Attachment 8

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Pesticide and Herbicide Residues in Sediments and Seagrasses from the Great Barrier Reef World Heritage Area and Queensland Coast

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Pesticides and herbicides including organochlorine compounds have had extensive current and past application by Queensland's intensive coastal agriculture industry as well as for a wide range of domestic, public health and agricultural purposes in urban areas. The persistent nature of these types of compounds together with possible continued illegal use of banned organochlorine compounds raises the potential for continued long-term chronic exposure to plants and animals of the Great Barrier Reef. Sediment and seagrass samples were collected from 16 intertidal and 25 subtidal sampling sites between Torres Strait and Townsville, near Mackay and Gladstone, and in Hervey and Moreton Bays in 1997 and 1998 and analysed for pesticide and herbicide residues. Low levels of atrazine (0.1–0.3 μg kg^{-1}), diuron (0.2–10.1 μg kg^{-1}), lindane (0.08–0.19 μg kg^{-1}), dieldrin (0.05–0.37 μg kg^{-1}), DDT (0.05–0.26 μg kg^{-1}), and DDE (0.05–0.26 μg kg^{-1}) were detected in sediments and/or seagrasses. Contaminants were mainly detected in samples collected along the high rainfall, tropical coast between Townsville and Port Douglas and in Moreton Bay. Of the contaminants detected, the herbicide diuron is of most concern as the concentrations detected have some potential to impact local seagrass communities. © 2000 Elsevier Science Ltd. All rights reserved.

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Introduction

Persistent organochlorine compounds such as PCBs, DDTs and HCHs are ubiquitous environmental pollutants in the global ecosystem (Loganathan and Kannan, 1991; Voldner and Li, 1995) and were produced and used extensively in Australia until the late 1980s (Kannan *et al.*, 1994). Compounds including lindane (γ-HCH), aldrin, heptachlor, chlordane, DDT and dieldrin were used extensively in Queensland agricultural applications (sugarcane, banana and dairying) for the control of insects and weeds (Hamdorf, 1992; Von Westernhagen and Klumpp, 1995) and for a wide range of domestic, public health and agricultural purposes in urban areas (Mortimer, 1998). As a consequence, these pollutants have been detected as contaminants in northern Australian groundwaters (Brodie *et al.*, 1984), seawater (Tanabe *et al.*, 1982; Kurtz and Atlas, 1990), freshwater biota (Thomson and Davie, 1974; Russell *et al.*, 1996a), freshwater and estuarine sediments (Dyall and Johns, 1985; Mortimer, 1998) and marine biota (McCloskey and Duebert, 1972; Olafson, 1978; Hamdorf, 1992; Von Westernhagen and Klumpp, 1995; Kannan *et al.*, 1995; Moss and Mortimer, 1996). Seafoods for human consumption collected along the tropical north-eastern Australian seaboard have also been shown to be contaminated with low levels of polychlorinated biphenyls, DDT and its metabolites, chlordane compounds and lindane isomers (Kannan *et al.*, 1994).

Although most organochlorine pesticides were banned for use in Queensland in the late 1980s, large quantities of farm chemicals including organochlorines in liquid formulation are still held on farming properties in Queensland (McGuffog *et al.*, 1996). It is estimated that at least 26 t of unwanted chlorinated pesticides (including DDT, endrin and lindane) are stored on farming properties in Queensland (McGuffog *et al.*, 1996). Over 50% of these are located in catchments adjacent to the Great Barrier Reef. In addition, a number of triazine (atrazine) and organochlorine and phenylurea herbicides (diuron and 2,4-D) and organophosphate pesticides (chlorpyrifos) are still in wide use

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by the Queensland sugarcane industry (Hamilton and Haydon, 1996). Sugarcane production is the largest intensive agricultural industry carried out in Queensland and industry sales generated approximately AUS $1.2 billion in 1991 (Anon., 1992). The industry is situated primarily along the coastal fringe and is concentrated in northern Queensland, between Gladstone and Cairns. The persistent nature of many pesticides together with possible continued illegal use of banned organochlorine compounds raises the potential for continued long-term chronic exposure to plants and animals, including mangroves, seagrasses and corals of the Great Barrier Reef. The data presented here represent the first survey of contemporary concentrations of organochlorine pollutants in marine sediment and seagrass samples collected along the Great Barrier Reef and greater Queensland coastline.

