

# **Reducing pesticide exposure risk on floodplain cotton farms by enhancing natural removal processes.**

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## **Abstract**

Concentrations of pesticide residues in floodplain cotton farm runoff are currently high enough for concern. Routine monitoring over 3 cotton seasons and subsequent risk analysis demonstrated the potential for toxicities to aquatic species. The insecticide endosulfan and the herbicide fluometuron were selected as targets for accelerated removal from cotton tailwater in field trials. Aquatic plants were shown to increase sedimentation and significantly reduce the first-order, total endosulfan half-life from 1.35 to 0.77 days. Basalt gravel filter beds were also beneficial, removing up to 41% of fluometuron and 26% of total endosulfan over a 20 m distance during peak mass loading periods. Manipulation of farm design is also discussed as an option for enhancing pesticide removal processes.

## **Keywords**

Cotton farm; insecticide; herbicide; artificial wetland; risk; sedimentation

## INTRODUCTION

The ecological importance of river floodplains and associated wetlands in inland Australia has been well documented. Briefly, these areas support diverse populations of mammal, reptile, amphibian, invertebrate and plant species (Smith *et al.*, 1998; Brock, 2003). They are especially important for the breeding of native fish and migratory birds (Gehrke and Harris, 2000; Kingsford and Auld, 2005). A number of floodplain wetlands are protected under the Ramsar convention. As well as providing habitat, wetlands also provide a number of indirect (non-habitat) ecosystem services such as groundwater recharge, prevention of soil erosion, climate regulation and filtration and polishing of surface water (Finlayson and Rea, 1999).

Despite their ecological significance, the inland river floodplains of New South Wales and Queensland have been successively developed and modified since non-indigenous settlement, most noticeably since the damming of numerous headwaters in the 1950's and 1960's. Advances in machinery and pest management around the same time led to the establishment of the Australian cotton industry, which now dominates agricultural enterprises along most the inland rivers of central and northern New South Wales and central and southern Queensland. Cotton production is relatively intensive and involves considerable earthworks for irrigation purposes and fertilizer and pesticide application for high quality and yields. With regard to the structure and function of the natural ecosystem, this has led to more regulated river systems, loss of habitat for native species and a greater incidence of pollutant exposure (Arthington and Pusey, 2003).

Nevertheless, the Australian cotton industry has demonstrated commendable effort in promoting environmental care and stewardship over the last decade. The flagship has been the best management practice (BMP) program and manual (Williams and Williams, 2000), which outline the current best knowledge and recommendations for improved environmental outcomes. Documented results have included increases in water use efficiency (Goyne and McIntyre, 2003), a shift toward integrated pest management and less reliance on pesticides (Fitt, 2000) and the management of off-farm pesticide movement (Kennedy *et al.*, 2003). The last two steps have been reflected in decreasing concentration and incidence of detection of cotton chemicals in river waters (Muschal, 2001).

There is now a real industry desire to enhance the agricultural floodplain for environmental outcomes, including habitat improvement for native species and encouraging on-farm biodiversity (Reid *et al.*, 2003; CRDC, 2005). This is set to continue with the establishment of a cooperative research centre for cotton catchment communities. However, for successful implementation of research findings without detriment to native fauna and flora, pesticide residue concentrations must now be reduced *on-farm*.

Pesticides naturally dissipate in water bodies through a combination of transport and degradation pathways (Stangroom, 2000). The rate and extent of these varies from pesticide to pesticide depending heavily on their physicochemical properties. From an environmental protection perspective, our goal is to identify and accelerate the major

removal pathways, thus reducing pesticide concentrations as rapidly as possible and limiting exposure.

The aim of this project was to accelerate the removal of pesticide residues from runoff water, which was deemed to be a high risk exposure route to native species in previous work. Specifically, we aimed to use natural processes rather than more industrial remediation techniques, due to the perceived lower cost, greater flexibility in response to change and ease of integration with current farming practices. This paper documents the nature and concentration of pesticide residues in tailwater on several representative cotton farm, the environmental risk of these residues and the potential for risk reduction using phytoremediation and (bio)filtration. The insecticide endosulfan and the herbicide fluometuron are used as illustrative examples.

