



Concentration and degradation of alternative biocides and an insecticide in surface waters and their major sinks in a semi-enclosed sea, Japan



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HIGHLIGHTS

- Surface water analysis of alternative biocides and an insecticide was done.
- The pesticides were photodegraded and biodegraded.
- Data generated and literature information were used in a mass distribution model.
- Pesticide degradation in river water was significantly faster than that of seawater.
- Sediments were found to be the major sink for the pesticides.

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ABSTRACT

A mass distribution model was used to predict the fate of Diuron, Irgarol 1051 and Fenitrothion in Seto Inland Sea which is located in western Japan. This was done by using concentration, degradation, and literature data. Diuron and Irgarol 1051 in Seto Inland Sea are mainly derived from antifouling paints used for ships and boats. On the other hand Fenitrothion exclusively comes from land via rivers and atmospheric deposition. The total inputs/yr to Seto Inland Sea were found to be 104 tons, 7.65 tons and 5.14 tons for Diuron, Irgarol 1051 and Fenitrothion, respectively. The pesticide residence times were 0.26 yr, 0.36 yr and 0.17 yr for Diuron, Irgarol 1051 and Fenitrothion, respectively. Photodegradation was faster than biodegradation. In seawater, the half-life ranges were 37.9–57.3 d for photodegradation. In the same seawater the half-life ranges were 1650–2394 d for biodegradation. Photodegradation is effective in surface water (0–5 m depth) while biodegradation occurs throughout the entire water column. Plankton and fishes accumulate these pesticides significantly. The pesticides are deposited (sorbed and buried with) sediments (between 74 and 87% of total input amounts). The open ocean is an important sink accounting for between 8 and 17% of the total pesticide input amounts while photo- and biodegradation accounts for a small percentage.

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1. Introduction

The widespread usage of pesticides has led to their amounts increasing significantly in water bodies making them major pollutants of the aquatic environment (Aaron and Oturan, 2001). Pesticides include Diuron (3-[3,4-dichlorophenyl]-1,1-dimethylurea), Irgarol 1051 (2-[tert-butylamino]-4-[cyclopropylamino]-6-[methylthio]-1,3,5-triazine) and Fenitrothion

(O,O-Dimethyl O-(3-methyl-4-nitrophenyl) phosphorothioate). These pesticides have various uses. Diuron and Irgarol 1051 are used as alternative biocides in antifouling paint (Ferrer et al., 1997; Lamoree et al., 2002). Diuron is also used as an herbicide (Blanchoud et al., 2004) while Irgarol 1051 is also used in the protection of structures as it is added to roof sealants (Mascarelli, 2009). Fenitrothion is used as an insecticide (Tsuda et al., 1998). Due to their high usage rates, these pesticides have been reported in the environment. For example in Japan Diuron, Irgarol 1051 and Fenitrothion have been reported in rivers (Numabe et al., 1992; Tanabe et al., 2001; Sakai, 2007; Kitada et al., 2008) and also the Seto Inland Sea (Okamura et al., 2003; Harino et al., 2005;

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Balakrishnan et al., 2012; Kaonga et al., 2015a). Fenitrothion is very volatile such that it has also been reported in rainwater in Japan (Suzuki, 1996; Nohara et al., 1997; Sakai, 2006).

In the environment, these pesticides are removed through degradation (Cardeal et al., 2011). Two major degradation pathways are photo transformation by sunlight and biodegradation (Tixier et al., 2001). The breakdown of a pesticide by an organism (biodegradation) is a primary strategy for its own survival (Singh, 2008) because microorganisms use it as a source of carbon and nitrogen (Cullington and Walker, 1999; Barragan-Huerta et al., 2007). Biodegradation has been reported in a number of studies. For example the bacteria *Bacillus cereus* and *Burkholderia ambifaria* are able to degrade Diuron (Ngigi et al., 2011): White rot fungus, *Phanerochaete chrysosporium* is able to degrade Irgarol 1051 (Ogawa et al., 2004): Fenitrothion was found to be degraded by the bacteria *Flavobacterium* sp. and *Pseudomonas putida* (Uobe, 2005). Photodegradation which involves production of OH radicals is one of the major transformation processes for pesticides. In natural waters •OH is generated by a number of mechanisms for example photolysis of nitrite and nitrate (Mopper and Zhou, 1990) and its concentration is dependent on water composition as well as on the spatial and temporal variations in solar irradiance (Zepp et al., 1992). While hydrolysis also destroys organic compounds, it has been found to be insignificant at pH common to natural waters (Lacorte and Barcelo, 1994).

