

# Occurrence and Distribution of Antifouling Biocide Diuron along the Coastal Areas of Zanzibar Island

Hassan R. Ali<sup>1,2\*</sup>, Fatma Kh. Ali<sup>3</sup>, Miza A. Kombo<sup>1</sup>

<sup>1</sup>School of Natural and Social Sciences, The State University of Zanzibar, Zanzibar, Tanzania

<sup>2</sup>Tropical Research Centre for Oceanography, Environment and Natural Resources, The State University of Zanzibar, Zanzibar, Tanzania

<sup>3</sup>Karume Institute of Science and Technology, Mbweni, Zanzibar, Tanzania

Email: \*hassan.ali@suza.ac.tz, \*hassan7418@gmail.com

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## Abstract

Diuron (N-(3,4-dichlorophenyl)-N,N-dimethylurea) is one among the booster biocides substituted tributyltin as an antifouling agent. It has continued to be used ever since, though little is known about their levels in the maritime setting of Zanzibar. This paper details the occurrence and distribution of diuron around the coastline of Zanzibar Island. The reported concentrations of diuron in water ranged from Below Detection Limit to  $1321.67 \pm 52.3$  ng/L at Bwawani. Majority of the sites showed levels above Environmental Risk Limit of 430 ng/L as proposed by the Dutch Authorities. The findings suggest the contamination is elevated especial in Harbor, Bwawani, Chapwani and even at Marine Protected Areas such Mnemba Island where the value of  $620.33 \pm 5.70$  ng/L was recorded. The main possible sources of the contamination are cargo shipping, passenger speedboats, fishing, and tourism activities taking places in these areas. Therefore, future studies on the antifouling biocide diuron particularly on long term monitoring, chronic exposure, risk assessment, organisms' responses and pollution models will add special value towards better understanding of the mechanisms and sustainable marine ecosystem health.

## Keywords

Diuron, Coral Reefs, Harbor, Marine Resources, Marine Protected Areas

## 1. Introduction

Diuron (N-(3,4-dichlorophenyl)-N,N-dimethyl-urea) is one among antifouling agents that was developed to replace more toxic biocide tributyltin. Apart from its

importance in the shipping industry, it has reported to pose some threat to aquatic ecosystem. Studies demonstrated the several effects of diuron on plants, corals and number of ecological effects as well as human being (Lewis et al., 2009; Jones & Kerswell, 2003; Jones, 2004; Jones, 2005). It has been classified as a hazardous substance by the European Commission (Malato et al., 2002; Malato et al., 2000a; Malato et al., 2000b; Malato et al., 2000c) as a result, some European countries, such as the United Kingdom, Sweden, Denmark, and France banned the use of diuron as an antifouling paints (Konstantinou & Albanis, 2004; Giacomazzi & Cochet, 2004).

Diuron was also used to control weeds on hard surfaces and crops, such as roads, railway tracks, pears, apple trees, forestry, ornamental trees and shrubs, pineapples, sugarcane, cotton, and wheat. For instance, sugarcane is one of the important food and commercial crops of Tanzania. Its production is concentrated mainly in three regions, Morogoro, Kagera, and Kilimanjaro for Tanzania mainland and Mahonda in Zanzibar Island. Furthermore, it is widely used as antifouling compound to inhibit photosynthesis in plants by binding site of photosystem II (PSII), which limits the electron transfer (Haynes et al., 2000). Diuron is very persistent in the environment, as it may remain from one month up to one year (Giacomazzi & Cochet, 2004). Diuron has been detected in marine environments from various regions such as western Japan (Okamura, 2002; Okamura et al., 2003; Sheikh et al., 2009), UK (Boxall et al., 2000), Spain (Ferrer & Barceló, 1999; Ferrer et al., 1997; Martinez & Barcelo, 2001; Martinez et al., 2000), The Netherlands (Lamoree et al., 2002), Sweden (Dahl & Blanck, 1996) and Malaysia (Ali et al., 2021; Ali et al., 2014). Diuron can undergo abiotic degradation such as hydrolysis, photo degradation as well as biotic degradation (Giacomazzi & Cochet, 2004). Occurrences of diuron in the marine environment require special attention in order to prevent further harm to marine life that is already stressed due to other environmental factors (Sheppard et al., 2002; Sathiyaseelan & Stella, 2011).

Zanzibar, with its extensive coastline, coral reefs, and new emphasis on coastal and marine-based tourism, has become a focal point for marine conservation efforts in Tanzania. Four marine protected areas were established in Zanzibar during the 1990s: Chumbe Island Coral Park, Mnemba Island Marine Conservation Area, Menai Bay Marine Conservation Area, and Misali Island Conservation Area (Levine, 2007). However, there exists a large gap between available information and desired information in most of the coastal as well as Marine Protected Areas (MPAs) of Zanzibar on contamination of biocide diuron. While there are plentiful of research works that has been carried out in many of the MPAs worldwide, the findings of these research works are seldom available locally. While there are already areas that have become seriously degraded by the direct or indirect effects of human activities and the rate of degradation is increasing rapidly (Agardy, 1997; UNEP-WCMC, 2023), it is important that the development of MPAs must give consideration for the continued welfare of people who have customarily used

these areas.

Due to the fact that the maritime environment is a global trade, and most developing countries have poor biocide monitoring strategies, diuron is still used in Tanzania including Zanzibar, while little attention has been paid to assess its environmental risk. By considering the fact that, Zanzibar depends on shipping activities and tourism as economic activities the chance of accumulation of antifouling agents such diuron is higher. The present study aimed to fill the knowledge gap by determines occurrence and distribution of diuron in seawater along the coasts of Zanzibar Island including MPAs such as Chumbe Coral Park and Mnemba Island Marine Conservation Area.

## 2. Material and Methods

### 2.1. Study Sites

Samples were collected from 11 different locations along Zanzibar Island (**Figure 1**). Sampling sites with a range of different characteristics were chosen to guarantee representativeness. The various socio-economic activities represented at the chosen sampling stations included ports, recreation, fishing, agriculture, tourism, shipyards, cargo shipping, and ferries. A summary of the activities for each sampling station is shown in **Table 1**.

