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In Situ Bioremediation Alternatives for Sediments Contaminated with Hexabromocyclododecane

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Background/Objectives. Hexabromocyclododecane (HBCDD) is an additive brominated flame retardant (BFR), which is widely used in extruded, expanded and high impact polystyrene foams for thermal insulation in buildings, and secondarily used in upholstery furniture, automobile interior textiles, car cushions and electrical equipment. Being produced since 1960s, HBCDD became the most used cycloaliphatic BFR. The consumption rate of HBCDD reached 12500 tons annually in 2013 in Europe. Due to the high usage rate, high HBCDD levels were found in soils, sediment and sewage sludge, which were mostly near to the regions of high industrialization and urbanization, and also found in human adipose tissue and blood. Toxicity tests of HBCDD have shown disruption in the thyroid hormone system, developmental neurotoxic effects and alteration in uptake of neurotransmitters in the brain. Owing to these properties of the chemical, HBCDD was listed among the persistent organic pollutants (POPs) by the Stockholm Convention in 2014, and the countries involved in the Convention started to phase out its use. Recent research has revealed high contamination levels in the environment; however, there is limited research focusing on HBCDD transformation. Biodegradation of HBCDD under aerobic and anaerobic conditions in soils, sediments, and sewage sludge were investigated in a few studies. The aim of this study is to investigate the fate of HBCDD in aquatic sediments under various conditions, thereby to assess effectiveness of possible in situ remediation strategies: natural attenuation, bioaugmentation, and biostimulation.

Approach/Activities. Natural attenuation was established with natural sediments spiked with HBCDD, with no amendments. Biostimulation, additional to the components of the natural attenuation set included a carbon source and electron donor rich organic medium. Bioaugmentation was prepared with *Dehalobium chlorocoercia* strain DF-1 additional to the components of the natural attenuation set. Control sets were prepared with i) culture media without bacteria cells, ii) autoclaved and mercury poisoned sterile sediments, iii) abiotic solid phase (i.e., kaolinite) instead of sediments, and iv) contaminant control, i.e., without HBCDD spike. Sediments used in microcosms were collected from a pond with no previous HBCDD contamination. Sacrificial operation of microcosms was adopted, hence for each set and each sampling time, duplicate 20 mL reactors were established. They were incubated in the dark at 25°C. Sediments were ultrasonically extracted and extracts were analyzed in gas chromatography – mass selective detector (GC-MS) as gamma-HBCDD.

Results/Lessons Learned. Biostimulation of sediments with a carbon source and electron donor rich organic medium demonstrated 86.3% reduction in HBCDD concentration in 36 days. The overall HBCDD decrease in natural attenuation and bioaugmentation sets were 36.9% and 34.9%, respectively. Biostimulation showed the highest rate of degradation with a first-order rate constant of 0.0542 per day. Natural attenuation and biostimulation sets, however, demonstrated rates of 0.0155 and 0.0123 per day, respectively. The rates found in this study were generally lower than previously reported values, except for one study which agrees well with the natural attenuation set conditions. Additionally, statistical analysis yielded no significant difference between natural attenuation and bioaugmentation sets, indicating the limited capability of DF-1 strain in degradation of a cycloaliphatic compound. Abiotic control and sterile microcosms, on the other hand, showed statistically no significant change in time. Hence, it can be assumed that HBCDD decreases observed in test reactors were only due to biodegradation, without the complicating effect of abiotic mechanisms. Application of three bioremediation techniques concurrently revealed a differentiation in terms of their biodegradation efficiency for HBCDD contaminated sediments.