The pesticides that are not degraded end up into aquatic organisms like plankton and fishes or other environmental segments. Plankton forms the basis of food chains in seawater. According to Fenchel (1988), organic production in the sea depends on phytoplankton. These are consumed by zooplanktons which are food for small fish which end up as prey for larger fish. Through these feeding linkages, organisms accumulate chemicals (Leppanen, 1995) such as pesticides. Organisms higher up in the food chain like fish accumulate more pesticides (Senthilkumar et al., 2001) than those on lower trophic levels. The fate of the fish that have accumulated pesticides includes the following: being caught through fishing; and being eaten by marine animals. Through excretion, death and then decomposition these organisms release pollutants to sediments and seawater. It is therefore important to determine the fate of chemicals in the environment.

According to Mackay et al. (2001), chemical fate can be predicted using models providing sufficient information is available on certain key chemical and environmental properties. This is because certain attributes of chemicals can be measured directly notably concentrations. However models become important when estimating chemical fate because in most cases this cannot be measured directly.

In this study, an assessment of alternative biocides (Diuron and Irgarol 1051) and an insecticide (Fenitrothion) in Kurose River water and marine samples (water, sediment, plankton and fishes) from the Seto Inland Sea, Japan was done. Degradation of these pesticides in river water and seawater samples was also done in the laboratory. Data obtained in laboratory analysis and literature information were used in a mass distribution model to predict the fate of these pesticides in Seto Inland Sea. The mass distribution model used in this study is a modification of box models which are useful tools for chemical risk assessment and management. Box models or Fugacity models were first introduced by Baughman and Lassiter (1978) in which the fate of chemicals in different environmental media is calculated based on physical chemical properties of a compound, environmental characteristics and emission data. This study is important because research combining biodegradation and photodegradation is rare. Additionally information on the total environmental quantities and fate of pesticides is important in managing their negative impacts.

2. Materials and methods

2.1. Sampling

Fig. 1a shows a map of the sampling area. The Seto Inland Sea is a semi-enclosed coastal sea located in the western part of Japan (Yoshie et al., 2011). Marine samples (water, sediments, plankton and fishes) were collected during four sea cruises in 2012–2014 using the Hiroshima University Research Ship – Toyoshio maru, which is equipped with CTD Sensors that measure salinity and photon intensity data used in this study. Kurose River (located in Higashi Hiroshima City, Japan) water samples were collected during 2013–2014.

2.2. Reagents

Diuron was obtained from Kanto Chemical Co. Inc. (Tokyo, Japan), Irgarol 1051 from Sigma Aldrich Laborchemikalien (Seelze, Germany) and Fenitrothion from Dr. Ehrenstorfer GmbH (Augsburg, Germany). HPLC grade acetonitrile (99.8%) and methanol (99.7%) were obtained from Nacalai Tesque Inc. (Kyoto, Japan).

2.3. River water and marine samples preparation for analysis

Sample preparation and analysis was done as described in Kaonga et al. (2015a, 2015b) which also have details of sampling stations, sample collection and identification of the pesticides. Briefly, samples were extracted using organic solvents and cleaned by Solid Phase Extraction (SPE). Analysis was done by an HPLC system described further below.

2.4. Biodegradation experiments

All glassware used were sterilized at 180 °C for 2 h using a Yamato Drying Sterilizer SH401. Unfiltered water samples (250 mL) were transferred to 300 mL conical flasks (three replications in each case) covered with plug silicone caps. These samples were spiked with Diuron, Irgarol 1051 and Fenitrothion to a final concentration of 0.5 mg L⁻¹ according to Derbalah et al. (2004). The samples were put in a Biotron incubator at 21 °C for three months according to Sakugawa et al. (2010). Samples were taken every ten days for HPLC analysis.

2.5. Photodegradation experiments

An Oriel solar simulator (Model 81160-1000) unit equipped with a 300 W Xenon lamp (Model 81160) and special glass filters restricting the transmission of wavelengths below 300 nm was used. Irradiation of samples was carried out in a quartz glass cell (60 mL) with the solution well mixed by magnetic stirring and temperature was kept at 20 °C. Irradiation was done in triplicate for each pesticide. The solution for the irradiated samples was removed at regular intervals for HPLC analysis.

2.6. Sample analysis

Pesticides analysis was done by a reverse phase HPLC system (Shimadzu Corporation, Japan) equipped with a UV–Vis detector using 60:40 acetonitrile and milli-q water as eluent with isocratic elution at a flow rate of 1 ml/min. A Supelcosil LC-18 (Supelco, particle size 5 µm) 250 mm × 4.6 mm ID column was used.

Additionally: Nitrate and nitrite in river water samples were analysed by Ion Chromatography (Dionex ICS 1600) while in seawater by Spectrophotometer (Shimadzu UV-1200). pH was measured by a Horiba D-24 pH/Conductivity meter. DOC was determined by Total Organic Carbon Analyser (TOC-VCSH,

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