Materials and Methods

Intertidal sediment and seagrass sampling

Sediment and seagrass samples were collected from 16 Queensland intertidal (<1 m deep) sites located between Cape York and Moreton Bay during February and May 1997 (Fig. 1). All sampling locations (with the exception of Cairns and Low Isles) are in the vicinity of important habitat of the dugong (*Dugong dugon*), a seagrass grazing marine mammal that has become endangered or exterminated over much of its range (Marsh and Saalfeld, 1989, 1990; Preen, 1993; Marsh *et al.*, 1995; Marsh and Corkeron, 1997). Three replicate 2 l sediment samples and three replicate 2 l quantities of seagrass were collected from each sampling location. All samples were collected in solvent-washed glass containers. Each sediment sample was a composite of multiple surficial sediment samples collected randomly over an area of approximately 400 m^2. A random sample of the dominant seagrass (*Cymodocea serrulata, Halodule uninervis* or *Zostera capricorni*) was also collected over the same area. Entire plants (leaves, roots and rhizomes) were sampled. Sediment and seagrass samples were frozen following collection.

Subtidal sediment sampling

Sediment samples were also collected from 25 subtidal Queensland locations between Torres Strait and Gladstone between June and November 1998 (Fig. 1). All sampling sites were located in shallow (typically <5 m) water in major estuaries and northward facing bays along the northern and central Queensland coast. Two replicate 2 l sediment samples were collected from each sampling location. Each replicate sample was composited from 3 random grab samples collected from bottom sediments using a modified Van-Veen grab sampler. Replicate samples were collected approximately 500–1000 m apart. All samples were collected in solvent-washed glass containers. Sediment samples were frozen following collection.

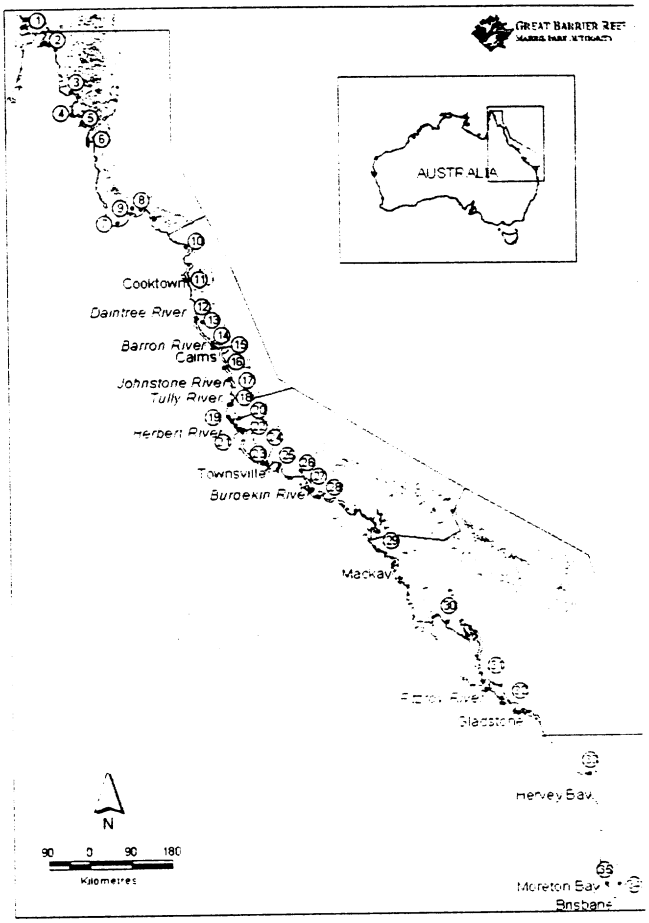

Fig. 1 Sediment and seagrass sampling sites: 1. Horn Island, 2. Newcastle Bay, 3. Shelburne Bay, 4. Temple Bay, 5. Weymouth Bay, 6. Lloyd Bay, 7. Princess Charlotte Bay, 8. Bathurst Bay, 9. Flinders Island, 10. Cape Flattery, 11. Walker Bay, 12. Daintree River, 13. Low Isles, 14. Barron River, 15. Cairns, 16. Russell River, 17. Johnstone River, 18. Tully River, 19. Cardwell, 20. Hinchinbrook, 21. Herbert River, 22. Lucinda, 23. Halifax Bay, 24. Pallarenda, 25. Cleveland Bay, 26. Bowling Green Bay, 27. Burdekin River, 28. Upstart Bay, 29. Newry Bay, 30. Shoalwater Bay, 31. Fitzroy River, 32. Gladstone, 33. Hervey Bay, 34. East Moreton Bay, 35. West Moreton Bay.

