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Toolset for assessment of natural recovery from legacy contaminated sediment: Case study of Pallanza Bay, Lake Maggiore, Italy

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ABSTRACT

The aim of this study was to develop a toolset that can be used by site managers to assess and monitor natural attenuation processes in sediments contaminated with legacy hydrophobic organic contaminants. The toolset is composed of sediment traps to measure quality and deposition rate of incoming sediment under different hydrodynamic conditions, sediment cores to show trends in sediment bed concentrations over time, and passive samplers attached to a porewater probe frame to assess the mobility of buried contaminants and possible contaminant flux from sediment. These three tools were used together for the first time to assess the mobility of dichlorodiphenyltrichloroethane (DDT) contaminants in sediment in Pallanza Bay, Lake Maggiore, Italy. Depositing sediment and sediment cores were consistent in showing that DDT-contaminated sediment is undergoing burial by cleaner sediment. Elevated DDT concentrations from historical contamination seemed to be effectively buried and immobilized by ongoing deposition by cleaner sediment, because the positive flux from the elevated DDT concentration in the sediment porewater should not advance towards the sediment surface. The monitoring toolset introduced in this study enabled us to more effectively assess ongoing natural attenuation processes and provide more risk relevant data than traditional methods used in monitored natural recovery projects, such as bulk sediment concentrations from sediment cores. Our field assessment results suggest that incoming sediment from the Toce River have reduced DDT concentrations in the sediment compared to historic levels, and will continue to do so in locations where higher DDT concentrations are found within the bioactive layer.

1. Introduction

Monitored natural recovery (MNR) of historically contaminated sediments can be a viable and cost-effective remediation strategy provided one can scrupulously measure and document site-specific processes that destroy or strongly immobilize contaminants within a reasonable timeframe (U.S. EPA-SAB, 2001; Magar and Wenning, 2006). Because contaminants are left in place, MNR requires multiple lines of evidence to support an MNR management decision, and to show that relevant biological, physical, and chemical processes are effectively reducing contaminant risks. While chemical or biological processes can transform and remove contaminants, these processes can be slow compared to immobilization processes that occur through sediment deposition and contaminant burial. Currently, the toolbox for assessing burial and deposition as well as sediment sorption rely heavily on sampling of sediment cores and subsequent analysis ex situ (Magar and Wenning, 2006). Unfortunately, this approach lacks the benefits of in situ monitoring, which represents local, site specific conditions. Moreover, freely dissolved contaminant concentrations measured using passive sampling techniques are a better predictor of toxicity and bioavailability than bulk sediment concentrations, and therefore crucial to making better-informed sediment management decisions than traditional methods (Lydy et al., 2014; Witt et al., 2013; Greenberg et al., 2014). Our current study is, to the best of our knowledge, the first study to combine traditional methods with in situ porewater passive sampling techniques to address the processes of natural recovery. The application of in situ techniques fills an important knowledge gap in field studies and strengthens the current toolbox by applying

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three different in situ measurement tools to provide multiple lines of evidence to quantify, assess, and document MNR.

1.1. Field site

Lake Maggiore is an alpine lake formed by glacial scour. The lake is characterized by steep side slopes, with depths in Pallanza Bay reaching a plateau of approximately 120 m depth, and then descending rapidly where the Bay connects to the rest of the Lake to depths of nearly 400 m deep. DDT contamination in the Lake was first reported in 1996 (CIPAIS, 1999; Bettinetti et al., 2006), and was traced to a DDT manufacturing facility located 30 km upstream of Pallanza Bay on the Toce River (Fig. 1). The plant was dismantled in 1997, and since then, studies indicate that DDT contamination had spread throughout the lake (Riva et al., 2010), and studies have measured lipid total DDT and DDT metabolite concentrations in zooplankton, mussels, and fish in the ppm range (Bettinetti et al., 2010). At the time of our field investigations, there was no consensus as to whether the DDT concentrations in the Lake represented an ecological or human health risk. In this manuscript, we will refer to DDT as a generic representative compound for DDT and DDT metabolites.

Hydrodynamics in Pallanza Bay are driven by river inflows, winds, seasonal heating and cooling, and rotation of the Earth. River inflow from the Toce River is driven by snowmelt in the spring and by storms in the fall, as well as the Prata Dam upstream of Pieve Vergonte and reservoirs (Sea Engineering, Inc., 2009; Scheu et al., 2015). Stratification is affected by summer heating and winter cooling, although the lake remains stratified year round and overturns rarely (Ambrosetti and Barbanti, 1999).

