NORTHERN TERRITORY GOVERNMENT

DEPARTMENT OF LAND RESOURCE MANAGEMENT

Pesticide Monitoring in the Douglas-Daly Region during the 2014 and 2015 Dry Seasons

Julia Schult

Aquatic Health Unit

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For further information contact:

Julia Schult

Department of Land Resource Management

PO Box 496, Palmerston, NT, 0831

Australia

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1. Summary

Seven sites in the Douglas-Daly and lower Daly region were investigated for pesticide and herbicide contamination in the dry seasons of 2014 and 2015, when flows in the streams are supplied by groundwater. Two sites were situated on the middle and lower Daly River, two sites on the upper and lower Douglas River, and one site each on Stray Creek, Hayes Creek and Green Ant Creek (Figure 1). Passive samplers were deployed in the streams for one month from August to September 2014 and September to October 2015 (lower Daly only) to detect trace levels of anthropogenic chemicals.

Pesticide and herbicide contamination of the streams was very low. Of 122 chemicals tested only 12 substances were found in the streams. Nine of these were herbicides and herbicide derivatives, one pesticide, the insect repellent DEET and a fragrance used in personal care products. Between 2 and 9 compounds were detected at any one site. The most commonly detected substances were herbicides of the triazine group (atrazine and simazine) that are highly water soluble and are known to have a high risk of leaching into groundwater. The herbicide atrazine or one of its break-down products was detected at all sites, while simazine was found at 5 of the seven sites.

The concentrations of all the detected chemicals were very low and none exceeded ANZECC guidelines for the protection of slightly to moderately disturbed ecosystems.

However, the detection of any pesticides and herbicides serves to highlight that the use of these chemicals should meet best practice to minimise environmental contamination.

2. Introduction

During the dry season, flows in the perennial streams of the Top End of the Northern Territory are supplied almost entirely from groundwater sources. When anthropogenic contaminants enter the groundwater, they can impact on river ecosystems. Monitoring and maintaining good groundwater quality is therefore important to protect the health of river environments.

The two main aquifers that underlie the Douglas-Daly region are the Tindall Limestone and Oolloo Dolostone aquifers. These karst aquifers support development in the Douglas-Daly region and supply most of the dry season flows in the area. The main land uses in the region are forestry, grazing and some horticulture, most of which use groundwater for irrigation, and stock and domestic use.

Groundwater is vulnerable to contamination when chemicals are applied to plants or soils. Water soluble substances in particular can filter through into deeper soil layers, carried by irrigation or rain water while other chemicals, including many pesticides, adhere to soil and particles and tend to remain in the top part of the soil profile. Direct contamination of bores can occur due to faulty fertigation or chemigation equipment, leading to unintentional backflow, or through runoff entering inadequately sealed bore heads. Other potential

sources of contamination include inadequately stored or dumped chemicals, industrial waste, faulty septic systems and runoff from fertilised fields, livestock or industrial areas.

Previous studies in the in the Darwin and Katherine regions have detected small amounts of herbicides and pesticides in springs and dry season stream flows. The results of these studies have been described in technical reports (Schult 2012 and 2014). These previous reports also provide additional background information on groundwater-fed streams, pesticide use and contamination pathways. This study is the third in a series of investigations to assess whether pesticides and herbicides are currently present in groundwater-fed streams of the Top End.

