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Biological effects of gold mine tailings on the intertidal marine environment in Nova Scotia, Canada

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1. Introduction

Nova Scotia experienced its first gold rush in the 1860s after the discovery of gold in a quartz vein at Mooseland in 1858 (Bates, 1987). This was followed by two additional gold rushes which accounted for the development of >60 gold districts throughout Nova Scotia from the 1860s to the mid 1900s. During this time, total gold production for Nova Scotia was estimated to be 1.2 MOZ. The fine grained wastes from the milling process (tailings), were contaminated by mercury and arsenic and these contaminated tailings were generally deposited without any form of control into natural water bodies or into low lying areas next to water bodies.

In Nova Scotia, mercury was used in gold mining for over 100 years, and the primary method used to extract the majority of gold during the three gold rushes was mercury amalgamation until the 1880s (Smith and Kontak, 1996), when it was supplemented by cyanidation (Bates, 1987). In general, 1 oz. of mercury was used for each ounce of gold recovered (Parsons et al., 2004). Of the total mercury used in the extraction process, 10–30% is typically lost per season (Alpers and Hunerlach, 2000) while another study found that up to 40% was lost

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ABSTRACT

From 1861 to the 1940s, gold was produced from 64 mining districts in Nova Scotia, where mercury amalgamation was the dominant method for the extraction of gold from ore until the 1880s. As a result, wastes (tailings) from the milling process were contaminated by mercury and were high in naturally occurring arsenic. In 2004 and 2005, sediments, water and mollusc tissues were collected from 29 sampling stations at nine former gold mining areas along the Atlantic coastline and were analysed for arsenic and mercury. The resulting data were compared with environmental quality guidelines. Samples indicated high potential risk of adverse effects in the intertidal environments of Seal Harbour, Wine Harbour and Harrigan Cove. Arsenic in Seal Harbour was bioavailable, resulting in high concentrations of arsenic in soft-shell clam tissues. Mercury concentrations in tissues were below guidelines. This paper presents results of the sampling programs and implications of these findings. Crown Copyright © 2016 Published by Elsevier Ltd. All rights reserved.

> when panning for gold (Ogola et al., 2002). The excess mercury was distributed with the tailings or lost through airborne emissions. Consequently, elevated mercury concentrations have been found in the environment associated with abandoned mine sites in Nova Scotia, although many of them have been closed for 70 years or more (Parsons et al., 2004). Eisler (2004) suggested that mercury has no beneficial biological function. Instead, its presence in living organisms causes neurotoxicity, birth defects and other adverse effects. Its ability to transform to more toxic forms (methyl mercury) and biomagnify within the food chain poses risk of adverse biological effects in ecosystems, especially to organisms in higher trophic levels (for example, fish eating mammals and birds), and in humans who consume mercury contaminated species.

> Arsenic is a naturally occurring element in many bedrock types. In Canada, arsenic occurs in gold ore deposit minerals such as arsenopyrite (FeAsS) (Eisler, 2004; Wang and Mulligan, 2006). Arsenopyrite is the most abundant arsenic containing mineral found and is commonly associated with gold (Francesconi and Kuehnelt, 2002). In Nova Scotia, historical mining activities resulted in the release of approximately 3 Mt of tailings containing 20,700 kg of arsenic (Wong et al., 1999). Since arsenic is found abundantly in the gold ore, when disturbed and altered, it can have serious repercussions on the environment and human health. The World Health Organization (WHO, 1992) reported that inorganic arsenic is a documented human carcinogen. The Agency for Toxic Substances and Disease Registry (2007) reports a variety of adverse effects of arsenic exposure in humans, including irritation of the stomach

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and intestines, blood vessel damage, skin changes, reduced nerve function, decreased production of red and white blood cells, skin cancer, and increased risk of cancer in the liver, bladder, and lungs. Environment Canada and Health Canada (1993) concluded that arsenic and its compounds are considered to be "toxic" to the environment and constitute a danger to human life or health. Gomez-Caminero et al. (2001) found that marine biota tend to accumulate much higher concentrations of arsenic than freshwater species. Neff (1997) suggested that the highest concentrations of arsenic are found in tissues of marine animals that feed primarily on phytoplankton or macroalgae. These species include planktonic crustaceans, bivalve molluscs, herbivorus snails, and some polychaete worms. Much of the bioaccumulated arsenic is present in a relatively nontoxic organic form, such as arsenobetaine (Cullen and Reimer, 1989; Francesconi and Kuehnelt, 2002; Phillips and Depledge, 1985).

Mercury and arsenic have a long residence time in the environment although local concentrations may decline due to dispersion processes. Releases from gold mining operations pose potential risks to the ecosystems and human health either through direct exposure to tailings, or indirectly from contaminated air, water or through the consumption of arsenic or mercury contaminated organisms. Mollusc species can provide valuable insight on the availability of metal(oid)s to the food web. Dixon and Wilson (2000) suggest that mussels are a good choice for the detection of pollution because they accumulate a variety of chemical substances due to their filtration capacity, their contact with sediments and the water column, and because of their low mobility. The use of bivalves to assess the bioaccumulation of metal(oid)s has been used in many studies around the world. The National Oceanic and Atmospheric Administration's National Status and Trends Program for Marine Environmental Quality has been using mussels and oysters since 1986 to monitor spatial and temporal trends in contamination of the coastal USA (Valetee-Silver et al., 1999). The San Francisco Estuary Institute, after a five year period (1993–1997) of analyzing heavy metals in mussels, oysters, and clams, concluded that bivalves are effective tools for monitoring long-term trends and that they provide valuable information that water or sediment data alone would not supply (Hardin et al., 1999).