Recent investigations of sediment transport in the Toce River and Pallanza Bay show that sediments in Pallanza Bay are predominantly silts, with smaller fractions of fine sands and clays, and studies also show sediment bed DDT concentrations in Pallanza Bay have been decreasing since the 1980s (Sea Engineering, Inc., 2009; Marritt et al., 2011; Bizzotto and Colombo, 2014). Reconstruction of sediment core deposits through hydrodynamic modeling of sediment transport supports the understanding that the Toce River flood plains and Pallanza Bay are mainly depositional with minimal resuspension (Sea Engineering, Inc., 2009; Scheu et al., 2015). Sea Engineering, Inc. (2009) concluded that deposition rates in the Bay are highly variable, and range from low mm/year to low cm/year, with rates of tens of cm/year near the river mouth. Bay-wide average deposition rates also vary by an order of magnitude from year to year based on flow rates and events from low mm/y to almost 10 cm/y during years with extreme flood events (Sea Engineering, Inc., 2009).

Under these depositional processes, exposure to DDT in the water column and sediment to the biota are expected to decrease through the process of natural capping of contaminated sediment with cleaner sediments. To our knowledge, previous studies have not measured the total DDT concentrations in incoming sediment to Pallanza Bay. Therefore, it is difficult to predict whether the reductions in sediment DDT concentrations observed in the past (Sea Engineering, Inc., 2009; Marritt et al., 2011) will continue in the future. Furthermore, it is unclear how porewater concentrations will change due to ongoing deposition processes and how this will affect the contaminant flux from the sediment bed.

1.2. Field objectives

The main goal of this field study was to develop a toolset to provide multiple lines of evidence to evaluate natural recovery processes. And another goal was to use the toolset to evaluate whether incoming sediment in the Bay was clean and whether sediment deposition processes could be expected to continue reducing the bioavailability of DDT in the Bay, as measured by the freely dissolved concentration of DDT (Lydy et al., 2014) in the sediment porewater and the overlying water column in Pallanza Bay. The approach was to determine how effectively elevated freely dissolved concentrations in older sediments were isolated from the sediment surface.

In order to determine how well new sediments are isolating the older contaminated sediments, we measured the DDT concentration in the sediment bed and newly settling sediment, sediment deposition rate under different hydrodynamic conditions, the freely dissolved concentration profile in sediment and overlying water, and calculated the flux profile in the sediment and the overlying water.

2. Material and methods

2.1. Field sampling plan

Field studies were conducted in 2012 (Phase I, Sep.–Nov.) and 2014 (Phase II, Apr.–Jul. and Phase III, Jul.–Nov.), which are outlined in Table A.1. Location of sampling sites in Pallanza Bay are shown in Fig. 1 and equipment deployed at each site is shown in Table A.2. Phase I sampling sites were chosen to compare areas where sediment cores show faster rates of natural attenuation (Sites P1 and P3) with an areas where sediment cores showed slower rates of natural attenuation (Site P2). Sampling equipment was deployed in triplicate at each site during Phase I, and located within an area of 200 m^2, which was the smallest size possible to avoid interference between mooring ropes. One of the main goals of the Phase II study was to determine DDT concentrations in sediment deposited during the major spring flood event that occurs annually between May and June when snow melt from the Alps drives high water flow rates down the Toce River. Equipment was deployed from April to July, and was subsequently redeployed from July to November for the Phase III study in order to capture the autumn flood events. The three sampling phases were used to compare total DDT concentrations in depositing sediments and deposition rates from seasonal events (Fig. 2). Total DDT is defined here as ΣDDT = sum of 4,4′-DDT, 2,4′-DDT, 4,4′-DDE (dichlorodiphenyldichloroethylene), 2,4′-DDE, 4,4′-DDD (dichlorodiphenyldichloroethane), 2,4′-DDD, and DDMU (dichlorodiphenylmonochloroethylene).

2.2. Sediment traps

Sediment traps (Fig. A.8) were used to collect depositing sediment from the water column during the deployment periods to measure apparent sediment deposition rates and ΣDDT concentrations in the depositing sediment. During Phase I, triplicate sediment traps were deployed at 3 m above the sediment at Sites P1, P2, and P3. During Phase II and III, two sediment traps were placed at each location at Sites T4–T10 at 3 and 7, or 10 m above the sediment during both deployment periods (Table A.2). The thickness of the deposited sediment layer in the traps was recorded upon retrieval. Afterwards, supernatant in the tubes was gently siphoned out, and the remaining sediment was transferred to clean plastic buckets where the retained sediment was allowed to settle. After additional settling for a few days, supernatant was again siphoned out of the buckets, and final sediment samples were poured into 250-mL precleaned amber glass jars (I-CHEM series™). De-ionized (DI) water was used to rinse the buckets to recover sediment from sediment traps and settling buckets. Samples were dried and the total dry sediment mass weighed.

A hydrodynamic study conducted in parallel with Phase I of this field work showed that short periods of high flow events accounted