3. Methods

Seven sites were sampled in the Douglas-Daly region (

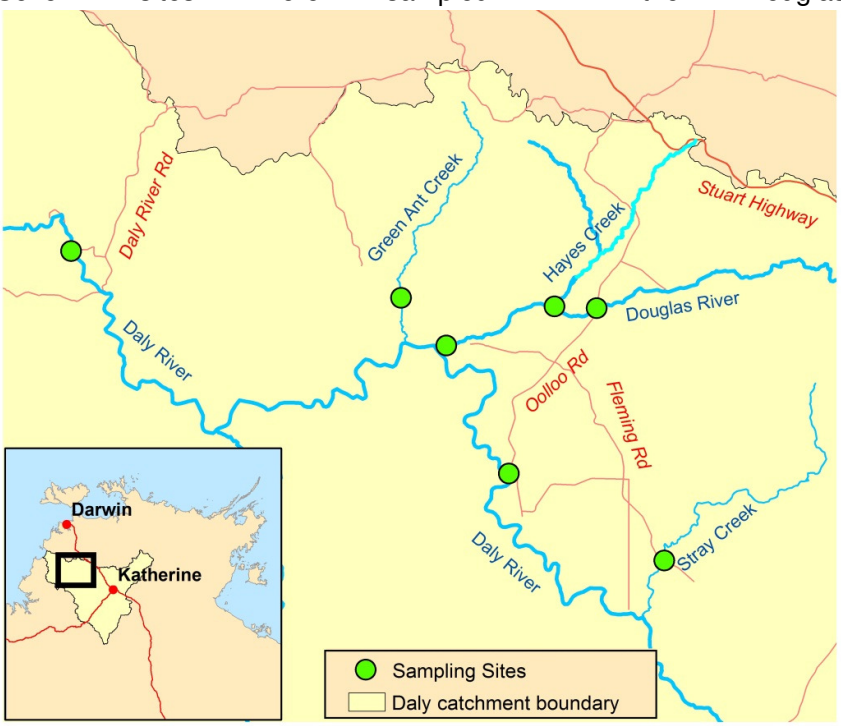

Figure 1) during the 2014 and 2015 dry seasons. With the exception of Site 5, sites were chosen to represent all the major perennial streams in the area that receive water from the Tindall and Oolloo karst aquifers. Dry season flows at Site 5, in the upper middle reaches of the Douglas River are predominantly sourced from a cretaceous sandstone aquifer but are also receiving some water from the karst aquifers.

Passive samplers were used to detect herbicide or pesticide contamination. These samplers can detect very low concentrations of chemicals that are not detectable with conventional grab sampling, by accumulating chemicals over a period of several weeks. The analysis provides an average concentration for the deployment period. Two different types of samplers were used to detect different classes of chemicals. The *Empore Disc* sampler (ED) accumulates hydrophilic (water soluble) substances, the second device, a

polydimethylsiloxane sampler (PDMS), attracts hydrophobic (water repellent) substances. Most herbicides belong to the former, most pesticides to the latter class.