## **METHODS**

### **Pesticide Analysis**

Time composite runoff water samples were collected from the taildrain exit following the irrigation of several cotton fields throughout the summer growing season in 2002, 2003 and 2005 in the Namoi Valley, NSW. Analyses were performed on pesticides applied to the field. Samples were extracted by liquid/liquid partitioning or solid phase extraction and analysed by a combination of HPLC/UV and GC/ECD (Baskaran and Kennedy, 1999; Crossan, 2002). Curve fitting to pesticide data was conducted using Origin 4.10 (Microcal, Northampton, MA).

### **Site Descriptions**

Two artificial treatment wetlands were constructed adjacent to cotton fields and receiving irrigation tailwater (Figure 1). The first system was ponded and consisted of an open and vegetated pond in series. It was designed to assess the contribution of aquatic plants to pesticide dissipation compared to open water. Samples were taken at the inlet initially during filling from an irrigation event, and then from both ponds every two days for 10 days. The second system was flow-through, consisting of two open channels and three sub-surface flow (SSF) gravel bed channels (basalt gravel, 12 mm mean size). It was designed to evaluate pesticide reduction by filtration and sorption processes. Time composite samples were taken over five hours from the inlet, middle and outlet of each channel. The nominal residence time of water was 5 h for the open channels and 4 h for the gravel beds. The nominal channel volumes were 50 m<sup>3</sup> for the open channels and 21 m<sup>3</sup> for the gravel beds. Both systems were used to treat one bed volume during three irrigations throughout one growing season.

INSERT FIGURE 1

### **Ecological Risk Analysis**

A Tier 1 Environmental Risk Assessment (ERA) was used to determine the risks associated with pesticide residues detected in water (Norton *et al.*, 1992). The aim of an ERA is to assess risk of adverse effects occurring to species that are potentially exposed

to pollutants (Solomon *et al.*, 2000). The ERA was carried out following the recognised framework (Norton *et al.*, 1992).

As a measure of ‘sustainability of practice’ we have included lowest observed effect concentrations (LOEC) for toxicity endpoints. Because of the different nature of action between the two pesticides we selected plant species toxicity data to assess fluometuron and animal species data to assess endosulfan. For quotients greater than one ( $Q > 1$ ) we infer that these concentrations are likely to have some effect on the ecosystem. A quotient less than one ( $0.01 < Q < 1$ ) indicates that the practice is reasonably sustainable, provided conditions do not significantly change. For quotients much less than 1 ( $Q < 0.01$ ) this indicates that the risk is very low and can easily absorb an order of magnitude change in conditions or use.

## RESULTS AND DISCUSSION

### Residues in Tailwater

Maximum concentrations of pesticides in the Namoi valley (Table 1) were similar to those found on other cotton farms in Australia (Silburn, 2003). These levels are approximately 5-10 times the corresponding maximum concentrations found in water of rivers in Australian cotton growing areas (Muschal and Warne, 2003). Our data reflects the general trend in pesticide use in the Namoi valley, which increased in the summer 2005 season because of high early insect pressure compared to the 2002 and 2003 seasons. Although insect pressure is difficult to predict prior to the growing season, it was speculated that the wet start to 2005 season contributed to the increased pressure. This rainfall occurred soon after fluometuron application, resulting in an initially high runoff loss. The amounts of fluometuron measured in irrigations later in that season were thus much lower.

INSERT TABLE 1

### Ecological risk of tailwater

Although it is important to quantify the concentration of chemical residues, this information is of limited use until some judgment of severity is made. We chose to conduct a first-tier risk assessment to put perspective on the pesticide concentrations detected (Table 1). The lack of readily available toxicological data for Australian aquatic species prevented in-depth or comparative analysis, as is often reported in the literature (Hose and Van den Brink, 2004).

