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Uptake of organic xenobiotics by benthic invertebrates from sediment contaminated by the pulp and paper industry

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ABSTRACT

Uptake of pulp and paper mill-derived pollutants by benthic invertebrates from sediment in Southern Lake Saimaa, eastern Finland, was studied. Two groups of benthic invertebrates (Diptera and Oligochaeta) were analyzed for their concentrations of resin acids (RAs), chlorophenolics (CPs) and β-sitosterol. The samples were collected 1 and 3 km downstream from the mill. In laboratory experiments *Chironomus plumosus* (a dipteran) and *Lumbriculus variegatus* (oligochaete) were exposed for 14 d to sediments collected from the same locations. The concentrations of RAs, CPs and β-sitosterol were higher in the areas downstream from the mill than those in the upstream reference area in both the feral and laboratory-exposed animals. Examination of the possible conjugation of contaminants revealed hydrolyzable fractions of RAs in Diptera, *C. plumosus* and *L. variegatus*. The results indicate both the bioavailability uptake of contaminants and uptake by benthic fauna when exposed to pulp and paper mill-contaminated sediment.

1. Introduction

Sediments serve as a sink for many industrial contaminants (Paasivirta et al., 1985; Kukkonen et al., 1996; Leppänen and Oikari, 1999; Lahdelma and Oikari, 2006); however, they can also act as a source for bioactive chemicals, thus posing a long-term hazard to the environment. Resin acids (RAs), wood sterols (WSs) and chlorophenolics (CPs) are all wood industry-derived contaminants, and are found in effluents, receiving waters, sediments and fishes living close to the pulp and paper industry (Kukkonen et al., 1996; Leppänen and Oikari, 1999; Lahdelma and Oikari, 2005). Because of the sorption–desorption cycle of many organic contaminants, not only is bioavailability in the short term reduced due to sorption to sediment organic material but also long-lasting exposure of benthic biota is evident (Paasivirta et al., 1985; Karels et al., 2001; Hyötyläinen and Oikari, 2004).

Benthic invertebrates are exposed to xenobiotic chemicals through several routes. One is direct integumental contact with sediment, its particles and pore water, and another is through ingestion of contaminated sediment (Landrum, 1989; Leppänen and Kukkonen, 2000). Pulp and paper mill effluents and sediments contain possibly bioavailable (Meriläinen et al., 2006) and toxic amounts of RAs, WSs (including β-sitosterol) and CPs (Kukkonen et al., 1996; Lahdelma and Oikari, 2005). Therefore, it is possible that benthic invertebrates are exposed to these sediment-borne contaminants, which are lipophilic, likely to absorb into organic particles of the sediment, and which have been deposited in high concentrations over several decades. Moreover, sediment

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organic carbon measures such as total organic carbon (TOC) are important factors affecting the bioavailability of lipophilic contaminants (Nikkilä et al., 2001). Biota-to-sediment accumulation factors (BSAFs) are widely used to describe the potential accumulation of organic contaminants in organisms (Spacie et al., 1995).

The invertebrates chosen for this study are important components of the benthic ecosystem, and they vary in their living environments, feeding habits and ability to metabolize xenobiotics. The sediment-dwelling oligochaete *L. variegatus* is a widely used organism in bioaccumulation studies, not least because of its ecological relevance (Chapman, 2001). Somewhat similarly, chironomids, a group of Diptera, are closely associated with the sediment surface, burrowing in the upper ca. 0–2 cm layer and feeding on particulate matter. Thus, chironomids, although in close contact with both the sediment and the overlying water, may be in contact with pore water. In addition, by bioturbation, larvae of *Chironomus riparius* can increase the bioavailability of contaminants (Goedkoop and Peterson, 2003), increasing the concentration of contaminants in the overlying water. Regarding the metabolic characteristics of invertebrates, *L. variegatus* is assumed to lack certain biotransformation capabilities (Verrengia Guerrero et al., 2002; Hyötyläinen and Oikari, 2004), whereas *C. plumosus*, as a member of Diptera, has a more developed metabolic system than that of *L. variegatus*.