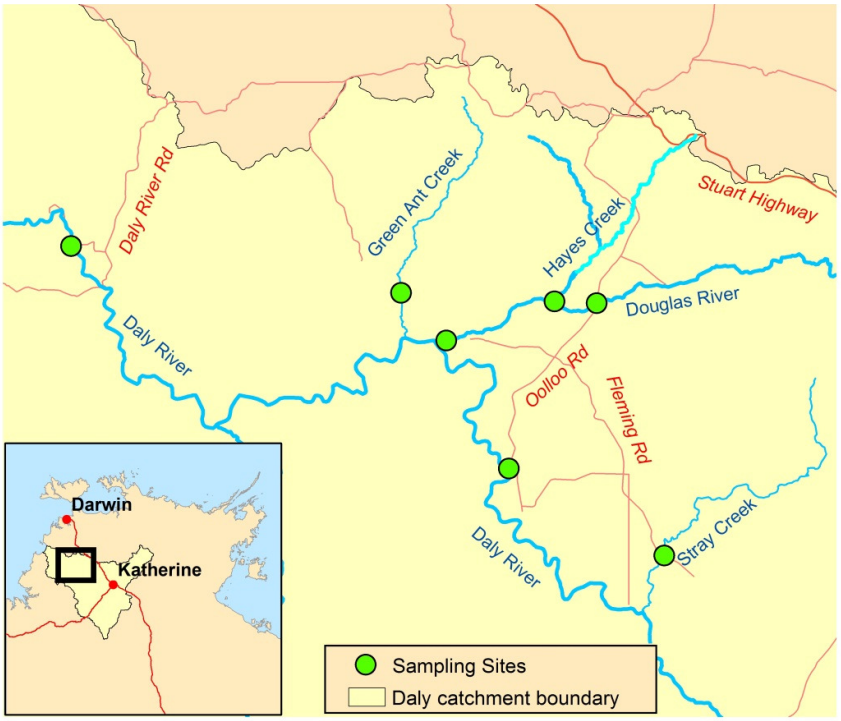

Figure 1.Location of sampling sites in the Douglas-Daly region

Samplers in the Douglas-Daly region were deployed for four weeks from mid-August to mid-September 2014 (Table 1), the sampler at the lower Daly River from early July to early August 2015. Depending on the water depth at the site, the samplers were either suspended from a rope where the water was deep enough, or attached to bricks or a weighted steel frame in shallower sites. Upon retrieval, the passive samplers were stored on ice and air-freighted to the laboratory for extraction and analysis to the National Research Centre for Environmental Toxicology (Entox, University of Queensland, Brisbane). The University of Queensland's standard suite of 122 chemicals were tested for. The full list of analytes is provided in Appendix A.

Table 1. Passive sampler locations and deployment dates.

Site No	Site	Latitude	Longitude	Date in	Date out
1	Daly River Esplanade (Site 2)	-14.0037	131.2390	11/8/14	8/9/14
2	Green Ant Creek	-13.7750	131.0989	13/8/14	9/9/14
3	Hayes Ck u/s of Douglas R. confluence	-13.7862	131.2983	12/8/14	9/9/14
4	Stray Creek	-14.1166	131.4413	12/8/14	8/9/14
5	Douglas River @ Oolloo Rd	-13.7885	131.3533	12/8/14	8/9/14
6	Douglas River @ Tipperary Crossing	-13.8369	131.1577	11/8/14	8/9/14
7	Lower Daly River (Banyan Farm)	-13.71417	130.66947	7/7/15	5/8/15

At the time of deployment and retrieval of the passive samplers, water quality measurements of temperature, electrical conductivity (EC), pH and dissolved oxygen (DO) were collected in-stream using a Quanta multi-parameter probe. Turbidity was measured with a Hach 2100Q turbidimeter, and water samples were collected for analysis of total nitrogen (TN) and phosphorus (TP), soluble nutrients (nitrite (NO_2), nitrate (NO_3), ammonia (NH_3) and filterable reactive phosphorus (FRP)) and chlorophyll a. Soluble nutrient samples were filtered through a 0.45 µm glass fibre filter in the field.

All samples were stored on ice during transport and frozen (nutrient samples) or refrigerated (major ions) upon return to the laboratory until further analysis at the Northern Territory Environmental Laboratories (Intertek NTEL). Analytical methods for nutrient analyses are listed in Appendix B.

4. Results

Herbicides and Pesticides

Of the 122 chemicals tested, 12 were detected in this study: seven herbicides, one pesticide, one insect repellent and one fragrance used in personal care products (Table 2). The herbicide *atrazine* and/or its breakdown products (*desisopropyl atrazine* and *desethyl atrazine*) were detected at all sites. The other four herbicides that were found (*tebuthiuron*, *simazine, metolachlor* and *ametryn*) occurred at 1 to 5 of the seven sites. The pesticide *chlorpyrifos* was found only at Site 5, the Douglas River at Oolloo Rd crossing, while the insect repellent DEET occurred at two sites (sites 5 and 7).

All of the estimated concentrations were very low and, where guideline values were available, did not exceed the ANZECC guideline values for 95% ecosystem protection, the level recommended for slightly to moderately disturbed ecosystems (ANZECC & ARMCANZ 2000), at any of the sites (Table 2). The ANZECC guidelines specify trigger values for a number of chemicals that are potentially toxic to aquatic organisms and recommend further actions if guideline values are exceeded.