Analyses of sediment and seagrass samples for pesticide residues and PCBs

Sediment and seagrass samples were analysed at the NATA-certified pesticide laboratory of Queensland Health Scientific Services, Brisbane, using routine laboratory methods with a series of QC/QA procedures. For the majority of compounds, which included aldrin, atrazine, chlorpyrifos, endosulfan (α, β and endosulfan sulfate), dieldrin, DDTs (*pp*-DDD, *pp*-DDE and *pp*-DDT), diuron, hexachlorobenzene, heptachlor, heptachlorepoxide, lindane and polychlorinated biphenyls (PCBs), aliquots were extracted using acetone/*n*-hexane (1:1). The extracts were centrifuged and the supernatant was subject to repeated liquid liquid partitioning with dichloromethane. The combined fractions of dichloromethane containing the compounds of interest were concentrated, transferred into *n*-hexane and made up to a volume of 3 ml. One ml of the extract was set aside for

analysis of diuron and atrazine, while the second fraction of 2 ml (67% of extract) was subject to a clean-up on Florisil™ (5% deactivated with H_2O). Compounds of interest were eluted in two separate fractions using *n*-hexane diethylether (94 6 v v) followed by *n*-hexane acetone (90 10 v v).

The individual fractions were concentrated to 1 ml and the first fraction was analysed for chlorpyrifos using a gas chromatograph equipped with a flame photometric detector (GC–FPD) for quantification and a gas chromatograph equipped with nitrogen phosphorous detector (GC–NPD) for confirmation. The first fraction (*n*-hexane diethylether eluate) was then subjected to a sulphur removal clean-up adding tetrabutyl ammonium hydrogen sulfate saturated with anhydrous sodium sulphite, 2 ml isopropanol and small amounts of extra anhydrous sodium sulphite. After shaking and phase separation, the top layer was carefully transferred using an extra 2 ml of *n*-hexane and concentrated under a gentle stream of nitrogen to 1 ml. Both the first fraction (after sulphur clean-up) and the second fraction directly from the Florisil™ column concentration, were then analysed for aldrin, endosulfan (α, β and endosulfan sulfate), dieldrin, DDTs (*pp*-DDD, *pp*-DDE and *pp*-DDT), hexachlorobenzene, heptachlor, heptachlorepoxide, lindane and polychlorinated biphenyls (PCBs) using a dual column gas chromatograph equipped with electron capture detectors.

For the 1 ml fraction (33% of extract) set aside for diuron and atrazine analysis, the solvent was exchanged by carefully evaporating the *n*-hexane and subsequently adding first water and then methanol. The samples were made up to a final volume of 1 ml and analysed on a high performance liquid chromatography system coupled with a triple stage quadropole mass spectrometer (API 300, Perkin–Elmer Sciex Instruments, Thornhill, Ont., Canada).

Samples were also analysed for 2,4-D and 2,4,5-T. Subsamples were extracted using 100 ml of 0.1 M NaOH in H_2O. The samples were centrifuged and the supernatant was decanted, acidified to a pH < 2 using concentrated H_2SO_4. If precipitation occurred after acidification, then the centrifugation was repeated. The compounds of interest were then extracted into diethylether (2 × 100 ml) using liquid–liquid partitioning. The diethylether was filtered through anhydrous sodium sulfate and the combined fractions are concentrated to approximately 2 ml. The compounds of interest were instantaneously methylated using freshly prepared diazomethane, which was collected in diethylether prior to use. The extract was then concentrated, transferred into *n*-hexane and made up to a final volume. The samples were analysed for 2,4,5-T using a gas chromatograph coupled to a mass-spectrometer. Quantification was performed in selective ion monitoring mode and confirmed using full ion scan.

Seagrass samples were thawed and one replicate from each sampling location was selected at random for analysis. Seagrass tissue was thoroughly washed in deionised water. Extraction, clean-up and quantification for the individual compound groups were similar to the methods described above for sediments. However, analysis of aldrin, chlorpyrifos, endosulfan (α, β and endosulfan sulfate), dieldrin, DDTs (*pp*-DDD, *pp*-DDE and *pp*-DDT), hexachlorobenzene, heptachlor, heptachlorepoxide, lindane and polychlorinated biphenyls (PCBs) in seagrass required an extra filtration step prior to the liquid partition through a Whatman 542 filter. Furthermore, after the liquid partitioning step and before the florisil clean-up, gel permeation chromatography (GPC) was used to purify the extracts. In brief, the 2-ml fraction after the liquid–liquid partitioning step was filtered through a 0.45-μm filter (Millex FH). The sample was then injected into the GPC (Envirogel) with DCM as the mobile phase. The fractionation containing the compounds of interest were collected, reduced, transferred into *n*-hexane, further purified on florisil and analysed as described for sediments.