### 2.2. Sample Collection

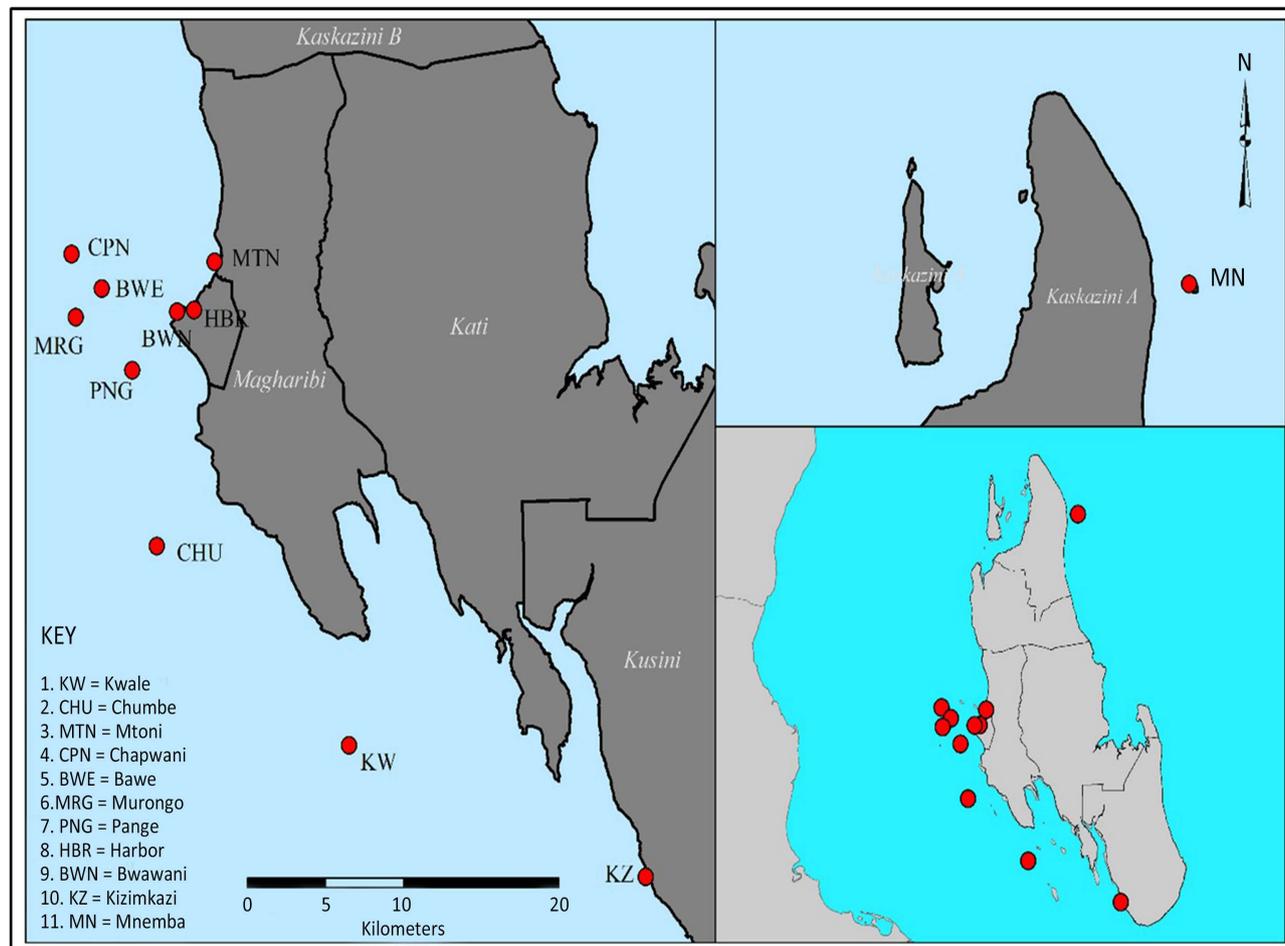
Total of 33 water samples were collected from 11 sampling sites of Zanzibar Island (**Figure 1**). In the process Niskin water sampler was used to collect 1 L of water at least 0.5 m down the subsurface layer in all stations to avoid taking the contaminated micro layer and poured in sampling bottles pre-cleaned with acetone. The samples were then kept under 4°C using ice box and brought to laboratory for diuron analysis within ten days from the sampling date.

### 2.3. Sample Extraction

Water samples were filtrated by vacuum filter with a glass fiber filter grade GF with nominal pore size of 0.7  $\mu\text{m}$  and diameter of 25 mm. Diuron in water was extracted following the solid phase extraction (SPE) and high performance liquid chromatography (HPLC) method of Diuron in crude oil as recommended by the Ministry of Plantation Industries and commodities, Malaysia (**Muhamad et al., 2010**).

Water samples were pre-concentrated in the solid phase extraction cartridges column (SUPELLEAN™, ENVI™-18 Sigma Aldrich, Germany). Prior to the extraction of diuron the columns were conditioned with 10 mL of acetonitrile, followed by methanol and milli Q water respectively. 10 mL of 0.2 M EDTA was added to 1 L of water and then pH of water was adjusted to 3.5. Then 1 mL of 1 mg/L diuron D-6 ( $\text{C}_9\text{H}_4\text{Cl}_2\text{D}_6\text{N}_2\text{O}$ ) was spiked as surrogate standard in order to monitor the recovery of Diuron. Water samples were automatically concentrated in the solid phase extraction column by using solid phase extraction controller

with flow rate of 20 mL/min. Supelclean™ ENVI™-18 cartridges were then dried under nitrogen gas for 5 min. Diuron was eluted from the column using 5 mL of acetonitrile. Finally, acetonitrile was evaporated to 0.2 mL with a stream of pure nitrogen gas.



**Figure 1.** Map of Zanzibar (Unguja) island showing sampling areas.

## 2.4. Sample Analysis

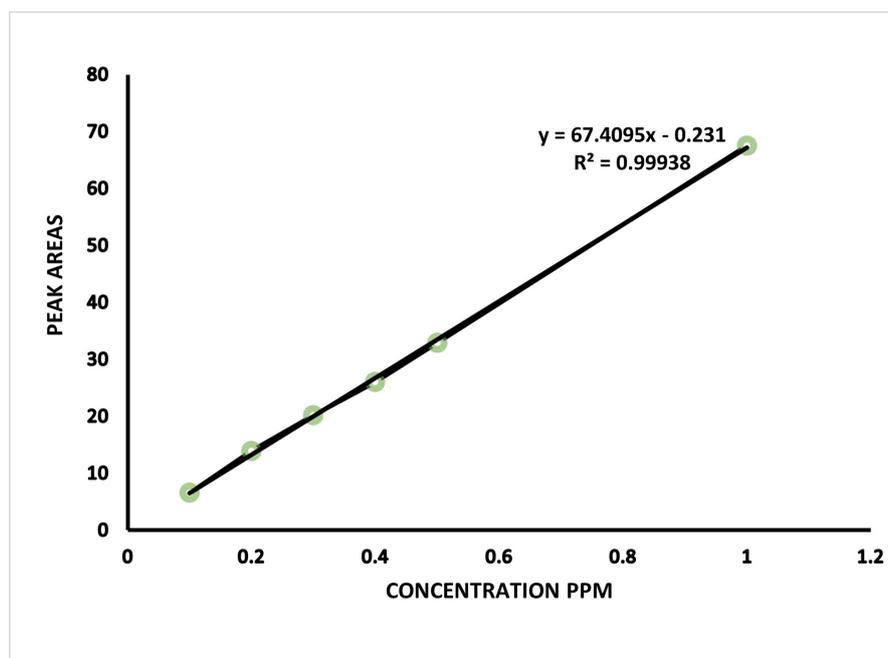
Diuron was analyzed using High Performance Liquid Chromatography (Agilent 1260 HPLC, G1315D Diode Array Detector). The Supelcosil™ LC-18 HPLC column (25 cm × 4.6 mm 5 μm) (Agilent, USA) was used. Separation was obtained using gradient elution at flow rate of 1 mL/min with solvent A (80% milli-Q water) and solvent B (20% Acetonitrile). The determination wavelength was set at 254 nm and the column temperature was kept at 40 °C, the retention time was 3.12 min and injection volume was 1.0 μL. The detection limit was 0.86 ng/L. The calibration data (**Table 1**) and the curve (**Figure 2**) are given below. Recovered concentrations from injected standards were collected from the following formula;

$$\text{Concentration } (\mu\text{g/L}) = (\text{Peak Area} + \text{y-intercept})/\text{Slope}$$

where y-intercept = 0.231 and slope = 67.4095 as indicated in calibration curve.