Environmental risk occurs whenever there is exposure of organisms to chemicals. Because the pesticide residues have been detected in farm water, concentrations were used directly as environmental exposures. These values provided a realistic temporal analysis of risk, as monitoring was conducted routinely throughout the cotton growing season. Whilst residue levels should also be monitored across numerous farms and regions to determine spatial exposure, we consider these data sufficient for our case study.

Our assessment demonstrated that there is a real risk of endosulfan toxicity to aquatic species, with hazard quotients exceeding unity by an order of magnitude for both chronic and acute exposure. Other recent reports also highlight the potential hazard of endosulfan residues (Muschal and Warne, 2003). Conversely, herbicides, because of their mode of action, did not show an acute ecological risk on our test species. Of greater concern is their potential for adverse *chronic* effects over time. A more in-depth assessment incorporating probabilistic approaches (Solomon *et al.*, 2000) is necessary for future work. Nevertheless, it can be seen that remediation efforts should focus on decreasing high pulse concentrations of endosulfan and minimizing chronic exposure to herbicides, particularly diuron.

### **Pesticide removal in ponded system**

The three irrigations monitored during this season occurred approximately 45 days after the heavy rainfall at the start of the season, resulting in lower-than-usual concentrations of fluometuron entering the ponded system. Fluometuron was only quantifiable in the first monitored irrigation. Dissipation was best described by a zero-order *decrease* in the vegetated pond, compared with an apparent *increase* in the open pond (Figure 2). Since the wetland passively receives irrigation runoff during non-monitoring times, it is likely that the wetland acted as a sink for the large concentrations of fluometuron in tailwater at the start of the season. It is therefore possible that upon receiving dilute concentrations during the monitoring period, higher concentrations of weakly-sorbed fluometuron have desorbed from the wetland bottom in the open pond. As the surface area available for sorption is greater in the vegetated pond (wetland bottom plus plant biofilms), the first-order decrease in the vegetated pond implies faster degradation of bound residues and/or plant uptake. Other studies have also shown an increased effectiveness in pesticide removal of vegetated wetlands compared with non-vegetated wetlands (Schulz *et al.*, 2003).

### INSERT FIGURE 2

Alvord and Kadlec (1996) have proposed that for atrazine, an herbicide of similar water solubility to fluometuron, mass transfer to biofilms on the wetland bottom is the rate-limiting removal step. They calculated mass transfer coefficients of 10-15 m per year for atrazine in a slow-moving wetland, similar to transfer coefficients for BOD and nutrients reported elsewhere. It follows that any increase in biofilm surface area with respect to water volume will reduce transfer distances and accelerate herbicide removal.

Using Alvord and Kadlec's models (1996) and data from this and a previous study of ours, we calculated an average mass transfer coefficient of 9.76 m per year for fluometuron in the ponded wetland. This agrees quite well the values given above, considering our system is static and movement is limited to wind- and bio-turbation. These data can now be manipulated to optimize wetland design criteria for enhanced removal of fluometuron, and similar herbicides, from runoff water. A simple example is given for a cotton farm turkey's nest dam (Figure 3).

### INSERT FIGURE 3

In contrast to fluometuron, the dissipation of total endosulfan was best described by a first-order exponential rate in the open pond and a second-order exponential rate in the vegetated pond. Calculated half-lives are presented in Table 2. A similar, but less pronounced, trend was also observed for suspended solid removal from tailwater entering the wetland. Suspended solids in the open pond fluctuated to a greater extent than in the vegetated pond.