In 2003, Natural Resources Canada initiated a study to examine the dispersion, speciation and fate of mercury and arsenic in terrestrial and shallow marine environments surrounding 14 abandoned gold mines in Nova Scotia (Parsons et al., 2012). The field studies confirmed that most of those sites contain large volumes of unconfined tailings and that gold mine tailings throughout Nova Scotia contain elevated concentrations of mercury and arsenic, which were widely dispersed in local sediments and surface waters. Based on the Natural Resources Canada findings, Environment Canada undertook studies in 2004 and 2005 to measure the magnitude and distribution of arsenic and mercury in sediments, water, and bivalve molluscs and this paper presents the results of these studies.

2. Methods and materials

2.1. 2004 study

The 2004 study focused on six intertidal sampling stations in Seal Harbour, Nova Scotia, located in the immediate vicinity of the mouth of West Brook, the main source of transport for tailings that were originally deposited approximately 1 km upstream (Fig. 1). At each sampling station, location was determined on a hand-held Global Positioning System device and is presented in Table 1. Sediment and soft-shell clam (*M. arenaria*) samples were taken from each sampling station. All samples were collected on 9 August 2004. Intertidal sediment was sampled from a depth of 0–10 cm using disposable polystyrene spatulas. Soft-shell clams were depurated in a bucket full of water from the site for approximately 10 h, after which they were measured, weighed, and the meat was removed from the shell with stainless

steel disposable scalpels. All sediment and tissue samples were then frozen for transport to laboratories for analyses of arsenic and mercury. Clams sold for human consumption were purchased from a Moncton, NB, fish market and used as reference to compare with clams collected from Seal Harbour.

2.2. 2005 study

The field program was expanded in 2005. In order to prioritize mining areas that could be having an impact on the marine environment, each of the 64 gold mining districts in Nova Scotia was assessed against the following criteria:

- Sites which had a relatively high ore throughput (>10,000 t);
- Sites located within 5 km of a marine receiving environment;
- Sites which have the potential for tailings to enter a receiving environment and that are "classified shellfish growing areas" where shellfish might be harvested for human consumption;
- Site accessibility;
- Sites that have any "species at risk" that potentially might be present;
- Shoreline should have intertidal sediments (i.e. favourable for clams, one of the indicator species to be collected during this study).

Nine former gold mining areas met the above criteria and were sampled in 2005, along with a reference area at New Harbour, for a total of 23 sampling stations. Their locations are shown on Fig. 1. The reference area was chosen at New Harbour, Nova Scotia, since it was from the same rock formation where the gold vein or lode deposits are predominantly located (the Meguma Supergroup slates and greywackes, Bates, 1987). No gold mining had been carried out in the New Harbour watershed area and so there was no influence from mine tailings. At Seal Harbour, because of the contamination identified in the 2004 study, a total of eight intertidal stations were chosen to examine the distribution of arsenic and mercury throughout the intertidal areas of the Harbour (Table 1), from 2.54 km west to 1.6 km east of the mouth of West Brook. Sampling occurred at low tide. At each sampling station, location was determined on a hand-held Global Positioning System device and is presented in Table 1. 100 mL of intertidal sediment was collected near the location of the clams from a depth of 0-10 cm using a new disposable polystyrene spatula. Water (250 mL) was collected adjacent to the blue mussels into an acid-washed plastic bottle. Five or more clams, and ten mussels (both >5 cm in length), were collected. Softshelled clams, and mussels were depurated in a food grade polyethylene bucket full of water from the station for approximately 6 h. Water samples and sediment samples were kept cool. At the end of the day, clams and mussels were measured, weighed, and the measurements recorded. The tissues were removed from the shell using new stainless steel disposable scalpels for each species at each station. The meat of all soft-shelled clams were pooled for each sampling station, and the meat of all blue mussels were pooled for each sampling station, and placed in acid-washed glass jars. Water samples were preserved using a few drops of nitric acid. All samples but water were then frozen for transport to laboratories for analyses. At each sampling station, samples were taken for: total arsenic and mercury in unfiltered seawater; total arsenic and mercury in sediments; total arsenic and mercury in tissues of soft-shell clam (M. arenaria) and blue mussel (Mytilus edulis); and inorganic arsenic speciation as arsenic (III) and arsenic (V) in tissues of clams and mussels. As a quality assurance measure and to ensure that data were comparable between the laboratories, homogenized and split samples of clam tissue (29% of samples, n = 4) and sediment (30% of samples, n = 7) were sent in 2005 to both the Environment Canada laboratory in Moncton, NB and to the laboratories used in 2005 for the majority of analyses for comparative purposes. The samples were thoroughly homogenized so that a representative sample could be used for analysis by each laboratory. The tissue samples were

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