**Table 1.** Calibration curve data for Diuron (ppm) analysis using HPLC.

Standard Name	Retention time	Peak Area
Standard 0.1	3.128	6.53677
Standard 0.2	3.128	13.928
Standard 0.3	3.128	20.18539
Standard 0.4	3.127	26.02167
Standard 0.5	3.126	32.92434
Standard 1.0	3.128	67.54146

**Figure 2.** Calibration curve of Diuron.

## 2.5. Quality Assurance of Diuron

For the purpose of quality control of the analytical procedure, 1 L of Milli-Q water was treated the same as the real water samples for each batch of analysis as the water sample procedural blank. The recovery tests were also conducted for the purpose of monitoring the efficiency of the methodology. 1 L of Milli-Q water was spiked with known concentrations of diuron D-6 to determine recovery. The sample was processed following same method as real water samples. The recoveries of diuron D-6 were  $92.34\% \pm 4.94\%$  in the spiked samples. The recoveries of Diuron D-6 are shown in **Table 2**.

**Table 2.** Recovery test data of Diuron spiked in remote samples.

S.N	Spiked value (ppm)	Observed value (ppm)	%Recovery
1.	0	0.00	100.00
2.	0.1	0.0875	87.5

**Continued**

3.	0.2	0.1794	89.7
4.	0.4	0.3684	92.1
5.	0.6	0.5322	88.7
6.	0.8	0.788	98.5
7.	1.0	0.899	89.9
		<b>Average</b>	<b>92.34</b>
		<b>SD</b>	<b>4.94</b>

## 2.6. Data Analysis

One-way repeated measures analysis of variance (ANOVA's F test at 5% level of significance) and Tukey's honest significance test (Tukey's HSD) was used to test whether the mean Diuron concentration differs by the study sites and to compare spatial variation of the data.

## 3. Results and Discussion

### 3.1. Levels of Diuron in Water

Diuron was detected in seven (64.6%) sites among 11 sampled sites in Unguja Island with Bwawani1 (BWN1) recorded a maximum value of 1366 ng/L (**Table 3**). The station is closer to the major dockyard in Zanzibar. It is also close to an overcrowded harbor in the archipelago. Furthermore, the station BWN1 is positioned in a bay area with little tides movement and water exchange, which may contribute to the elevation of Diuron. The lowest value of diuron among the detected sites was 260 ng/L at Bawe1 (BWE1). However, Diuron was below detection limit (BDL) in 4 (36.4%) sites, which are Kwale (KW stations), Chumbe (CHU stations), Mtoni (MTN stations), and Murongo (MRG stations) as seen in (**Table 3**).

Chumbe (CHU) is a coral park and one among the Marine Protected Areas (MPAs) in Zanzibar. Although the concentration of Diuron in Chumbe (CHU) was Below Detection Limit (BDL), ecological monitoring needs to be consolidated over a longer period and widely in the region. Monitoring and assessment methodology needs to be standardized and harmonized in order to make the results comparable. Much as the regional collaboration is desirable, it is important to recognize that institutional collaboration has to start at the national level for successful management of the MPAs in the region (**UNEP-WCMC, 2023**).

The detected values of diuron from each site are summarized in **Figure 3**. Results from one-way repeated measures analysis of variance indicated no significance difference in Diuron concentration among measured concentrations at each site (**Figure 3**). It is evident the distributions reflect anthropogenic activities as a possible source of diuron in the sampling areas (**Ali et al., 2021; Sheikh et al., 2016; Lam et al., 2005; Hall Jr. et al., 1999**).

**Table 3.** Location, characteristics of sampling stations and Levels of Diuron (ng/L).

Sites name	N	E	Characteristics of the stations	Samples	Concentration (ng/L)
Kwale (KW)	6.37431	39.28846	Tourism activities	KW1	BDL
				KW2	BDL
				KW3	BDL
Chumbe (CHU)	6.27454	39.17746	Tourism activities, MPA	CHU1	BDL
				CHU2	BDL
				CHU3	BDL
Mtoni (MTN)	6.13289	39.13289	Local cargo station	MTN1	BDL
				MTN2	BDL
				MTN3	BDL
Chapwani (CPN)	6.12437	39.12437	Tourism activities	CPN1	1097
				CPN2	1123
				CPN3	1159
Bawe (BWE)	6.14932	39.14932	Tourism, fishing, and shipyard	BWE1	260
				BWE2	268
				BWE3	263
Murongo (MRG)	6.16168	39.13140	Tourism activities	MRG1	BDL
				MRG2	BDL
				MRG3	BDL
Pange (PNG)	6.18566	39.16447	Tourism activities	PNG1	1096
				PNG2	1006
				PNG3	1040
Harbor (HRB)	6.15441	39.19415	Cargo shipping, passenger speedboats, fishing, and shipyard	HBR1	1026
				HBR2	1086
				HBR3	908
Bwawani (BWN)	6.15605	39.20052	Fishing, dockyards and cargo activities	BWN1	1366
				BWN2	1264
				BWN3	1335
Kizimkazi (KZ)	6.43944	39.45882	Fishing and tourism	KZ1	554
				KZ2	387
				KZ3	407
Mnemba (MN)	5.81637	39.38271	Tourism activities, MPA	MN1	625
				MN2	622
				MN3	614

Note: BDL = Below Detection Limit; MPA = Marine Protected Areas.

### 3.2. Level of Diuron in Sampling Sites Which Exceeding the Maximum Permissible Concentration (MPC)

The maximum permissible concentration of 430 ng/l for Diuron has been set by the appropriate Dutch authorities (Lamoree et al., 2002). Diuron is no longer approved for use in the UK as an active ingredient in antifouling paints, on any size of vessel. In this study the highest Diuron concentration in average is  $1321.67 \pm 52.30$  ng/L recorded at Bwawani (BWN) while the lowest is  $263.33 \pm 4.00$  recorded at Bawe, BWE (Table 4).