For 2,4-D and 2,4,5-T analysis, seagrass samples were extracted into acetone using a high speed blender. The acetone was then filtered through a Whatman 542 filter, concentrated, transferred to a conical flask using diethylether and hydrolysed with KOH (37% w v) on a water bath for 1 h. Before liquid–liquid partitioning the pH was reduced to < 2 using H_2SO_4 and the method was continued as described for sediment samples.

All analytical methods were subject to standard QA QC procedures. Reagent blanks and series of spikes, which contained known quantities of the analytes, were included in each batch (usually 12 samples). The reporting limit was defined as five times the average values of the baseline noise signals and or three times the concentration in a representative blank. QC QA data are presented in Table 1. No contaminants were detected in reagent blanks.

Sediment calcium carbonate and organic carbon and seagrass lipid analyses

Sediment calcium carbonate content was determined by a weight loss gravimetric method (Blakemore *et al.*, 1987). For total organic carbon content (TOC) quantification in sediment samples, inorganic carbonates were first removed using an acid catalyzed digestion (10%

TABLE 1

Percentage recovery of spiked samples.

Compound	Subtidal sediments	Intertidal sediments	Intertidal seagrass
OCs	68–114	70–100	70–100
PCBs	nd[a]	90	80
Chlorpyrifos	77	95	85
Atrazine	53	70	95
Diuron	96	70	90
2,4-D	67	85	nd[a]

[a] nd: not determined.

HCl, 1% $FeCl_2$ at 70°C). The remaining material was dried and subjected to a combustion procedure (LECO induction furnace) with subsequent detection of CO_2 (LECO WR12 CO_2 detector). In order to determine the lipid content of the seagrass, the nonpolar extract after liquid–liquid partitioning was reduced to dryness and the lipid content determined gravimetrically.

Results

Intertidal sediments

Detectable concentrations (0.5–1.7 μg kg^{-1}) of diuron were present in three of the 16 sediment samples collected from intertidal locations. The occurrence of diuron in intertidal sediments was confined to samples collected in the wet tropics region south of the Daintree River between Cairns and Cardwell and in Moreton Bay (Table 2). Organochlorines, PCBs, chlorpyrifos, atrazine and 2,4-D were not detected in intertidal sediment samples.

Subtidal sediments

Atrazine, diuron, lindane, dieldrin, DDT and DDE were detected in a series of sediment samples collected from subtidal sampling sites (Table 3), with their occurrence being associated predominantly with sampling sites located in the wet tropics south of the Daintree River. Chlorpyrifos (detection limit 1.0 μg kg^{-1}), HCB (detection limit 0.05 μg kg^{-1}), heptachlor (detection limit 0.05 μg kg^{-1}), aldrin (detection limit 0.05 μg kg^{-1}), endosulfan (detection limit 0.05 μg kg^{-1}), DDD (detection limit 0.05 μg kg^{-1}) and PCBs (detection limit 50 μg kg^{-1}) were not detected in any of the subtidal sediment samples analysed in this study. Considering only the sites at which the respective pesticide was above the limit of quantification, concentrations of atrazine ranged from 0.1 to 0.3 μg kg^{-1}, diuron from 0.2 to 10.1 μg kg^{-1}, lindane from 0.08 to 0.19 μg kg^{-1}, dieldrin from 0.05 to 0.37 μg kg^{-1} and DDT and DDE from 0.05 to 0.26 μg kg^{-1}. No herbicides or insecticides were detected at sites north of the Daintree River (10 locations), whereas many samples collected further south (11 locations) had quantifiable levels of one or more contaminants. The exceptions were samples collected from the Hinchinbrook Channel, Bowling Green Bay, Cleveland Bay and Shoalwater Bay, where no herbicides or insecticides were detected.

Intertidal seagrasses

Diuron was the only contaminant detected in intertidal seagrass samples, where its concentration ranged from 0.8 to 1.7 μg kg^{-1} (Table 4). Its occurrence was confined to samples collected between Townsville and Cairns and in western Moreton Bay. Seagrass tissue concentrations of diuron were usually higher than sediment concentrations. Organochlorines, PCBs, chlorpyrifos and atrazine were not detected in intertidal seagrass samples (Table 4).