Endosulfan has a high organic partition coefficient ( $K_{oc}$ ) and is associated strongly with sediments in runoff (Crossan *et al.*, 2002). The greater initial rate of sedimentation in the vegetated pond suggests accelerated removal of sediment-bound endosulfan, which would explain the different dissipation curves observed. If this is true, surface water exiting the vegetated pond will contain less endosulfan than water leaving the open pond. Moreover, our results support the hypothesis that aquatic plants assist in sediment stabilisation and decrease the likelihood of contaminated sediment resuspension (Braskerud, 2002).

### INSERT TABLE 2

#### **Pesticide removal in flow-through system**

In all irrigations, pesticide concentrations were observed to increase in the open channels from the inlet to outlet. This was identified as an artifact of the staggered time-composite sampling protocol, and, as a result, outlet concentrations of sub-surface flow channels were normalised with respect to open channel outlet concentrations.

Fluometuron residues in tailwater were low during the first monitored irrigation and outlet concentrations were not significantly different from inlet concentrations (Figure 4). However, in the second monitored irrigation, inlet concentrations were high (an average of 82 ug/l) and were significantly reduced by the SSF gravel beds at the outlet, by a mean of 41% ( $\pm 5\%$ ). Having already undergone two wetting cycles, it is likely that biological activity had increased in the gravel beds, providing greater sorptive capacity. Increased performance with aging has also been observed in SSF wetlands for nutrient removal because of increased biological activity. In the final irrigation, inlet and outlet concentrations were not significantly different although average outlet concentrations were slightly higher.

### INSERT FIGURE 4

Similar results were observed for endosulfan removal. No endosulfan was detected in the first monitored irrigation, but inlet concentrations were significantly reduced in the second monitored irrigation by an average of 26% ( $\pm 17\%$ ). Again, no significant difference was noted in the third monitored irrigation although average outlet concentrations were higher.

One of the main benefits of SSF is that contaminated water remains inaccessible to fauna during treatment, which is not the case with surface flow wetlands. Potential physical exposure is therefore greatly reduced. In this trial sub-surface flow-gravel beds were shown to reduce high pulse-concentrations of pesticide in irrigation tailwater. As with the ponded channel, residual pesticide sorbed to gravel from previous irrigations was probably washed out by 'cleaner' water subsequent irrigations. Nevertheless, this effect was not significant and the total mass removed over both irrigations would still comprise a significant overall reduction.

How can SSF systems be optimised? Media selection for the sub-surface bed composition is important. In this field-trial, basalt gravel was used as a sorbent filter; however experiments in our laboratory and others (Bras *et al.*, 1999) have demonstrated that organic media have a faster pesticide binding time and greater binding capacity. Cotton gin-trash may be a low-cost alternative, but field validation of its use is necessary. As with other wetland systems for pollutant removal, mass loading rates require further attention in future investigations. More research is also required into the degradation of bound residues in the SSF bed to prevent pesticide wash-off, as was observed in our trial. Because irrigations of cotton fields usually occur every 10-14 days in Australia, it is likely that multiple treatment beds would be useful. Alternating between beds would extend the time for degradation between irrigations to 20-30 days.

### **Application of findings**

Our results suggest that the greatest toxicity risk to aquatic species will be from high insecticide pulse concentrations. For a specific chemical, this will usually be a 'one-off' throughout the season and occur in the first irrigation or significant rainfall after application. The incorporation of sub-surface flow filter beds near to field exits is therefore highly desirable, as they can immediately reduce and spread out peak concentrations whilst physically preventing exposure to native species. Alternating between filter beds may be necessary to increase the reduction of bound residues. The removal of remaining pesticide residues can then be accelerated by aquatic plants and system design, through increased sedimentation and biofilm contact.

However, a 'silver-bullet' remains to be found, and probably does not exist. Despite our best efforts, it is unlikely that all irrigation tailwater can be rendered pesticide free under current practices. A further recommendation is therefore proposed: that 'contaminated' and clean tailwater be segregated as best possible and managed accordingly. Classification may as simple as conventional field runoff (high pesticide) versus genetically-modified field (low pesticide) runoff or more thorough, such as analytical monitoring. Contaminated water should be stored in dams that are unattractive to wildlife, such as covered dams, or deep open water bodies with no surrounding vegetation. Conversely, relatively clean water could be used to promote biodiversity, by storage in variably sloped dams with attractive features such as islands, aquatic vegetation, trees and mudflats (Jarman and Montgomery, 2001).