Table 2. Estimated concentrations (ng/L) of pesticides and herbicides detected in Douglas-Daly region streams

	ANZECC Guideline 95% protection	Daly River Esplanade	Douglas R. at Oolloo Rd	Green Ant Creek	Hayes Ck	Stray Ck	Tipperary Crossing	Lower Daly
Atrazine	13,000	**0.74**	ND	**1.2**	**0.18**	ND	**0.155**	**0.22**
Atrazine Desisopropyl	NL	ND	ND	**0.72**	**11**	ND	**2.35**	**0.25**
Atrazine Desethyl	NL	**1.3**	**0.53**	**8.2**	**13**	**0.63**	**2.8**	**0.85**
Tebuthiuron	2,200	**0.16**	ND	ND	ND	**0.24**	ND	**0.05**
Ametryn	NL	ND	ND	**0.11**	**0.052**	ND	ND	ND
Simazine	3,200	**0.1**	**0.21**	ND	**2.1**	ND	**0.74**	**0.09**
Metolachlor	ID	ND	ND	**0.033**	ND	ND	ND	ND
Chlorpyrifos	10	ND	**0.30**	ND	ND	ND	ND	ND
DEET	NL	ND	**36**	ND	ND	ND	ND	**39**
Diuron	ID	ND	ND	ND	ND	ND	ND	**0.17**
Bromacil	ID	ND	ND	ND	ND	ND	ND	**0.11**
Tonalid	NL	ND	ND	ND	ND	ND	ND	**0.03**

ID: insufficient data; NL: not listed in ANZECC guidelines; ND: not detected

General water quality

The general water quality of the sites was within expected ranges for the Douglas Daly region. The full range of field measurements and nutrient water quality data is provided in Appendix C.

Electrical conductivities ranged from 280-300 at Site 5 (Douglas River at Oolloo Rd) and from 500-700 at the other sites. Electrical conductivity can be used as an indicator for the groundwater origin of stream flows. The lower conductivity at Site 5 indicates that a larger proportion of the flow originates from low conductivity sandstone groundwater (estimated to be about 50%). All other sites had flows that were of predominately karst origin.

5. Discussion

Pesticides and herbicides are widely used in the community by gardeners, for domestic pest control, in agriculture and to control weeds and pests around commercial buildings, infrastructure and roadsides.

The chemicals that were detected in this study are commonly used pesticides, herbicides and their break-down products. Most of them are known to have a high risk of groundwater contamination due to their high water solubility (e.g. Infopest (2014), APVMA (2008), Waugh and Padovan (2004)). Table 3 contains a summary of the characteristics of the substances that were detected in the Douglas Daly region.

Table 3. Description of chemicals detected in Douglas-Daly region streams

Chemical name	Description	Examples of trade names
Ametryn	Triazine herbicide. Mainly used to control grasses and broadleaf weeds in sugarcane and pineapples.	Crop care Farmoz
Atrazine	Atrazine is one of the main herbicides used in Australia. Used before and after planting of crops to control broad-leaved weeds and grasses in crops such as sorghum, sugar cane, maize and canola. Also used in turf and non-agricultural sites such as lawns, industrial areas, rights-of-way and in orchards (APVMA 2008, US EPA 2014). Atrazine is a common contaminant of surface and groundwater in Eastern Australia (Shaw and Muller 2005) and has been banned in Europe after persistent contamination of groundwater was found (EC 2014)	Farmozine Nutrazine Gesaprim
Atrazine Desethyl	Breakdown product of atrazine	N/A
Atrazine Desisopropyl	Breakdown product of atrazine	N/A
Bromacil	Herbicide for the control of grasses and other weeds. Registered for use on farm buildings and commercial land, fenceline weed control and rights of way as well as some crops, including citrus. High risk of leaching	Bromacil Krovar Uragan
Chlorpyrifos	Commonly used organochlorine pesticide for termite control and the control of other insect pests on fruits, vegetables and other crops. Bioaccumulation can occur (Infopest 2014).	Lorsban Strikeout
DEET	A common personal insect repellent found in most tropical strength repellent products.	Bushman's Rid Aeroguard
Diuron	A general herbicide often used on hard surfaces. Breaks down very slowly in water. Can be toxic to fish and invertebrates. High risk of leaching.	Diuron Diurex
Metolachlor	Used for the control of grasses and broadleaf weeds in sorghum, maize, sugar cane and improved pastures.	Nufarm Farmoz
Simazine	Systemic triazine herbicide. Absorbed through roots. Stable in water, decomposed by UV light, binds to soil. Controls broad-leaf weeds in a variety of crops and at higher rates of application, grasses and broad-leaved weeds in other areas. Used in citrus and for non-crop weed control on roads, railways etc. High risk of groundwater leaching.	Accensi
Tebuthiuron	A general herbicide that is commonly used to control weeds. It is slightly toxic to aquatic vertebrates and invertebrates at higher concentrations but has little potential to accumulate in the environment.	Farmalinx Graslan Tebulan
Tonalide	A musk-fragrance commonly used in personal care products, laundry detergents and cosmetics	

