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Effects of bioturbation on the fate of oil in coastal sandy sediments – An *in situ* experiment

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ABSTRACT

Effects of bioturbation by the common lugworm *Arenicola marina* on the fate of oil hydrocarbons (alkanes and PAHs) were studied *in situ* during a simulated oil spill in a shallow coastal area of Roskilde fjord, Denmark. The fate of selected oil compounds was monitored during 120 d using GC–MS and bioturbation activity (feces production and irrigation) was measured regularly during the experiment and used as input parameters in a mechanistic model describing the effects of *A. marina* on the transport and degradation of oil compounds in the sediment. The chemical analytical data and model results indicated that *A. marina* had profound and predictable effects on the distribution, degradation and preservation of oil and that the net effect depended on the initial distribution of oil. In sediment with an oil contaminated subsurface-layer *A. marina* stimulated both the physical removal and microbial degradation of oil compounds in uniformly oil contaminated sediments especially in deeper sediment layers (10–20 cm below the surface), whereas the fate of oil compounds deposited in surface layers (0–5 cm) mainly was affected by removal processes induced by wave actions and other bioturbating infauna such as *Nereis diversicolor*, *Corophium volutator* and *Hydrobia* spp. present in the experimental plots.

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1. Introduction

Bioturbation by benthic infauna is known to influence physical and geochemical sediment properties and play important roles for sediment processes such as nutrient cycling and degradation of organic matter (Berner, 1980; Aller and Aller, 1998; Banta et al., 1999; Kristensen, 2001). Particle mixing and the irrigation of burrows with overlying water may enhance the transport of particles, water and solutes between different redox zones (Aller and Aller, 1998) and change the structure and porosity of the sediment (Kure and Forbes, 1997). The enhanced transport of oxygen and other electron acceptors into otherwise reduced environments may affect the rate at which mineralization occurs (Banta et al., 1999; Papaspyrou et al., 2006), the degradation pathway (Kristensen et al., 1985; Nielsen et al., 2004) and the composition of the microbial communities (Reise, 1981; Papaspyrou et al., 2006).

One class of materials that is affected by bioturbation is organic pollutants such as crude oil and oil constituents (Kure and Forbes, 1997; Christensen et al., 2002b). Given their high hydrophobicity

* Corresponding author. Address: National Environmental Research Institute, Aarhus University, Frederiksborgvej 399, P.O. Box 358, 4000 Roskilde, Denmark. Tel.: +45 46301296. many organic pollutants become associated with particles and accumulate in the organic fraction of the sediments where they interact with the benthic community. Whether organic pollutants in fact remain at the sediment surface, are buried, are released to the water column, or are degraded is highly depending on benthic macroinfauna activity (Gilbert et al., 1994; Schaffner et al., 1997).

Although most of our knowledge on the effect of bioturbation on the fate of contaminants comes from controlled laboratory experiments, data from *in situ* situations are required in order to understand and quantify the relative importance of bioturbation under natural conditions where weathering processes such as dissolution, evaporation and photo degradation are known to play a more important role (Wang and Fingas, 1995). The very few semi-controlled *in situ* experiments performed with bioturbated contaminated sediment do however demonstrate that bioturbation also may affect the fate of contaminants under natural conditions (Gilbert et al., 1996; Nobbs et al., 1997; Grossi et al., 2002).

Interactions between infauna and pollutants are often complicated making it difficult to predict the fate of contaminants in bioturbated sediments, especially under natural conditions and with natural infauna communities. Effects of bioturbation from multi species infauna communities demonstrate, however, that the effect of bioturbation is often controlled by one or a few "keystone" species (Levin et al., 1997; Sandnes et al., 2000; Mermillod-Blondin





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et al., 2005). Hence, focusing on the role of these keystone species is a good first step in understanding and predicting the effects of infaunal communities on sediment pollutants.

A good example of such a dominant, keystone bioturbator is the lugworm, Arenicola marina, common in shallow, sandy areas along the Atlantic coastline, which has been shown to dramatically alter the biological, chemical and physical characteristics of the sediments it inhabits (Volkenborn et al., 2007). A. marina is a head down conveyor belt feeder, ingesting large quantities of sediment 10-20 cm below the sediment surface and defecating at the surface (Riisgård and Banta, 1998). This feeding behaviour results in a vertical transport of particles downward in the sediment. Irrigation by A. marina results in a water transport in the opposite direction. Water is pumped down the tail shaft and injected directly into the sediment at feeding depth resulting in an upward advective transport of water through the feeding funnel. Laboratory experiments (Kure and Forbes, 1997: Rasmussen et al., 1998: Christensen et al., 2002b; Banta and Andersen, 2003) have demonstrated the significant effects of A. marina on the fate of organic and inorganic contaminants but the importance of A. marina bioturbation for the fate of contaminants under natural conditions has however not been investigated and the objectives of the present study were to examine the in situ effects of A. marina on the fate of selected oil compounds from a simulated oil spill.