## **CONCLUSIONS**

Current levels of pesticides in tailwater on cotton farms pose a toxicity risk to non-target species. These residues need managing prior to encouraging on-farm biodiversity, especially for aquatic habitats. Our results demonstrate a significant potential for accelerating pesticide removal using aquatic plants and filter beds. We have shown that the design of farm water systems can be manipulated for further reductions in toxicity risk.

## ACKNOWLEDGEMENTS

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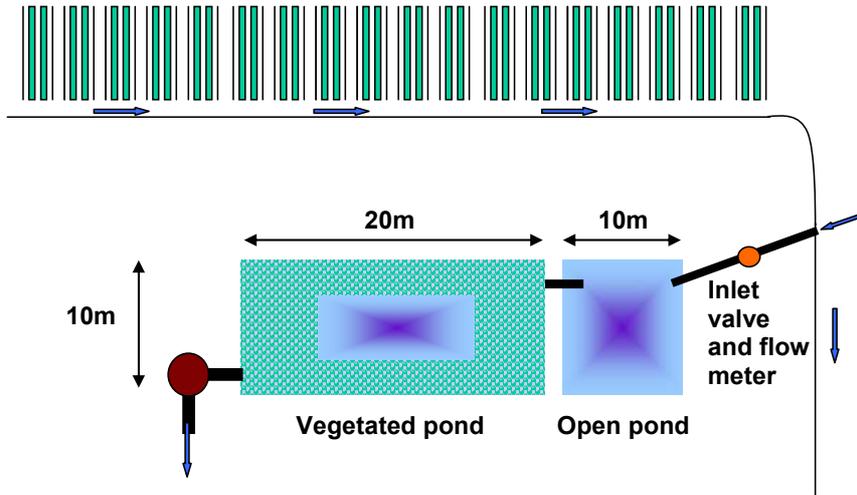
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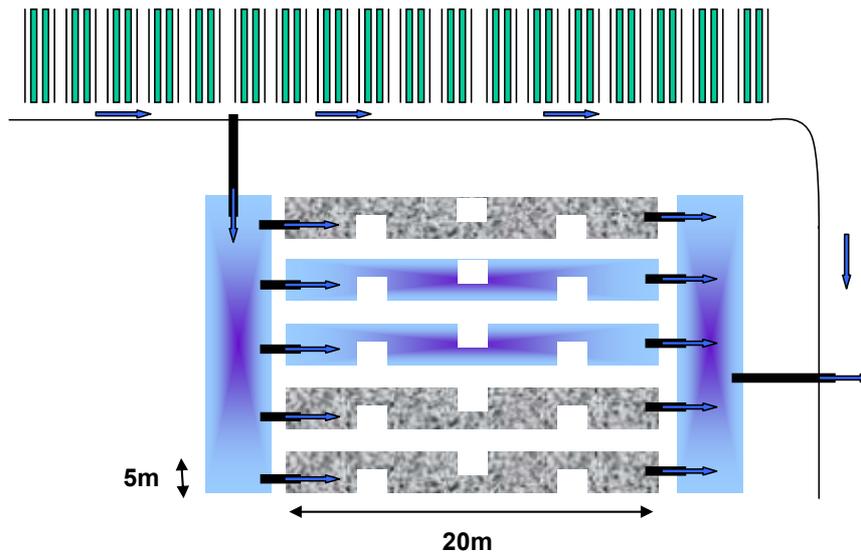
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A.



B.



**Figure 1** Schematic diagram of A. open pond system and B. flow-through system. Blue arrows represent tailwater flow.

**Table 1** Maximum measured pesticide concentration in tailwater during cotton seasons (ug/L), and chronic and acute risk quotients calculated using those concentrations.