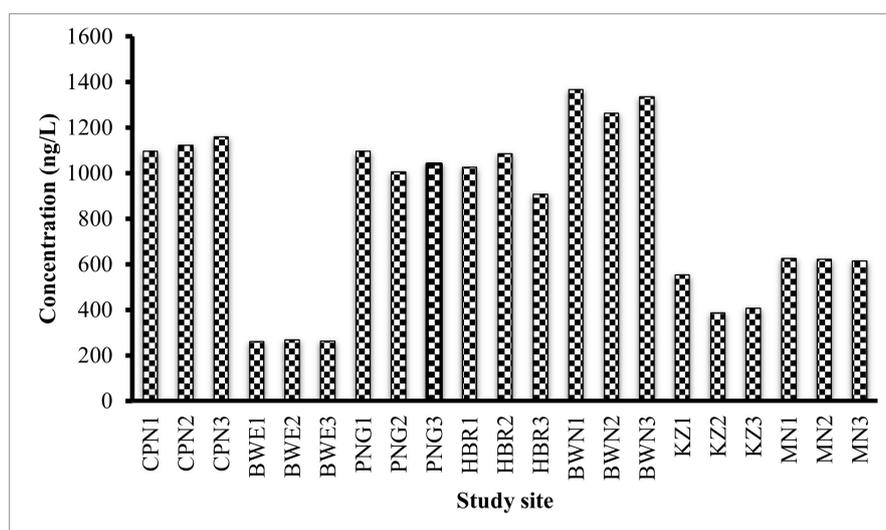
**Table 4.** Mean diuron concentration by study sites.

SITES	Mean $\pm$ S.D (ng/L)	Maximum	Minimum
Bawe (BWE)	$263.67 \pm 4.00$	268	260
Bwawani (BWN)	$1321.67 \pm 52.30$	1366	1264
Chapwani (CPN)	$1126.33 \pm 31.10$	1159	1097
Harbour (HBR)	$1006.67 \pm 90.60$	1086	908
Kizimkazi (KZ)	$449.33 \pm 91.20$	554	387
Mnemba (MN)	$620.33 \pm 5.70$	625	614
Pange (PNG)	$1047.33 \pm 45.40$	1096	1006

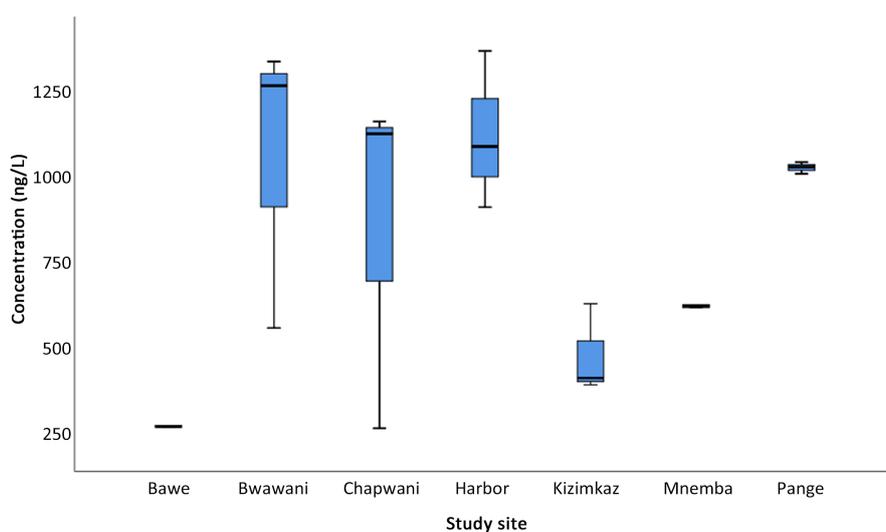
Spatial variations in diuron mean concentrations were noted at Harbour (HBR), Bwawani (BWN) and Kizimkazi (KZ) with little variation observed in Pange (PNG) (Figure 4). However, all sites except Bawe (BWE) regardless of those below detected limits, have shown Diuron concentration above maximum permissible concentration (MPC) of 430 ng/L, as proposed by Dutch National Institute of Public Health and Environment (Giacomazzi & Cochet, 2004) (Figure 5). The average Diuron concentrations detected around Unguja Island were considerably different ( $p = 0.05$ ; at 5% significance level) from the allowed Diuron value of 430 ng/L (Figure 5).

Over the years, increasing pressure has been placed on the marine environment including Marine Protected Areas. The development in technology has brought about an increase in the range of use of marine resources and access to marine environments. For example, the development of large scale fishing industries, tourism, aquaculture and the development of new forms of drugs from marine biodiversity has led to exploitation of marine resources and destruction of the marine Environment (National Research Council, 1994). It should be noted that Mnemba Island (MN site), which is Marine Conservation Area in Zanzibar, has recorded the diuron concentration of up to 625 ng/L (Figure 5), which is above MPC. The main reason may be due to tourism activities taking place in this area. Although diuron concentrations were not record in several study sites, the results of this study indicating that diuron in Zanzibar is a little higher since the majority of the sites (54.6%) records the value above the maximum permissible concentra-

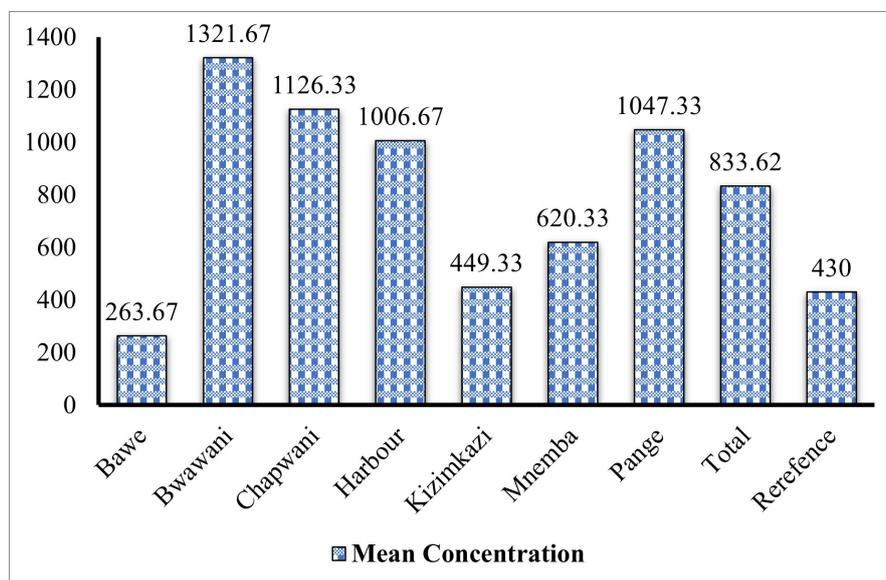
tion (MPC) of 430 ng/L. The ubiquitous presence of diuron is unsurprising since it appears one of the most commonly used antifouling paint booster biocides whereas also having wide spread non-antifouling uses. Predominantly, the use of diuron has been associated with weed control in non-agricultural applications. Several studies have investigated the impact of non-antifouling use of diuron release into the aquatic environment (Albanis et al., 2002; De Almeida Azevedo et al., 2000; Field et al., 2003). It is suggested that the source of diuron in estuaries and coastal areas is supplemented by non-antifouling paint inputs since the estuaries are subject to relatively low volumes of shipping and yachting when compared to marinas and ports water. There is the possibility that some freshwater and seawater from ports and marinas near estuaries might receive considerable agricultural drainage, containing diuron [9].