The traces of herbicides and pesticides found in this study were very low and do not pose a risk to the aquatic ecosystems they were found in. Had samples been collected by conventional grab sampling, rather than the passive sampler accumulation technique, the concentrations would have been mostly undetectable.

Previous studies of groundwater-fed ecosystems by this department in the Darwin and Katherine region detected a similar suite of chemicals dominated by highly water soluble herbicides (Schult 2012 and 2014). Table 4 and Figure 2 provide a summary of results of the two previous studies and the current study.

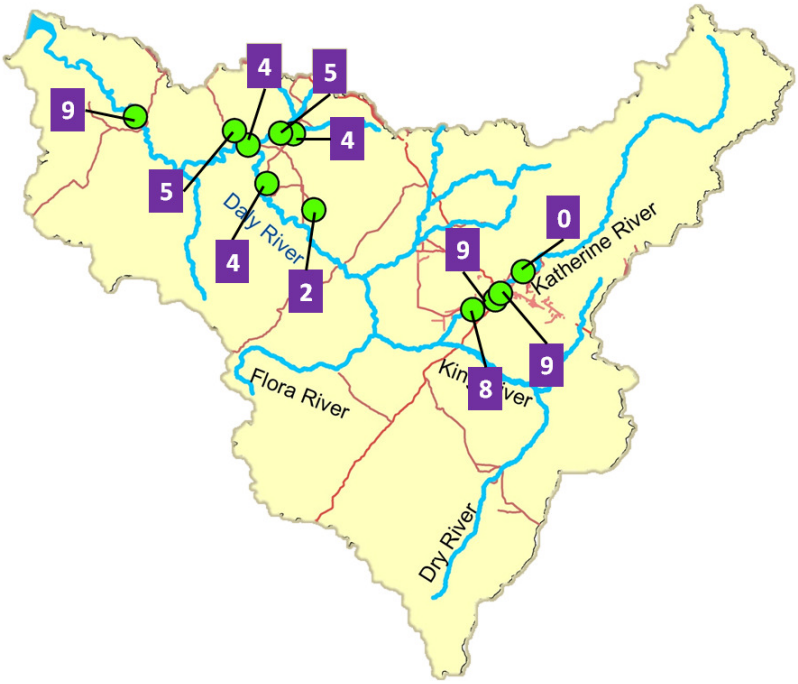

Figure 2. Overview of the number of pesticides and herbicides found at different sites in the Daly River catchment. Sampling took place during the dry season in 2011 and 2014/15.

Table 4. Summary of pesticide and herbicide detection in springs and streams of the Top End 2011-2015.