Pesticide	Maximum detected concentration (ug/L)	Chronic endpoint concentration (ug/L)	Chronic Risk Quotient	Endpoint	Indicator Species	Reference
Endosulfan	6.6	0.3	22	NOEC <sup>1</sup>	<i>Jappa kutera</i>	(Leonard <i>et al.</i> , 2001)
Fluometuron	90.1	220	0.41	EC50 <sup>2</sup> (population)	<i>Lemna gibba</i>	(EPA, 2000)
Aldicarb	89.6	0.5	179	LOEC <sup>3</sup> EC50	<i>Daphnia magna</i>	(Moore <i>et al.</i> , 1998)
Prometryn	3.2	11.8	0.27	(population)	<i>Lemna gibba</i>	(EPA, 2000)
Diuron	52.2	5	10	NOEC	<i>Lemna gibba</i>	(Teisseire <i>et al.</i> , 1999)

Pesticide	Maximum detected concentration (ug/L)	Acute endpoint (LC50) <sup>4</sup> (ug/L)	Acute Risk Quotient	Indicator Species	Reference
Endosulfan	6.6	0.4	16.5	<i>Macquaria ambigua</i>	(Hose and Van den Brink, 2004)
Fluometuron	90.1	30000	0.003	<i>Oncorhynchus mykiss</i>	(Herbicide Handbook, 1994)
Aldicarb	89.6	1500	0.0597	<i>Lepomis macrochirus</i>	(Kidd and James, 1991)
Prometryn	3.2	5050	0.0006	<i>Oncorhynchus mykiss</i>	(Orme and Kegley, 2004)
Diuron	52.2	9921	0.0053	<i>Oncorhynchus mykiss</i>	(Orme and Kegley, 2004)

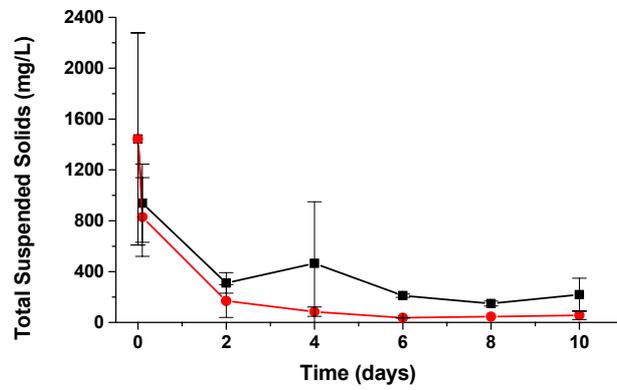
<sup>1</sup> no observable effect concentration

<sup>2</sup> concentration at which the specified endpoint is observed in half the population

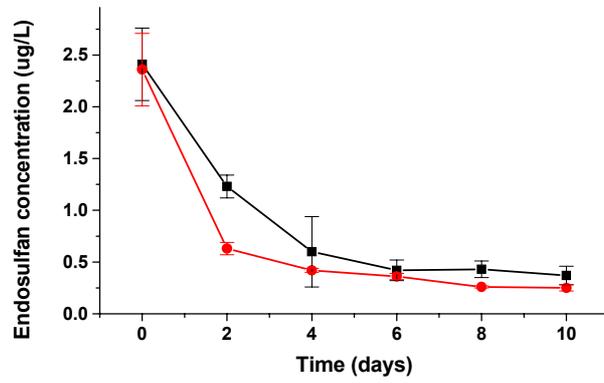
<sup>3</sup> lowest observable effect concentration