**Figure 3.** Diuron concentration (ng/L) recorded from each study site.



**Figure 4.** Spatial variation of average diuron concentration (ng/L) in coastal waters of Unguja Island.



**Figure 5.** Comparison of average diuron concentration (ng/L) in coastal waters of Zanzibar Island with Maximum Permissible Concentration (Reference).

### 3.3. Comparison of Diuron Level from Different Clusters and Effects from Other Studies

Diuron residues showed significant differences between average concentrations of the study sites (Table 5) with least of 0.000 for  $p < 0.05$  (ANOVA). It is evident that, distributions reflect anthropogenic activities as a possible source of diuron in the sampling areas (Ali et al., 2021; Sheikh et al., 2016; Lam et al., 2005; Hall Jr. et al., 1999). Moreover, statistical analysis of the results between the clusters; Reef (CPN and MN); Islands (BWE and PNG); Harbor (HBR and BWN); and Coast (KZ) showed a significant difference ( $p < 0.05$ ) of the concentration of Diuron between the clusters (Table 6). The findings showed that the distribution of Diuron was based on the characteristics of the relative site. The highest average concentration of  $1164.17 \pm 184.78$  ng/L was found at the area of dockyards and harbor because the economic activities conducted like cargo shipping, passenger speed boat, fishing, tourism activities such as snorkeling and local cargo increase the level of diuron contamination (Figure 6). Liu et al. (1999) reported that fishing boats require heavy applications of antifouling paint to prevent the growth of fouling organisms, due to the faster wear off of the antifouling chemicals through their heavy use. The lowest average concentration ( $449.33 \pm 91.19$  ng/L) was detected at the coastal area. This is probably due to the fact that the most economic activities in this area are agriculture, which have low possibility of producing high amount of contamination of diuron (Figure 6).

The average concentration in coral reefs areas was  $925.20 \pm 276.29$  ng/L (Figure 6). The coral reefs were positioned at the intermediate contamination of Diuron. The elevated value of Diuron in Reef areas (CPN and MN) is probably due to high level of tourism activities taking place daily compared to other clusters such as coast and Island.

**Table 5.** Spatial variation on concentration of diuron in ng/L using Tukey HSD.

(I) Study site	(J) Study site	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
					Lower Bound	Upper Bound
CPN	BWE	862.667*	46.121	0.000	705.18	1020.15
	PNG	79.000	46.121	0.619	-78.49	236.49
	HBR	119.667	46.121	0.199	-37.82	277.15
	BWN	-195.333*	46.121	0.011	-352.82	-37.85
	KZ	677.000*	46.121	0.000	519.51	834.49
	MN	506.000*	46.121	0.000	348.51	663.49
BWE	CPN	-862.667*	46.121	0.000	-1020.15	-705.18
	PNG	-783.667*	46.121	0.000	-941.15	-626.18
	HBR	-743.000*	46.121	0.000	-900.49	-585.51
	BWN	-1058.000*	46.121	0.000	-1215.49	-900.51
	KZ	-185.667*	46.121	0.017	-343.15	-28.18
	MN	-356.667*	46.121	0.000	-514.15	-199.18
PNG	CPN	-79.000	46.121	0.619	-236.49	78.49
	BWE	783.667*	46.121	0.000	626.18	941.15
	HBR	40.667	46.121	0.970	-116.82	198.15
	BWN	-274.333*	46.121	0.001	-431.82	-116.85
	KZ	598.000*	46.121	0.000	440.51	755.49
	MN	427.000*	46.121	0.000	269.51	584.49
HBR	CPN	-119.667	46.121	0.199	-277.15	37.82
	BWE	743.000*	46.121	0.000	585.51	900.49
	PNG	-40.667	46.121	0.970	-198.15	116.82
	BWN	-315.000*	46.121	0.000	-472.49	-157.51
	KZ	557.333*	46.121	0.000	399.85	714.82
	MN	386.333*	46.121	0.000	228.85	543.82
BWN	CPN	195.333*	46.121	0.011	37.85	352.82
	BWE	1058.000*	46.121	0.000	900.51	1215.49
	PNG	274.333*	46.121	0.001	116.85	431.82
	HBR	315.000*	46.121	0.000	157.51	472.49
	KZ	872.333*	46.121	0.000	714.85	1029.82
	MN	701.333*	46.121	0.000	543.85	858.82
KZ	CPN	-677.000*	46.121	0.000	-834.49	-519.51
	BWE	185.667*	46.121	0.017	28.18	343.15
	PNG	-598.000*	46.121	0.000	-755.49	-440.51
	HBR	-557.333*	46.121	0.000	-714.82	-399.85
	BWN	-872.333*	46.121	0.000	-1029.82	-714.85
	MN	-171.000*	46.121	0.030	-328.49	-13.51

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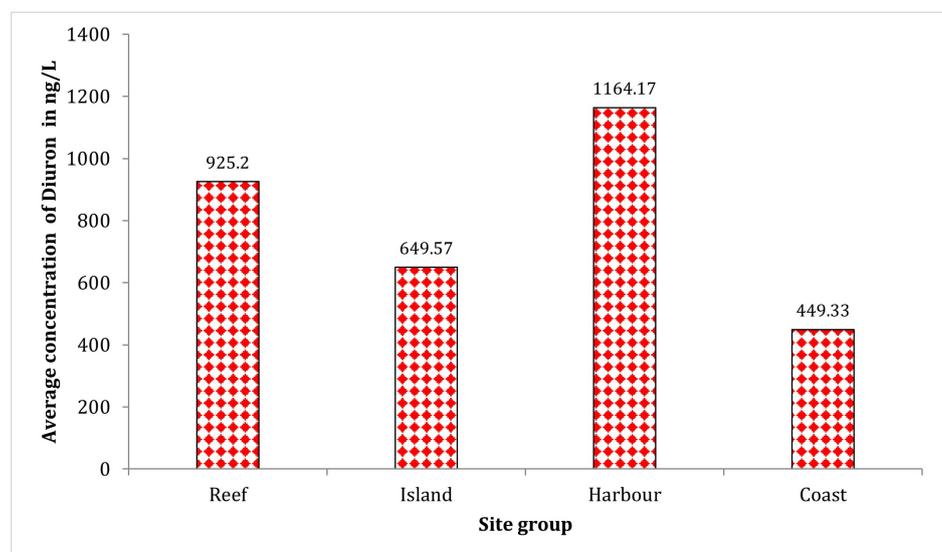
MN	CPN	-506.000*	46.121	0.000	-663.49	-348.51
	BWE	356.667*	46.121	0.000	199.18	514.15
	PNG	-427.000*	46.121	0.000	-584.49	-269.51
	HBR	-386.333*	46.121	0.000	-543.82	-228.85
	BWN	-701.333*	46.121	0.000	-858.82	-543.85
	KZ	171.000*	46.121	0.030	13.51	328.49

Note: \*The mean difference is significant at the 0.05 level.