Chemical name	ANZECC Guideline Value** (ng/L)	Katherine Region (2011)				Darwin Region Springs (2014)					Douglas-Daly Region (2014/15)						
		Donkey Camp Pool inflow	Katherine R near Northbank spring	Katherine R near Springvale Spring	Galloping Jack's	Howard Springs	Whitewood Jungle	Melacca Spring	Berry Springs u/s swimming area	Palm Creek	Daly River Esplanade	Douglas River at Oolloo Rd	Green Ant Ck	Hayes Ck	Stray Ck	Douglsa River at Tipperary Crossing	Lower Daly River
3,4-dichloroaniline	3,000	-	-	-	-	-	**0.06***	-	-	-	-	-	-	-	-	-	-
Ametryn	NL	-	-	-	-	-	-	-	-	-	-	-	**0.11**	**0.052**	-	-	-
Atrazine	13,000	-	**0.39**	**0.18**	**0.35**	-	-	-	-	**0.07***	**0.74**	-	**1.2**	**0.18**	-	**0.155**	**0.22**
Atrazine desethyl	NL	-	**1.48**	**1.10**	**0.65**	-	-	-	-	-	**1.3**	**0.53**	**8.2**	**13**	**0.63**	**2.8**	**0.85**
Atrazine Desisopropyl	NL	-	-	-	-	-	-	-	-	-	-	-	**0.72**	**11**	-	**2.35**	**0.25**
Bromacil	ID	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	**0.11**
Chlorpyrifos	10	-	**0.5**	**0.6**	-	-	-	-	**0.7**	-	-	**0.30**	-	-	-	-	-
DEET	NL	**189**	**79**	**93**	**103**	-	-	-	-	-	-	**36**	-	-	-	-	**39**
Diuron	ID	-	-	-	-	-	**0.10***	-	-	**0.13***	-	-	-	-	-	-	**0.17**
Galaxolide	NL	-	-	**21.1**	**22.3**	-	-	-	-	-	-	-	-	-	-	-	-
Hexazinone	ID	-	**0.07**	**0.15**	**0.10**	-	-	-	-	-	-	-	-	-	-	-	-
Imidacloprid	NL	-	-	-	-	-	-	-	-	**0.21***	-	-	-	-	-	-	-
Methamidaphos	NL	-	**42.0**	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Metolachlor	ID	-	-	-	-	-	-	-	-	-	-	-	**0.033**	-	-	-	-
Simazine	3,200	-	**0.11**	**0.07**	**0.10**	-	-	-	-	-	**0.1**	**0.21**	-	**2.1**	-	**0.74**	**0.09**
Tebuthiuron	2,200	-	**0.38**	**0.30**	**0.24**	-	-	-	-	**1.1**	**0.16**	-	-	-	**0.24**	-	**0.05**
Tonalide	NL	-	**3.1**	**6.2**	**3.1**	-	-	-	-	-	-	-	-	-	-	-	**0.03**

ID: insufficient data
NL: not listed in ANZECC guidelines
" – " : Not detected

It is difficult to determine whether contamination found today originates from current or historical land use practices. Most of the herbicides found in streams in this and other studies have been registered for use in the NT for more than 40 years (Stuart Smith, DPIF, pers comm) and are still widely used today. The only insecticide that was detected, c*hlorpyrifos,* is an organophosphorus pesticide that is not usually considered to have a high risk of leaching into groundwater. However, it is moderately persistent in soil. Chlorpyrifos was used extensively for domestic termite control for 10 years after 1995, after other termidicides were banned (Stuart Smith p.c.) but is now mainly used to control pests in fruit and vegetable crops.

Groundwater consists of waters that are a mix of ages, so that the exact age of the groundwater, and therefore the age of the contamination, is difficult to determine. The time it takes for groundwater recharge to reach the streams can vary from a few days to many years or decades, depending on the location and pathways of groundwater flows (Karp 2005, Steven Tickell DLRM pers.comm.)

This sampling program was not designed to detect the sources of contamination for pesticides. More extensive testing of groundwater directly from domestic and production bores in the region would be required to provide a more comprehensive picture of groundwater quality.

However, the detection of any pesticides and herbicides in our waterways serves to highlight that the use of these chemicals should meet best practice to minimise environmental contamination.