<sup>4</sup> concentration at which mortality of half the population is observed

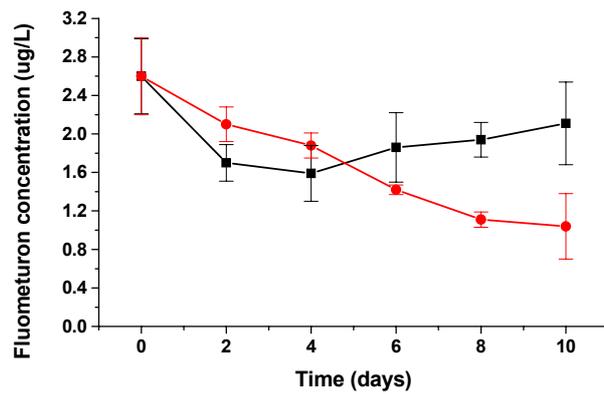
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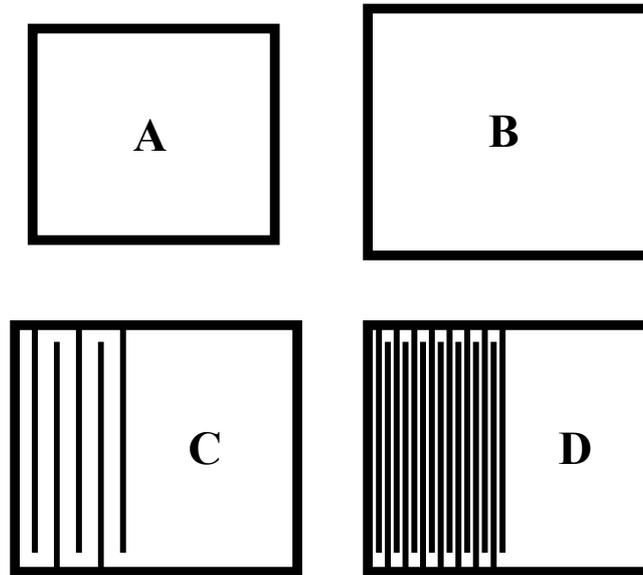
C.



**Figure 2** Concentrations of A. total suspended solids, B. endosulfan, and C. fluometuron in the open pond (squares, black lines) and vegetated ponds (circles, red lines) following the first monitored irrigation. Error bars represent 95% confidence intervals (n=3).

**Table 2** Calculated half-lives for endosulfan and fluometuron (days) in the open pond (OP) and vegetated pond (VP) of the ponded system. Numbers in brackets represent 95% confidence levels (n=3).

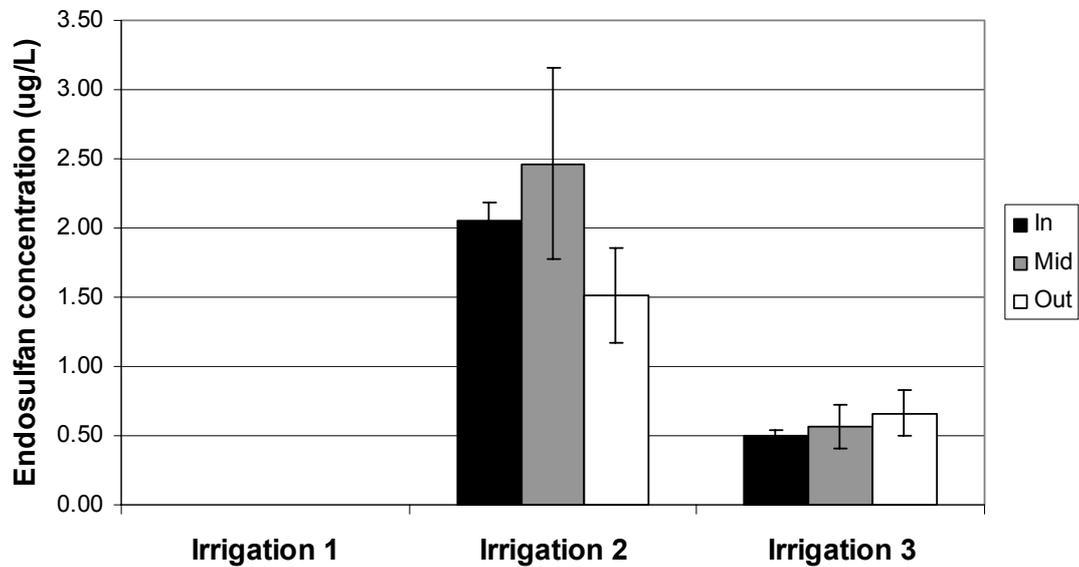
	Zero-order		First-order		Second-order	
	OP	VP	OP	VP	VP	
	t <sub>1/2</sub>	t <sub>1/2</sub>	t <sub>1/2</sub>	t <sub>1/2</sub>	1 <sup>st</sup> t <sub>1/2</sub>	2 <sup>nd</sup> t <sub>1/2</sub>
Endosulfan	-	-	1.35 (0.26)	0.77 (0.10)	0.42 (0.07)	3.26 (0.97)
Fluometuron	-	7.8	-	-	-	-