We used a combination of field experiments and mechanistic modelling to test the potential impact of *A. marina* on the fate of crude oil. The model describes the effects of *A. marina* bioturbation on transport and degradation processes and has previously been validated in controlled laboratory experiments (Timmermann et al., 2003). The model is extrapolated to field conditions in the present study to test if it can predict the *in situ* fate of oil considering *A. marina* bioturbation alone given that we expect physical factors and other organisms, factors not included in the model, to play important roles in the field.

2. Materials and methods

2.1. Experimental site

The experiment was conducted in a shallow and sheltered bay of Roskilde fjord. This location has large populations of *A. marina, Nereis diversicolor, Corophium volutator* and *Hydrobia* spp. The water depth varies between 0 and 1 m. Tide has limited influence in this enclosed fjord system, and water depth is primarily controlled by wind action. The sediment is poor in organic matter (~1%, determined by loss on ignition) and can be classified as permeable sandy sediment. Vegetation is patchy, and was absent at the experimental site. Mean salinity was 14‰, but varied between 10‰ and 16‰.

2.2. Sediment contamination

A mixture of four North Sea crude oils obtained from the National Environmental Research Institute, Denmark was artificially weathered by air bubbling for 48 h resulting in a mass loss of 27% due to evaporation. Sediment from the experimental site was sieved through a 1 mm mesh and mixed with the weathered crude oil for 24 h. A sediment portion with an oil concentration of approximately 2000 mg kg⁻¹ was prepared for later use in subsurface-layer plots. Another portion of sediment was prepared with an oil concentration of 500 mg kg⁻¹ for later use in plots with uniform oil concentration.

2.3. Experimental plots

A total of four experimental plots were constructed when the sediment surface was exposed (low tide and off shore wind) by removing sediment from an area with a diameter of 37 cm and a depth of 40 cm using a polycarbonate cylinder. The resulting holes were lined with a special sewn thin plastic lining. Two plots designated subsurface-layer plots were filled to approximately 4 cm from the top with uncontaminated and sieved (1 mm) sediment from the same location. Oil contaminated sediment (2000 mg kg⁻¹) was carefully distributed in a 2 cm layer on top of the uncontaminated sediment. To avoid immediate removal of the contaminated layer by wind and wave action an additional layer (approx. 2 cm) of uncontaminated sediment was added. Two plots with a uniform oil concentration (designated uniform plots) were prepared by filling the plastic lined holes with contaminated sediment (500 mg kg^{-1}) until the sediment within the plots levelled with the surroundings. Parts of the plastic lining still sticking up, was then cut of just below the surface to avoid hydrodynamic disturbances due to "wall effects". Hereafter the presence of the plots was only hardly visible. The artificial weathering as well as initial distribution of oil in these experimental plots was chosen to simulate two common scenarios in oil spills, namely a recent settlement of the heavy fraction of an oil spill on and near the sediment surface (subsurface) and an older, chronic situation where the oil contamination has been present for some time and is prevalent throughout the system (uniform).

Lugworms (6 in.) collected at the location were added to each of two plots (one uniform plot and one subsurface-layer plot) resulting in a density of 56 worms m^{-2} which is in the upper end of the natural density range (Riisgård and Banta, 1998).

2.4. Experimental procedure

2.4.1. Maintaining of plots

During the experimental period sediment plots were checked and maintained weekly. The number of lugworms present in each plot was estimated as the number of fecal casts. New worms were added if <6 casts were observed due to the assumption that nondefecating worms were either dead or had escaped the plots. The number of fecal casts in plots with *A. marina* never exceeded 6 and fecal casts were never observed in *A. marina* free plots indicating that *A. marina* did not invade the experimental plots.

2.4.2. Sediment reworking

Feces produced by the lugworms was measured by collecting fecal casts deposited at the sediment surface, 4–6 h after the surface had been carefully levelled to avoid collection of previously deposited fecal casts. Volume of the fecal casts was measured and returned to the surface to avoid artificial removal of sediment and oil. Feces production was measured six times during the experimental period however during one occasion strong wind and wave action moved the produced feces preventing measurement of the fecal material.

2.4.3. Solute transport

Irrigation (water transport) was measured using bromide as a passive solute tracer. Prior to sediment sub-sampling the water column above each plot was isolated by placing one end of a plastic cylinder designed for sediment flux measurements into the sediment around the plot and with the other end above the water surface. Bromide ($[Br^-] = 10 \text{ mM}$) was added to the isolated water column and incubated during continuously stirring with battery driven air pumps for 4–7 h.

2.4.4. Sediment sampling

One sediment sub-sample was taken from each plot at day 1, 20, 32, 60 and 120 using a plastic core with a diameter of 5 cm and a length of 40 cm. To avoid collapse of the sediment surrounding the sub-sampling location, removed sediment was replaced

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