**Table 6.** Mean pairwise comparison for sites group.

(I) Site group	(J) Site group	Mean Difference (I-J)	Std. Error	Sig.	95% Confidence Interval	
					Lower Bound	Upper Bound
Reef	Island	275.629	169.202	0.390	-205.34	756.59
	Harbor	-238.967	174.978	0.536	-736.35	258.42
	Coast	475.867	211.032	0.148	-124.00	1075.74
Island	Reef	-275.629	169.202	0.390	-756.59	205.34
	Harbor	-514.595*	160.766	0.024	-971.58	-57.61
	Coast	200.238	199.406	0.749	-366.59	767.06
Harbor	Reef	238.967	174.978	0.536	-258.42	736.35
	Island	514.595*	160.766	0.024	57.61	971.58
	Coast	714.833*	204.331	0.013	134.01	1295.66
Coast	Reef	-475.867	211.032	0.148	-1075.74	124.00
	Island	-200.238	199.406	0.749	-767.06	366.59
	Harbor	-714.833*	204.331	0.013	-1295.66	-134.01

Note: \*The mean difference is significant at the 0.05 level.



**Figure 6.** Mean concentration of Diuron from four categories.

In addition to that, the statistical information between each category varied, where their mean variation was significant in some stations. For example, the average concentration of Diuron between Reef category and Small Island showed statistically there is no significant variation ( $p > 0.05$ ). On the other hand, the average concentration between Reef and Harbour, Reef and Coast, and between Coast and Island showed there were not significant. However, the average concentration between Island and Harbour and between Harbour and Coast statistically showed significant variation (**Table 6**). Variation was probably due to difference of the activities and boat density (Ali et al., 2021; Sheikh et al., 2016). The main sources of the biocides could come from increasing in activities in harbor/dockyard such as passengers, cargo and fishing as well as tourism boating activities during snorkeling and dolphin watching. It is important to continue monitoring of the biocides.

The maximum concentrations measured from different regions are much higher compared to the maximum concentration found around coastal areas of Unguja Island. For example concentration up to 3,050 ng/L was detected in Seto Inland Sea, Japan (Okamura et al., 2003); 42,000 ng/L in Lagoon water Italy (Genaro et al., 1995); 6742 ng/L estuaries UK, (Thomas et al., 2002); 2,000 ng/L in Mediterranean coast, Spain, (Martinez et al., 2000). In contrast to the said examples, however, the findings of the current study demonstrate that the level of Diuron around coastal Unguja Island is relatively low. Different researches have verified numerous effects of diuron on corals, such as the loss of symbiotic algae in *M. digitata* and *S. hystrix* (Jones, 2004), reduction the calcification rate of *G. fascicularis* (Sheikh et al., 2009), tissue retraction of *P. damicomis* (Negri et al., 2004), reduction of respiration of *P. cylindrical* (Råberg et al., 2003), the detachment of soft tissue of juvenile of *Acropora tenuis* (Watanabe et al., 2006) and the reduction of  $^{14}\text{C}$  incorporation in *Madracis mirabilis* (Owen et al., 2003). Definitely, detection of diuron in marine environments causes a number of ecological effects.

#### 4. Conclusion

This paper presents basic information of N'-(3,4-dichlorophenyl)-N,N-dimethylurea (Diuron) contamination as a fingerprint of anthropogenic impacts on coral reef ecosystems around coastal area of Zanzibar Island. The study specifically, focused on spatial variation and quantification of Diuron concentration among the study sites. Different sites such as Harbour (HBR), Pange (PNG), Chapwani (CPN) and Bwawani (BWN) recorded higher level of greater than 1000 ng/L. Additionally, results of this study were compared with the proposed Maximum Permissible Concentration (MPC) of 430 ng/L, as recommended by Dutch National Institute of Public Health and the Environment. It was found that six sites among eleven (54.6%) had higher value above MPC. Finally, the findings were compared to other reported concentrations from different countries as well as the acute exposure experimental data to corals and other marine organisms. The maximum value in this study was  $1321.67 \pm 52.3$  ng/L at Bwawani which is located near to harbor, but also fishing,

dockyards and cargo activities are taking place as well.

Apart from the fact that the concentration of diuron in Marine Protected Areas (MPAs) in Zanzibar such as Chumbe Island is below detected limit, overall results from other sites suggest that Zanzibar Island might be contaminated with Diuron from various sources such as agriculture, urban uses and shipping activities while coral reef Islands, might be contaminated from high boating activities. The results reveal that Diuron contamination at Zanzibar is at levels of causing concern since most recorded values (54.6%) were above the maximum permissible concentration of 430 ng/L set by Dutch Authorities. It can be concluded that, although the data obtained are far below the lowest observable effects concentration (LOEC) to corals as shown by laboratories of eco-toxicological studies in the world, but still are undefined how the chronic exposure of ambient concentration of Diuron will affect corals in Zanzibar Island.

Based on the current data of diuron residue found in Zanzibar the followings are recommended:

1. It is important for Zanzibar Maritime Authorities and other stakeholders to pay particular attention and explore the existence of Diuron in other coastal regions of Zanzibar including MPAs. This will give clear understanding of the environmental assessment around coastal areas of the Islands.
2. Determination of diuron in other sources such as sediments and organisms should be given priority in future studies.
3. Short and long term exposure on the antifouling biocide Diuron particularly on monitoring, chronic exposure, risk assessment, organism's responses and pollution models would add special value towards better understanding of the mechanisms and sustainable marine ecosystem health.
4. The findings of this study should be considered useful for the formulation of technical strategies to control new antifouling contamination, risk assessment and develop new alternative antifouling chemicals in coastal waters.
5. Data should be shared to many parties especially government agencies, private sectors, researchers and other stake holders for future plan and conserving the ecological marine biodiversity especially in MPAs and coral reef Islands.

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

## References

Agardy, T. S. (1997). *Marine Protected Area Typologies* (p. 244). Elsevier.