6. Conclusion

This study provides baseline information for pesticide and herbicide contamination of streams in the Douglas-Daly region, and can be used for comparison with future monitoring results to assess long-term contaminate trends.

The concentrations of all the detected chemicals (12/122) were very low and none exceeded the ANZECC environmental guidelines for the protection of slightly to moderately disturbed ecosystems.

The detection of any pesticides and herbicides in our waterways serves to highlight that the use of these chemicals should meet best practice to minimise environmental contamination.

7. References

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Appendix A: List of analytes for passive samplers

Table 5. List of hydrophobic compounds tested

Is 1	Terbuphos	Fipronil	Propiconazol isomer
Methomyl	Diazinon	Isophenophos	Endosulfan sulphate
Tep	N-butyl benzene sulfonamide	Chlorfenvinphos e+z isomers	Ddt p,p
Oxydemeton methyl	Tcpp isomers	Endosulfan lactone	Hexazinone
Sur1 2-nitro-m-xylene	Phosphamidon peak1 **200**	Allethrin	Dicofol o,p
Is 2	Is4	Triadimenol isomers	Diclofop methyl
Methamidophos	Anthracene-d10	Oxychlor	Propagite
Dichlorvos	Chlorothalonil	Heptachlor epoxide	Tebuconazole
Diuron bd	Disulfoton	Fenthion ethyl	Haloxyfop, 2-ethoxyethyl
Nicotine	Chlordene	Thiabendazole	Sur4 triphenylphosphate
1-methyl-1h-benzotriazole	Etrimiphos	Furalaxyl	Piperonyl butoxide
4-chloro-3,5-dimethylphenol (dettol)	Triallate	Procymidone	Bioresmethrin
Mevinphos z+E	HCH-d	1-hydroxy-2,3-epoxychlordene	Iprodione
Acephate	Pirimicarb	Captan	Tetramethrin isomers
3,4-dichloroaniline	Galaxolide	Methoprene	Bifenthrin
2,6-di-t-butylphenol	Musk xylene	Haloxyfop methyl	Phosmet
1h-benzotriazole	Tonalid	Bromophos ethyl	Methoxychlor
Is3	Phosphamidon peak2 **800**	Methidathion	Is5
2,4-di-t-butylphenol	Endosulfan ether	Triclosan	Dicofol p,p
2,6-di-t-butyl-p-cresol (bht)	Propanil	Tetrachlorvinphos	Phenothrin isomers
Demeton-o-methyl	Chlorpyrifos me	Dde o,p	Tetradifon
Tebuthiuron	Metribuzin	Chlordane trans	Cyhalothrin isomers
Molinate	Vinclozalin	Triclosan methyl ether	Azinphos methyl
5-methyl-1h-benzotriazole	Transfluthrin	Chlordane cis	Amitraz
DEET	Parathion methyl	Endosulfan alpha	Pyrazaphos
Omethoate	n-butyltoluenesulfonamide	Fenamiphos	Azinphos ethyl
Propoxur	Metalaxyl	Sur3 pyrene -d10	Bitertanol isomers
Demeton-s-methyl	Ametryn	Nonachlor trans	Permethrin isomers
Fluometuron	Carbaryl	Prothiophos	Coumaphos
Phosphate TRI-n-butyl	Prometryn	Bisphenol A	Cyfluthrin isomers
Ethoprop	Fenchlorphos	Profenophos	Cypermethrin isomers
Desisopropylatrazine	Heptachlor	Oxadiazon	Sur5 decachlorobiphenyl
Trifluralin	Pirimiphos methyl	Dde pp	Is6
Desethylatrazine	Chlordene epoxide	Oxyfluorfen	Fenvalerate isomers
Bendiocarb	Terbutryn	Ddd o,p	Fluvalinate isomers
Monocrotophos	Fenitrothion	Fluazifop butyl	Deltamethrin isomers
Cadusaphos	Bromacil	Dieldrin	Dimethomorph e,z isomers
Phorate	Malathion	Endrin	Temephos
HCH-a	Chlorpyrifos oxon	Ethion	Rotenone
HCB	Musk ketone	Nonachlor cis	Dicofol o,p bd
Demeton-s	Metolachlor	Endosulfan beta	Flutriafol
Dimethoate	Chlorpyrifos	DDD p,p	Icaridin
Simazine	Fenthion methyl	DDT o,p	Praziquantel
Atrazine	Aldrin	Moclobemide	Benzenesulfonanilide
Propazine	Parathion ethyl	Endrin aldehyde	2-benzyl-4-chlorophenol
TCEP	Triadimefon	Sulprofos	Quintozene
HCH-b	Chlordene, 1-hydroxy	TDCPP isomers	Terbuthylazine
Dioxathion	Dicofol p,p bd	Famphur	Pendimethalin
Lindane (HCH-g)	Sur2 dibromobiphenyl	Benalaxyl	Carbophenothion

