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Antifouling herbicides in the coastal waters of western Japan

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Abstract

Residue analyses of some antifouling herbicides (Diuron, Irgarol 1051 and the latter's degradation product M1, which is also known as GS26575), were conducted in waters collected along the coast of western Japan. In total, 142 water samples were collected from fishery harbours (99 sites), marinas (27 sites), and small ports (16 sites) around the Seto Inland Sea, the Kii Peninsula, and Lake Biwa, in August 1999. A urea-based herbicide, Diuron, was positively identified for the first time in Japanese aquatic environments. Diuron was detected in 121 samples (86%) up to a highest concentration of 3.05 μg/l, and was found in 86% of samples from fishery harbours, 89% from marinas, and 75% from ports. Four freshwater samples out of 11 collected at Lake Biwa contained Diuron. Neither Irgarol 1051 nor M1 was found in the lake waters, but both were found in many coastal waters. Irgarol 1051 was found in 84 samples (60%) at a highest concentration of 0.262 μg/l. The concentrations detected were of similar magnitude to those in our previous surveys, taken in 1997 and 1998. M1 was found in 40 samples (28%) up to a highest concentration of 0.080 μg/l. The concentrations detected were generally lower than those found in our previous surveys. The detection frequency among fishery harbours, marinas, and ports was 57–70% for Irgarol 1051 and 25–30% for M1. Ninety-five per cent of the coastal waters in which M1 was detected also contained Irgarol 1051, and 93% of the samples in which Irgarol 1051 was detected also contained Diuron. These results clearly suggest that commercial ship-bottom paints containing both Diuron and Irgarol 1051 are used extensively in the survey area.

Keywords: Antifouling; Coastal waters; Degradation; Diuron; Herbicide; Irgarol 1051

1. Introduction

Organotin antifouling compounds have been regulated internationally since the late 1980s and early 1990s. Alternative biocides are intended as replacements for organotins. Voulvoulis et al. (1999) reviewed 11 alternative antifouling biocides and concluded that there was not enough information on such chemicals to perform a sound environmental risk assessment. Recently, the environmental fate and behaviour of antifouling paint booster biocides such as chlorothalonil, dichlofluanid, Diuron, Irgarol 1051, Sea Nine211, TCMTB, zinc pyrithione, and Zineb, were reviewed by Thomas (2001). Among the compounds that are currently used as antifouling biocides, Irgarol 1051 (2-methylthio-4-*tert*-butylamino-6-cyclopropylamino-*s*-triazine) has been detected in the estuarine, coastal, and lake waters and sediments in Europe since 1992 (Readman et al., 1993; Gough et al., 1994; Toth et al., 1996; Scarlett et al., 1997; Penalver et al., 1999; Thomas et al., 2000). There are a few reports on Irgarol residues outside Europe, for example, in the Australian Great Barrier Reef (Scarlett et al., 1999), in the Seto Inland Sea of Japan (Liu et al., 1999), and in Hamilton Harbour, Bermuda (Connelly et al., 2001). Irgarol 1051 and its degradation product M1 (2-methylthio-4-*tert*-butylamino-6-amino-*s*-triazine) were found in Japanese coastal waters with highest concentrations of 296 and 1870 ng/l, respectively (Okamura et al., 2000). Solar degradation of Irgarol in the upper water column has been proposed as a possible mechanism for the formation of M1 (Okamura et al., 1999).

Recently, 17 new antifouling compounds, including Irgarol 1051 and Diuron (3-(3,4-dichlorophenyl)-1, 1-dimethylurea), were summarized in a report published

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