the outfall due to the high current and suspended matter loads in this stretch of the river. Caged mussels (30 per cage) were submerged in the St. Lawrence River for 90 days, from the beginning of June until the beginning of September 2001. Unfortunately,