Discussion

Studies of the distribution of terrigenous biomarkers such as the lignin phenols derived from terrigenous vascular plants suggests that the bulk of terrigenous input from river runoff into Great Barrier Reef waters is confined to within 10 km of the Queensland coast (Johns *et al.*, 1988; Currie and Johns, 1989; Johnson, 1996). Organochlorine compounds tend to rapidly partition to fine sediments or be bio-accumulated into lipids in biota (Olsen *et al.*, 1982; Miyamoto *et al.*, 1990). As a consequence, estuarine and nearshore samples are likely to contain the highest concentrations of any contaminants resulting from anthropogenic activity in coastal catchments. The herbicides atrazine and diuron and the pesticides lindane, dieldrin and DDT (and its breakdown

TABLE 2

Pesticide concentrations, intertidal sediments 1997.[a]

Site	Site No.	OCs	PCBs	Chlorpyrifos	Diuron	Atrazine	2,4-D	%$CaCO_3$	% Organic carbon
Lloyd Bay	6	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	9.2	0.34
PCB	7	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	24.6	0.38
Bathurst Bay	8	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	32.3	0.76
Flinders Island	9	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	50.5	0.72
Low Isles	13	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	93.0	0.03
Cairns	15	< 1.0	< 50	< 1.0	0.5	< 0.5	< 10	3.6	0.30
Cardwell	19	< 1.0	< 50	< 1.0	1.7	< 0.5	< 10	1.4	2.2
Pallarenda	24	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	2.1	0.11
Cleveland Bay	25	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	4.2	0.29
Upstart Bay	28	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	0.3	0.10
Newry Bay	29	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	2.9	0.68
Shoalwater Bay	30	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	7.4	0.18
Gladstone	32	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	5.9	0.07
Hervey Bay	33	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	3.5	0.06
Moreton Bay East	34	< 1.0	< 50	< 1.0	< 0.5	< 0.5	< 10	< 0.1	0.26
Moreton Bay West	35	< 1.0	< 50	< 1.0	0.6	< 0.5	< 10	< 0.1	0.52

[a] All concentrations are μg kg^{-1} dry weight.

TABLE 3

Pesticide concentrations, subtidal sediments 1998.[a]

Site	Site No.	Depth (m)	Atrazine	Diuron	Lindane	Dieldrin	DDT	DDE	$CaCO_3$	Organic carbon
Horn Island	1	3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	70	0.8
		3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	68	0.7
Newcastle Bay	2	5	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	9	0.3
		6	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	4	0.3
Shelburne Bay	3	3.5	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	37	0.2
		3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	37	0.4
Temple Bay	4	4	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	6	0.4
		4	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	8	0.6
Weymouth Bay	5	3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	< 1	0.1
		4.5	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	< 1	na[b]
Lloyd Bay	6	4	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	17	1.5
		4	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	15	1.6
PCB	7	4.7	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	12	0.6
		3.8	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	13	0.6
Bathurst Bay	8	4.2	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	23	1.1
		4.2	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	28	0.9
Cape Flattery	10	3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	23	0.2
		4	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	10	0.1
Walker Bay	11	3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	12	1.3
		3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	17	1.0
Daintree River	12	na[b]	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	3	< 0.1
		na[b]	< 0.1	*0.2*	< 0.05	< 0.05	< 0.05	< 0.05	3	0.4
Barron River	14	3.6	< 0.1	*0.3*	< 0.05	< 0.05	< 0.05	*0.15*	5	0.6
		3.3	< 0.1	*0.4*	< 0.05	*0.09*	0.05	0.26	6	0.9
Russell River	16	5.1	< 0.1	*1.6*	< 0.05	< 0.05	< 0.05	< 0.05	5	1.2
		4.2	< 0.1	*0.5*	< 0.05	< 0.05	< 0.05	< 0.05	6	0.6
Johnstone River	17	3	< 0.1	*10.1*	0.08	0.15	< 0.05	*0.16*	3	2.5
		2.2	< 0.1	*9.8*	0.19	0.37	< 0.05	*0.25*	2	3.5
Tully River	18	4.3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	6	1.3
		2.8	< 0.1	*1.4*	< 0.05	< 0.05	< 0.05	*0.06*	4	1.2
Cardwell	19	3.4	< 0.1	*0.8*	< 0.05	< 0.05	< 0.05	< 0.05	7	1.7
		3.5	< 0.1	*0.8*	< 0.05	< 0.05	< 0.05	< 0.05	5	1.7
Hinchinbrook	20	4.8	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	4	0.6
		3.6	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	14	1.8
Herbert River	21	1.5	*0.1*	1.1	< 0.05	< 0.05	< 0.05	< 0.05	3	2.2
		1.5	*0.3*	2.8	< 0.05	< 0.05	< 0.05	< 0.05	4	3.4
Lucinda[c]	22	4.6	< 0.1	*1.6*	< 0.05	< 0.05	< 0.05	< 0.05	5	1.5
Halifax Bay	23	2.9	< 0.1	< 0.1	< 0.05	*0.05*	< 0.05	*0.05*	11	0.5
		2.6	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	4	0.7
Cleveland Bay	25	2.7	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	6	0.1
		2.7	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	11	0.2
B. Green Bay	26	2.3	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	7	0.8
		2.6	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	2	0.9
Burdekin River	27	8	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	*0.11*	4	0.6
		8.8	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	*0.1*	6	0.9
Shoalwater Bay	30	na[b]	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	< 1	0.2
		na[b]	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	9	0.2
Fitzroy River	31	na[b]	< 0.1	*0.9*	< 0.05	< 0.05	< 0.05	*0.10*	33	0.7
		na[b]	< 0.1	< 0.1	< 0.05	< 0.05	< 0.05	< 0.05	55	0.4