- Albanis, T. A., Lambropoulou, D. A., Sakkas, V. A., & Konstantinou, I. K. (2002). Antifouling Paint Booster Biocide Contamination in Greek Marine Sediments. *Chemosphere*, *48*, 475-485. [https://doi.org/10.1016/s0045-6535\(02\)00134-0](https://doi.org/10.1016/s0045-6535(02)00134-0)
- Ali, H. R., Ariffin, M. M., Omar, T. F. T., Ghazali, A., Sheikh, M. A., Shazili, N. A. M. et al. (2021). Antifouling Paint Biocides (Irgarol 1051 and Diuron) in the Selected Ports of Peninsular Malaysia: Occurrence, Seasonal Variation, and Ecological Risk Assessment. *Environmental Science and Pollution Research*, *28*, 52247-52257. <https://doi.org/10.1007/s11356-021-14424-1>
- Ali, H. R., Arifin, M. M., Sheikh, M. A., Shazili, N. A. M., Bakari, S. S., & Bachok, Z. (2014). Contamination of Diuron in Coastal Waters around Malaysian Peninsular. *Marine Pollution Bulletin*, *85*, 287-291. <https://doi.org/10.1016/j.marpolbul.2014.05.049>
- Boxall, A. B. A., Comber, S. D., Conrad, A. U., Howcroft, J., & Zaman, N. (2000). Inputs, Monitoring and Fate Modelling of Antifouling Biocides in UK Estuaries. *Marine Pollution Bulletin*, *40*, 898-905. [https://doi.org/10.1016/s0025-326x\(00\)00021-7](https://doi.org/10.1016/s0025-326x(00)00021-7)
- Dahl, B., & Blanck, H. (1996). Toxic Effects of the Antifouling Agent Irgarol 1051 on Periphyton Communities in Coastal Water Microcosms. *Marine Pollution Bulletin*, *32*, 342-350. [https://doi.org/10.1016/0025-326x\(96\)84828-4](https://doi.org/10.1016/0025-326x(96)84828-4)
- de Almeida Azevedo, D., Lacorte, S., Vinhas, T., Viana, P., & Barceló, D. (2000). Monitoring of Priority Pesticides and Other Organic Pollutants in River Water from Portugal by Gas Chromatography-Mass Spectrometry and Liquid Chromatography-Atmospheric Pressure Chemical Ionization Mass Spectrometry. *Journal of Chromatography A*, *879*, 13-26. [https://doi.org/10.1016/s0021-9673\(00\)00372-1](https://doi.org/10.1016/s0021-9673(00)00372-1)
- Ferrer, I., & Barceló, D. (1999). Simultaneous Determination of Antifouling Herbicides in Marina Water Samples by On-Line Solid-Phase Extraction Followed by Liquid Chromatography-Mass Spectrometry. *Journal of Chromatography A*, *854*, 197-206. [https://doi.org/10.1016/s0021-9673\(99\)00506-3](https://doi.org/10.1016/s0021-9673(99)00506-3)
- Ferrer, I., Ballesteros, B., Marco, M. P., & Barceló, D. (1997). Pilot Survey for Determination of the Antifouling Agent Irgarol 1051 in Enclosed Seawater Samples by a Direct Enzyme-Linked Immunosorbent Assay and Solid-Phase Extraction Followed by Liquid Chromatography-Diode Array Detection. *Environmental Science & Technology*, *31*, 3530-3535. <https://doi.org/10.1021/es970292c>
- Field, J. A., Reed, R. L., Sawyer, T. E., Griffith, S. M., & Wigington, P. J. (2003). Diuron Occurrence and Distribution in Soil and Surface and Ground Water Associated with Grass Seed Production. *Journal of Environmental Quality*, *32*, 171-179. <https://doi.org/10.2134/jeq2003.171>
- Gennaro, M. C., Abrigo, C., Giacosa, D., Rigotti, L., & Liberatori, A. (1995). Separation of Phenylurea Pesticides by Ion-Interaction Reversed-Phase High-Performance Liquid Chromatography Diuron Determination in Lagoon Water. *Journal of Chromatography A*, *718*, 81-88. [https://doi.org/10.1016/0021-9673\(95\)00659-1](https://doi.org/10.1016/0021-9673(95)00659-1)
- Giacomazzi, S., & Cochet, N. (2004). Environmental Impact of Diuron Transformation: A Review. *Chemosphere*, *56*, 1021-1032. <https://doi.org/10.1016/j.chemosphere.2004.04.061>
- Hall Jr., LW., Giddings, J. M., Solomon, K. R., & Balcomb, R. (1999). An Ecological Risk Assessment for Use of Irgarol 1051 as an Algaecide for Antifoulants Paints. *Critical Reviews in Toxicology*, *29*, 367-437.
- Haynes, D., Ralph, P., Prange, J., & Dennison, B. (2000). The Impact of the Herbicide Diuron on Photosynthesis in Three Species of Tropical Seagrass. *Marine Pollution Bulletin*, *41*, 288-293. [https://doi.org/10.1016/s0025-326x\(00\)00127-2](https://doi.org/10.1016/s0025-326x(00)00127-2)

- Jones, R. (2004). Testing the 'Photoinhibition' Model of Coral Bleaching Using Chemical Inhibitors. *Marine Ecology Progress Series*, 284, 133-145. <https://doi.org/10.3354/meps284133>
- Jones, R. (2005). The Ecotoxicological Effects of Photosystem II Herbicides on Corals. *Marine Pollution Bulletin*, 51, 495-506. <https://doi.org/10.1016/j.marpolbul.2005.06.027>
- Jones, R., & Kerswell, A. (2003). Phytotoxicity of Photosystem II (PSII) Herbicides to Coral. *Marine Ecology Progress Series*, 261, 149-159. <https://doi.org/10.3354/meps261149>
- Konstantinou, I. K., & Albanis, T. A. (2004). Worldwide Occurrence and Effects of Anti-fouling Paint Booster Biocides in the Aquatic Environment: A Review. *Environment International*, 30, 235-248. [https://doi.org/10.1016/s0160-4120\(03\)00176-4](https://doi.org/10.1016/s0160-4120(03)00176-4)
- Lam, K., Cai, Z., Wai, H., Tsang, V. W., Lam, M. H., Cheung, R. Y. et al. (2005). Identification of a New Irgarol-1051 Related S-Triazine Species in Coastal Waters. *Environmental Pollution*, 136, 221-230. <https://doi.org/10.1016/j.envpol.2005.01.014>
- Lamoree, M. H., Swart, C. P., van der Horst, A., & van Hattum, B. (2002). Determination of Diuron and the Antifouling Paint Biocide Irgarol 1051 in Dutch Marinas and Coastal Waters. *Journal of Chromatography A*, 970, 183-190. [https://doi.org/10.1016/s0021-9673\(02\)00878-6](https://doi.org/10.1016/s0021-9673(02)00878-6)
- Levine, A. (2007). Staying Afloat: State Agencies, Local Communities, and International Involvement in Marine Protected Area Management in Zanzibar, Tanzania. *Conservation & Society*, 5, 562-565.
- Lewis, S. E., Brodie, J. E., Bainbridge, Z. T., Rohde, K. W., Davis, A. M., Masters, B. L. et al. (2009). Herbicides: A New Threat to the Great Barrier Reef. *Environmental Pollution*, 157, 2470-2484. <https://doi.org/10.1016/j.envpol.2009.03.006>
- Liu, D., Pacepavicius, G. J., Maguire, R. J., Lau, Y. L., Okamura, H., & Aoyama, I. (1999). Survey for the Occurrence of the New Antifouling Compound Irgarol 1051 in the Aquatic Environment. *Water Research*, 33, 2833-2843. [https://doi.org/10.1016/s0043-1354\(98\)00501-6](https://doi.org/10.1016/s0043-1354(98)00501-6)
- Malato, S., Blanco, J., Cáceres, J., Fernández-Alba, A. R., Agüera, A., & Rodríguez, A. (2002). Photocatalytic Treatment of Water-Soluble Pesticides by Photo-Fenton and TiO<sub>2</sub> Using Solar Energy. *Catalysis Today*, 76, 209-220. [https://doi.org/10.1016/s0920-5861\(02\)00220-1](https://doi.org/10.1016/s0920-5861(02)00220-1)
- Malato, S., Blanco, J., Richter, C., & Maldonado, M. I. (2000a). Optimization of Pre-Industrial Solar Photocatalytic Mineralization of Commercial Pesticides. *Applied Catalysis B: Environmental*, 25, 31-38. [https://doi.org/10.1016/s0926-3373\(99\)00114-9](https://doi.org/10.1016/s0926-3373(99)00114-9)
- Malato, S., Blanco, J., Richter, C., Fernández, P., & Maldonado, M. I. (2000b). Solar Photocatalytic Mineralization of Commercial Pesticides: Oxamyl. *Solar Energy Materials and Solar Cells*, 64, 1-14. [https://doi.org/10.1016/s0927-0248\(00\)00037-4](https://doi.org/10.1016/s0927-0248(00)00037-4)
- Malato, S., Blanco, J., Fernández-Alba, A. R., & Agüera, A. (2000c). Solar Photocatalytic Mineralization of Commercial Pesticides: Acrinathrin. *Chemosphere*, 40, 403-409. [https://doi.org/10.1016/s0045-6535\(99\)00267-2](https://doi.org/10.1016/s0045-6535(99)00267-2)
- Martinez, K., & Barceló, D. (2001). Determination of Antifouling Pesticides and Their Degradation Products in Marine Sediments by Means of Ultrasonic Extraction and HPLC-APCI-MS. *Fresenius' Journal of Analytical Chemistry*, 370, 940-945. <https://doi.org/10.1007/s002160100904>
- Martinez, K., Ferrer, I., & Barceló, D. (2000). Part-Per-Trillion Level Determination of Antifouling Pesticides and Their Byproducts in Seawater Samples by Off-Line Solid-Phase Extraction Followed by High-Performance Liquid Chromatography-Atmospheric Pressure Chemical Ionization Mass Spectrometry. *Journal of Chromatography A*, 879, 27-37.