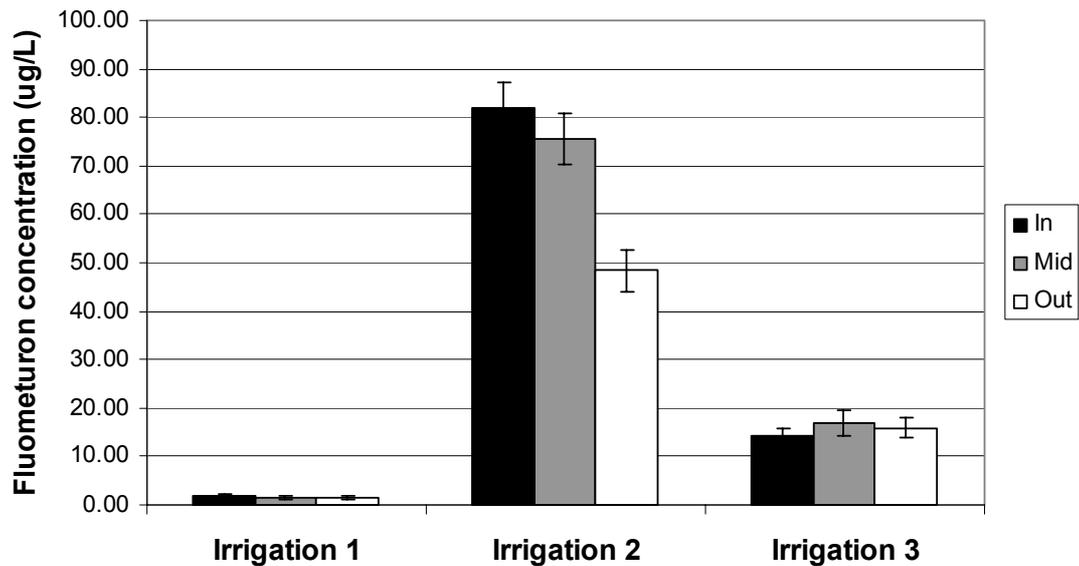
Design	Area/Volume (m <sup>-1</sup> )	Half-life Days)
A	0.38	69.7
B	0.54	49.4
C	0.59	45.1
D	0.80	33.1

**Figure 3** Fluometuron half-life in representative cotton farm turkey's nest dams of 20 ML capacity. A. Side length 82 m, depth 3 m. B. Side length 100 m, depth 2 m. C. Side length 100 m, depth 2 m, 5 × 2.5 width square earth berms. D. Side length 100 m, depth 2 m, 15 × 2.5 width square earth berms.

A.



B.



**Figure 4** Concentrations of A. endosulfan, and B. fluometuron in the inlet, middle, and outlet of filter gravel beds following three irrigation events. Mid and outlet concentrations have been normalised with respect to corresponding open channel concentrations. Error bars represent 95% confidence intervals (n=6).