[a] All concentrations are μg kg^{-1} dry weight, organic carbon %, $CaCO_3$ %.
[b] na: not available.
[c] Only one site sampled.

product DDE) were all detected in nearshore marine samples collected along the Queensland coast.

Atrazine (2-chloro-4-ethylamino-6-isopropyl-amino-s-triazine), diuron (3,4-dichlorophenyl-1,1-dimethyl-urea) and 2,4-D (2,4-dichlorophenoxy) acetic acid) are currently the three most widely used herbicides for the pre- and post-emergence control of weeds in Queensland catchments. An estimated 331 t of atrazine, 197 t of diuron and 141 t of 2,4-D are applied annually to Queensland cane fields (Hamilton and Haydon, 1996). Of the three, only atrazine and diuron were detected in marine sediment or seagrass samples.

Atrazine was only detected in sediments collected in the vicinity of the mouth of the Herbert River (Fig. 1). Atrazine has a relatively high aqueous solubility (30–33 mg l^{-1} at 20°C) and only a moderate ability to adsorb onto soils (Huber, 1993; Hamilton and Haydon, 1996). As a consequence, an average of <3% (and up to 18%) of applied atrazine is lost to aqueous environments through runoff (Huber, 1993), where it can persist through freshwater and estuarine environments to contaminate marine ecosystems (Readman *et al.*, 1993). The low concentrations of atrazine detected in this study (0.1–0.3 μg kg^{-1}) are likely to be a consequence of

TABLE 4

Pesticide concentrations, intertidal seagrasses 1997.[a]

Site	Site No.	Species	OCs	PCBs	Chlorpyrifos	Diuron	Atrazine	% Lipid
Lloyd Bay	6	*H. uninervis*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.90
PCB	7	*H. uninervis*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	1.4
Bathurst Bay	8	*C. serrulata*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	na[b]
Flinders Island	9	*C. serrulata*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	1.6
Low Isles	13	*C. serrulata*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.40
Cairns	15	*Z. capricorni*	< 1.0	< 50	< 1.0	0.6	< 0.5	1.2
Cardwell	19	*H. uninervis*	< 1.0	< 50	< 1.0	1.1	< 0.5	0.90
Pallarenda	24	*H. uninervis*	< 1.0	< 50	< 1.0	0.8	< 0.5	0.40
Cleveland Bay	25	*C. serrulata*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.40
Upstart Bay	28	*Z. capricorni*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.90
Newry Bay	29	*Z. capricorni*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.30
Shoalwater Bay	30	*Z. capricorni*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.40
Gladstone	32	*Z. capricorni*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.30
Hervey Bay	33	*Z. capricorni*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.30
Moreton Bay East	34	*C. serrulata*	< 1.0	< 50	< 1.0	< 0.5	< 0.5	0.50
Moreton Bay West	35	*Z. capricorni*	< 1.0	< 50	< 1.0	1.7	< 0.5	0.40

[a] All concentrations are μg kg^{-1} dry weight.
[b] na: not available.

herbicide degradation rates being considerably enhanced by sunlight and saline conditions (Brambilla *et al.*, 1993), resulting in a half-life of generally less than 30 days in estuarine (tidal) environments (Huber, 1993). This is supported by microcosm trials, which demonstrated rapid (15–20 day half-life) breakdown of atrazine in estuarine sediments (Jones *et al.*, 1982). Rapid breakdown in tropical marine ecosystems together with the enhanced decomposition of atrazine under tropical soil conditions (herbicide half-life of 6–150 days (Obien and Green, 1969; Akinyemiju, 1991; Korpraditskul *et al.*, 1992; Korpraditskul *et al.*, 1993) imply that the Queensland tropical marine ecosystems should remain relatively uncontaminated by agriculturally applied atrazine.

