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Concentrations of Antifouling Biocides in Sediment and Mussel Samples Collected from Otsuchi Bay, Japan

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Abstract. Organotin compounds (OTs) and representative booster biocides were measured in sediment and mussels from Otsuchi Bay, Japan. The mean amounts of tributyltin (TBT) and triphenyltin (TPT) compounds in sediment were 13 μg kg^{-1} dry and 3 μg kg^{-1} dry, respectively. Representative booster biocides (Sea-Nine 211, Diuron, Dichlofluanid, Irgarol 1501, M1, which is a degradation compound of Irgarol 1051, and Copper pyrithione) were also detected in sediment from Otsuchi Bay. OT concentrations were higher than those of the measured booster biocides. Otsuchi Bay was divided into four parts by cluster analysis based on OT concentrations in sediment sampled from the bay. These areas included the vicinity of a shipyard, a small fishing port, the closed inner area of the bay, and the mouth of the bay. Higher concentrations of TBT and TPT and a higher ratio of TBT to total BTs were observed in the vicinity of the shipyard. A higher concentration of TPT in comparison with TBT was detected in a small fishing port. Furthermore, OT concentrations in the mouth of the bay were higher than those in the closed-off section. OT concentrations in mussels decreased with distance from the shipyard. Otsuchi Bay was then divided into three parts by cluster analysis based on the concentrations of representative booster biocides found in the bay's sediment. These areas included the vicinity of a shipyard, a small fishing port, and other sites. Concentrations of Diuron and Irgarol 1051 in the vicinity of a shipyard and a small fishing port were dramatically high in comparison with the other sites. Copper pyrithione and Dichlofluanid in addition to Diuron and Irgarol 1051 were also detected in the area of a small fishing port. The concentrations of antifouling biocides were highest in the water in front of the shipyard and showed a marked decrease with distance from the shipyard.

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Organotin (OT) compound leaching from antifouling paints has caused many deleterious effects on nontarget aquatic organisms, including imposex and abnormal shell (Gibbs *et al.* 1988; Waldock and Thain 1983). As a result, OTs have led to a decrease in aquatic products (Bryan *et al.* 1986), and thus OT application to large vessels has been banned or restricted in some countries. In Japan, in 1990, bis(tributyltin)oxide (TBTO) was designated a Class 1 Specified Chemical Substance under the Law Concerning the Examination and Regulation of the Manufacture of Chemical Substances, and 7 triphenyltin (TPT) species and 13 tributyltin (TBT) species, excluding TBTO, were designated as Class 2 Specified Chemical Substances under the same law. Actually, in the past the sale and use of OTs was regulated. In spite of the regulations, OT compounds have nonetheless been detected at higher concentrations in water, sediment, and biota from harbours, marinas, and estuaries, particularly where boat activity is high and water movement is restricted (e.g., Harino *et al.* 1998; Tselentis *et al.* 1999). In October 2001, the International Maritime Organization (IMO) adopted the International Convention on the Control of Harmful Antifouling Systems (AFS Convention), which prohibited the use of OTs as active ingredients in antifouling systems for ships. Following the international restrictions on the use of OT-based antifoulants, paint manufacturers have developed many products as alternatives to the use of OTs. In Japan, more than 20 chemical substances have been used or proposed as alternative compounds. When these antifouling biocides from the hulls of ships, fishing nets, and so on, are released into the aquatic environment, these chemicals are distributed among water, sediment, and aquatic organisms. In fact, aquatic pollution by these booster biocides has been reported already in some countries. Sea-Nine 211, Diuron, and Irgarol 1051, which were representative booster biocides, were detected at the levels of sub ng L^{-1} and sub μg kg^{-1} dry in water and sediment from coastal areas, respectively (Harino 2004).

The Sanriku coastal area is one of the areas of Japan that is rich in marine products. In particular, Otsuchi Bay, located in