The metabolism of pulp industry-derived contaminants has been studied widely in fish, but not to a significant extent in benthic invertebrates (Oikari et al., 1984; Leppänen and Kukkonen, 2000; Verrengia Guerrero et al., 2002). Moreover, there is very little information on the possible bioaccumulation of bleached kraft mill effluent (BKME)-related bioactive contaminants in benthic fauna. Such knowledge would be important to understand the transfer of xenobiotics in the food chain. Biotransformation consists of two phases: phase I, in which lipophilic xenobiotics are transformed into primary metabolites, and phase II, which continues the biotransformation by conjugation processes, turning primary metabolites into highly hydrophilic metabolites, as these are easier to eliminate than the original compound (Livingstone, 1998). This study presents observations on the uptake and possible conjugation of RAs, β-sitosterol and CPs in benthic freshwater invertebrates. The information gathered helps in assessing whether BKME-contaminated sediments pose a long-term ecotoxicological risk to lake ecosystems.

The objectives of this study were (1) to measure the uptake of pulp and paper industry-derived contaminants in sorted invertebrate groups collected from an area that had been receiving BKME for decades; (2) to assess the presence of hydrophilic metabolites derived from these contaminants in animal tissue; and (3) to validate the uptake of RAs and β-sitosterol by benthic invertebrate species subjected to laboratory exposure.

2. Materials and methods

2.1. *Sampling of benthic fauna from a polluted lake area*

Benthic animals were collected in October 2002 from Southern Lake Saimaa in two areas, located 1 and 3 km downstream from a pulp and paper mill (Fig. 1). The spatial distribution of contaminants in the sediment is well known in Southern Lake Saimaa (e.g. Lahdelma and Oikari, 2005), and therefore only three sampling sites were decided to be used in this study. Information on the hydrography of the area is provided in Fig. 1, and in more detail by Lahdelma and Oikari (2005). In 2001, the mill produced 164 000 tons of mechanical pulp and 678 000 tons of soft- and hardwood-based kraft pulp, as

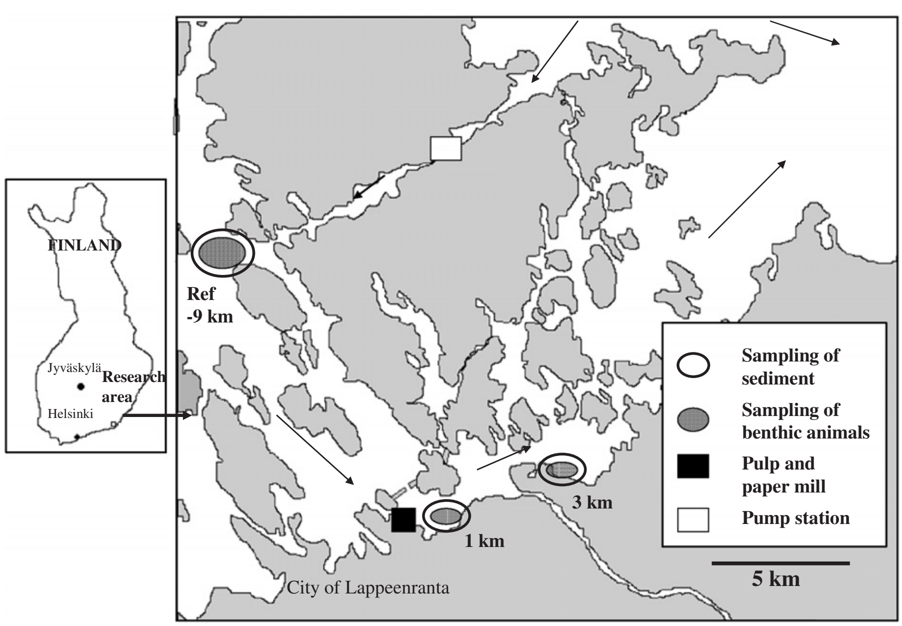

Fig. 1 – The sampling areas of sediments and benthic animals in Southern Lake Saimaa, Finland. Area Ref −9 km is the reference area; area 1 and 3 km are located downstream from the pulp and paper mill. The black arrows indicate the water flow in the area.