In contrast to atrazine, diuron was widely distributed in marine sediments along the wet-tropics coastline (Townsville to Port Douglas, Fig. 1). It was detected in both subtidal and intertidal samples. Diuron is moderately mobile in soil (Lewis and Gardiner, 1996) and has a soil half-life of 100–300 days (Hamilton and Haydon, 1996; Lewis and Gardiner, 1996). It has an aquatic half-life of approximately 120 days and will degrade more rapidly in organically rich aquatic sediments (Howard, 1991). Photolysis is not a major degradation pathway for the herbicide (Lewis and Gardiner, 1996). The widespread occurrence of diuron in Queensland coastal sediments is therefore likely to be a consequence of high local agricultural usage combined with moderate soil mobility and a relatively long aquatic half-life.

The use of a majority of organochlorine compounds in Australia was banned by the 1990s (ANZEC, 1991; Hamdorf, 1992; Richardson, 1995). Information on historic usage in Australia (and Queensland) of organochlorine compounds such as DDT is limited (Hamdorf, 1992; Hunter, 1992; Voldner and Li, 1995), although it is estimated that up to 10 000 t of DDT was used in Australia for insect control prior to its ban in the 1970s (Voldner and Li, 1995; Mortimer, 1998). Figures for Australian sales or usage of cyclodiene organochlorines including dieldrin are not available (Hamdorf, 1992), although dieldrin had broad usage in Australia until the late 1980s as a termiticide and for the control of soldier fly (*Inopus* spp) in Queensland cane fields. Highest usage of dieldrin occurred in Queensland catchments south of the Burdekin River (Olafson, 1978). Lindane (γ-hexachlorocyclohexane) was also used extensively for the control of *Inopus* spp and cane grubs (*Lepidiota* spp) in Queensland catchments between the 1950s and the 1990s (Chessels *et al.*, 1988; Just *et al.*, 1990; Rayment *et al.*, 1997), with highest usage occurring in northern catchments (Olafson, 1978). Lindane, dieldrin and DDT and its metabolites were all detected in low concentrations in subtidal sediment samples.

Lindane was only detected in sediments from the vicinity of the mouth of the Johnstone River. Lindane has not been detected in water or riverine sediment samples collected in the Johnstone catchment in the 1990s (Hunter *et al.*, 1999). However, past monitoring has indicated that lindane was a widely distributed contaminant in northern Queensland with the pesticide detected in groundwaters (Brodie *et al.*, 1984), air and seawater samples (Tanabe *et al.*, 1982; Kurtz and Atlas, 1990), marine sediments (Dyall and Johns, 1985) and in freshwater and marine biota (Olafson, 1978; Kannan *et al.*, 1994; Kannan *et al.*, 1995; Russell *et al.*, 1996b). More recent monitoring has failed to detect the pesticide in freshwater and marine fish samples from northern Queensland (Russell *et al.*, 1996a; Von Westernhagen and Klumpp, 1995; Russell *et al.*, 1996b), although the pesticide is still detectable in northern Queensland agricultural soils and in sediments from irrigation drains (Müller *et al.*, 1999; Cavanagh *et al.*, 1999). Its limited distribution in nearshore sediments may, in part, be due to its relatively high vapour pressure and rapid volatili-

ization in tropical regions (Chessels *et al.*, 1988; Kannan *et al.*, 1995).

Dieldrin was detected in sediments collected from the mouth of both the Barron and Johnstone Rivers and in sediments from Halifax Bay. Dieldrin was a widely distributed contaminant of Queensland waterways and estuaries in the past (Clegg, 1974; Russell *et al.*, 1996a; Kannan *et al.*, 1995; Rayment *et al.*, 1997). It is still consistently detected in mud crabs (*Scylla serrata*) collected from estuaries adjacent to agricultural catchments between Moreton Bay and Cairns (Mortimer, 1999), although concentrations present in freshwater fish have declined by an order of magnitude between the 1970s and 1990s (Russell *et al.*, 1996b). It is also detectable in marine fish tissue (liver) collected from the central Queensland coast adjacent to agricultural activity (Von Westernhagen and Klumpp, 1995).