Table 6. List of hydrophilic compounds tested

Desisopropyl Atrazine	Terbutryn	Trifluoxysulfuron
Imidacloprid	Flumeturon	Imazethapyr
Desethyl Atrazine	Atrazine	Metsulfuron-Methyl
Tebuthiuron	Diuron	Clopyralid
Ametryn	Metolachlor	Picloram
Hexazinone	3,4 Di Cl Analine	Terbuthylazine
Bromacil	Metribuzin	Terbuthylazine desethyl
Simazine	Imazapic	
Prometryn	Asulam	

Appendix B: Analytical methods and APHA standard method numbers

Parameter	Method	APHA (1998) number
NO_3/NO_2	Automated cadmium reduction method	4500-NO_3-F
NH_3	Automated Phenate method	4500-NH_3 F
Total N	Persulfate method	4500-N C
Filterable reactive P	Flow injection analysis for orthophosphate	4500-P F (B1)
Total Phosphorus	Flow injection analysis for orthophosphate	4500-P F (B3)
Silica (SiO_2)	Flow injection analysis	4500-SiO2 F

Appendix C: Water Quality Data

Site	Daly River Esplanade (Site 2)		Douglas River @ Oolloo Rd		Green Ant Creek		Hayes Ck u/s of Douglas R. Confluence		Stray Creek		Tipperary Crossing	Lower Daly
Date	11/08/14	8/09/14	12/08/14	8/09/14	13/08/14	9/09/14	12/08/14	9/09/14	12/08/14	8/09/14	8/09/14	7/7/15
Temp (^{o}C)	26.6	27.8	26.0	28.3	22.6	24.5	22.5	24.0	22.7	26.3	27.8	24.9
EC (µS/cm)	672	638	300	283	569	547	625	592	598	562	508	556
pH	7.8	7.9	7.0	7.2	7.9	8.0	7.8	7.9	8.0	8.1	8.0	8.5
DO (mg/L)	8.05	8.05	5.24	5.72	7.09	7.16	7.84	7.82	8.5	8.3	7.8	8.06
DO (%)	101	103	65	74	84	86	91	30	99	104	100	98
Turbidity (NTU)	NS	1.68	1.98	1.84	2.51	2.00	1.56	1.24	0.97	1.01	1.81	4.1
NO_2_N (µg/L)	3	<1	4	3	4	5	3	5	<1	4	<1	NS
NO_3_N (µg/L)	26	20	21	13	13	7	22	22	4	4	57	NS
NH_3_N (µg/L)	<1	1	5	1	<1	2	<1	5	5	2	4	NS
FRP (µg/L)	8	9	7	6	7	5	6	7	6	6	5	NS
Total N (µg/L)	80	80	80	80	110	140	80	80	60	70	110	NS
Total P (µg/L)	23	8	13	16	10	17	15	16	17	16	13	NS
CHL-a (µg/L)	0.46	0.55	0.52	1.25	2.28	2.22	0.96	0.64	0.4	0.27	0.66	NS

NS: not sampled