- [https://doi.org/10.1016/s0021-9673\(00\)00307-1](https://doi.org/10.1016/s0021-9673(00)00307-1)
- Muhamad, H., Ramli, M. I., Zakaria, Z., & Sahid, I. (2010). *Determination of Diuron in Crude Palm Oil and Crude PALM kernel Oil by Solid Phase Extraction and High Performance Liquid Chromatography Using Ultra Violet Detection. Malaysian Palm Oil Board Information Series*. Ministry of Plantation Industries and commodities.
- National Research Council (1994). *Restoring and Protecting Marine Habitat: Role of Engineering and Technology* (p 193).
- Negri, A. P., Hales, L. T., Battershill, C., Wolff, C., & Webster, N. S. (2004). TBT Contamination Identified in Antarctic Marine Sediments. *Marine Pollution Bulletin*, 48, 1142-1144. <https://doi.org/10.1016/j.marpolbul.2004.03.004>
- Okamura, H. (2002). Photodegradation of the Antifouling Compounds Irgarol 1051 and Diuron Released from a Commercial Antifouling Paint. *Chemosphere*, 48, 43-50. [https://doi.org/10.1016/s0045-6535\(02\)00025-5](https://doi.org/10.1016/s0045-6535(02)00025-5)
- Okamura, H., Aoyama, I., Ono, Y., & Nishida, T. (2003). Antifouling Herbicides in the Coastal Waters of Western Japan. *Marine Pollution Bulletin*, 47, 59-67. [https://doi.org/10.1016/s0025-326x\(02\)00418-6](https://doi.org/10.1016/s0025-326x(02)00418-6)
- Owen, R., Knap, A., Ostrander, N., & Carbery, K. (2003). Comparative Acute Toxicity of Herbicides to Photosynthesis of Coral Zooxanthellae. *Bulletin of Environmental Contamination and Toxicology*, 70, 541-548. <https://doi.org/10.1007/s00128-003-0020-6>
- Råberg, S., Nyström, M., Erös, M., & Plantman, P. (2003). Impact of the Herbicides 2, 4-D and Diuron on the Metabolism of the Coral *Porites Cylindrica*. *Marine Environmental Research*, 56, 503-514. [https://doi.org/10.1016/s0141-1136\(03\)00039-4](https://doi.org/10.1016/s0141-1136(03)00039-4)
- Sathiyaseelan, K., & Stella, D. (2011). Isolation, Identification and Antagonistic Activity of Marine Actinomycetes Isolated from the Muthupet Mangrove Environment. *International Journal of Pharmaceutical and Biological Achieves*, 2, 1464-1466.
- Sheikh, M. A., Fujimura, H., Miyagi, T., Uechi, Y., Yokota, T., Yasumura, S. et al. (2009). Detection and Ecological Threats of PSII Herbicide Diuron on Coral Reefs around the Ryukyu Archipelago, Japan. *Marine Pollution Bulletin*, 58, 1922-1926. <https://doi.org/10.1016/j.marpolbul.2009.09.010>
- Sheikh, M. A., Juma, F. S., Staehr, P., Dahl, K., Rashid, R. J., Mohammed, M. S. et al. (2016). Occurrence and Distribution of Antifouling Biocide Irgarol-1051 in Coral Reef Ecosystems, Zanzibar. *Marine Pollution Bulletin*, 109, 586-590. <https://doi.org/10.1016/j.marpolbul.2016.05.035>
- Sheppard, C. R. C., Spalding, M., Bradshaw, C., & Wilson, S. (2002). Erosion Vs. Recovery of Coral Reefs after 1998 El Niño: Chagos Reefs, Indian Ocean. *AMBIO: A Journal of the Human Environment*, 31, 40-48. [https://doi.org/10.1639/0044-7447\(2002\)031\[0040:evrocr\]2.0.co;2](https://doi.org/10.1639/0044-7447(2002)031[0040:evrocr]2.0.co;2)
- Thomas, K. V., McHugh, M., & Waldock, M. (2002). Antifouling Paint Booster Biocides in UK Coastal Waters: Inputs, Occurrence and Environmental Fate. *Science of the Total Environment*, 293, 117-127. [https://doi.org/10.1016/s0048-9697\(01\)01153-6](https://doi.org/10.1016/s0048-9697(01)01153-6)
- UNEP-WCMC (2023). *Protected Area Profile for United Republic of Tanzania from the World Database on Protected Areas*. <https://www.protectedplanet.net>
- Watanabe, T., Yuyama, I., & Yasumura, S. (2006). Toxicological Effects of Biocides on Symbiotic and Aposymbiotic Juveniles of the Hermatypic Coral *Acropora tenuis*. *Journal of Experimental Marine Biology and Ecology*, 339, 177-188. <https://doi.org/10.1016/j.jembe.2006.07.020>