DDT and its metabolites were detected in low concentrations at the mouth of the Barron, Johnstone, Tully, Burdekin and Fitzroy Rivers and in Halifax Bay. Concentrations of DDE exceeded those of DDT at all sampling sites. Low concentrations of DDT and its metabolites have been detected in agricultural soils in the Herbert and Burdekin areas (Cavanagh *et al.*, 1999) and DDT has been consistently detected in mud crabs (*S. serrata*) collected from Queensland estuaries adjacent to agricultural catchments (Mortimer, 1999), although concentrations of DDT have declined in freshwater fish collected in northern Queensland waterways over the last 20 years (Russell *et al.*, 1996b).

Biological effects were not measured during this study. However, the concentrations of most detected pollutants were below the concentration believed to evoke a toxic response in marine benthic organisms (Table 5). The exceptions were dieldrin and diuron. These were detected in subtidal sediments at concentrations, which may present an environmental threat to the nearshore flora and fauna of the Great Barrier Reef region.

The herbicidal action of diuron is a consequence of inhibition of photosynthetic transport of electrons in photosystem II (Molander *et al.*, 1992). Diuron concentrations of 2, 10–170 and 10 μg l^{-1} have been shown to result in reduction in photosynthesis in marine periphyton (Molander *et al.*, 1992), reduction in growth in marine phytoplankton (Mayer, 1987) and inhibition of seagrass photosynthesis (Ralph, 2000), respectively. Predicted chronic water column diuron concentrations near the mouths of most wet tropics rivers range from 0.1 to 1.0 μg l^{-1} (Table 6) and concentrations are likely to be higher during monsoon rainfall periods which occur over the summer months (November–April). The potential to impact phytoplankton community structure and to inhibit photosynthesis and growth of seagrasses and coral zooxanthellae therefore exists. The influence of elevated water temperatures and reduced salinity during summer months on diuron toxicity to phytoplankton and seagrass is unknown.

Dieldrin is a cyclodiene pesticide with neurotoxic properties (Ware, 1989). Where dieldrin was detected, its

TABLE 5

Comparison of ER-L and ER-M concentrations and Great Barrier Reef (GBR) sediment pollutant concentrations.

Compound	GBR intertidal sediment range (μg kg^{-1})	GBR subtidal sediment range (μg kg^{-1})	ER-L[a] (μg kg^{-1})	ER-M[a] (μg kg^{-1})
Atrazine	< 0.5	< 0.1–0.3		
Diuron	< 0.5–1.7	*< 0.1–10.1*		
Lindane	< 1.0	0.19		
Dieldrin	< 1.0	*< 0.05–0.37*	*0.02*	8
DDT	< 1.0	0.05	1	7
DDE	< 1.0	0.26	2	15
Σ DDT	< 1.0	0.31	3	350

[a] Kennicutt et al. (1994).

TABLE 6

Potential diuron water column concentrations.

	Site No.	Diuron (μg kg^{-1})	Organic carbon (%)	Diuron C_{soc}[a]	Diuron K_{oc}[b]	Diuron C_w[c] (μg l^{-1})[d]
Barron R	14	0.4	0.9	44	398	0.1
Russell R	16	1.6	1.2	133	398	0.3
Johnstone R	17	10.1	2.5	404	398	1.0
Tully R	18	1.4	1.2	117	398	0.3
Cardwell	19	0.8	1.7	47	398	0.1
Herbert R	21	2.8	3.4	82	398	0.2
Lucinda	22	1.6	1.5	107	398	0.3
Fitzroy R	31	0.9	0.7	129	398	0.3

[a] C_{soc} – concentration in sediments expressed in terms of organic carbon.
[b] K_{oc} – partitioning coefficient between organic carbon and water.
[c] C_w – water concentration.
[d] $C_w = C_{soc} K_{oc}$ (Connell, 1990).

sediment concentration exceeded both the effects range low (ER-L) and effects range median (ER-M) for observed biological impacts to marine infauna (Long and Morgan, 1995). As a consequence, it may also present a localised threat to nearshore marine organisms along the wet tropics Queensland coast.

In conclusion, northern Queensland nearshore sediments are contaminated with a range of pesticides. Contamination is associated with intensive agricultural land use (primarily sugarcane production) carried out along the coast in high rainfall regions. In particular, marine sediment concentrations of diuron and dieldrin are high enough to result in ecosystem impacts.

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