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Evaluation of a Polyacrylamide Soil Additive to Reduce Agricultural-Associated Contamination

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Abstract Polyacrylamide is an effective water treatment product used to reduce suspended sediment and associated contaminants. An anionic polyacrylamide-containing product was tested for sediment and associated contaminant reduction and potential toxicity in agricultural irrigation and rainfall runoff. The product effectively reduced turbidity, total suspended solids, and phosphate concentrations in the field when compared to the untreated runoff waters. Acute survival of *Ceriodaphnia dubia* and *Pimephales promelas* was not decreased compared to laboratory controls. No significant increases in toxicity were measured in 10-d sediment toxicity tests with *Chironomus dilutus*. Product applications were effective in controlling sediment and nutrient contamination without increasing measured toxicity.

Keywords Agricultural runoff · Polyacrylamide · Acute toxicity · Water quality

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Non-point source contamination contributes to water quality impairments within agriculturally dominated watersheds. Contaminants associated with agriculture include nutrients (e.g. nitrate and orthophosphate), pathogens, suspended sediment, organic matter, and pesticides. Leonard (1990) listed climatic effects as one of the four major factors affecting the transport of pesticides and nutrients from target areas. Effects of rainfall duration, amount and intensity, concurrent with air and rainfall temperature, are believed to be directly related to the volume of contaminants associated with runoff from adjacent fields. Other factors consist of soil characteristics, including texture, organic matter content, compaction and stability, along with slope and topography of the field. In addition to weather, sprinkler and furrow irrigation applications have the potential to promote soil dispersion and movement of agricultural chemicals from production fields.

Since agriculture is cited as the single largest source of contaminants in surface water (34.5%), with an additional 3.4% credited to non-point source runoff such as road construction and maintenance (US EPA 2000a), the use of soil amendments to minimize this source of non-point source contamination is of great investigational importance.

Throughout the 1950s, polymer soil stabilizers such as polyacrylamide (PAM) were tested for their ability to reduce soil erosion (Weeks and Colter 1952). As soil stabilizers, polymers limit detachment of soil particles, thereby decreasing runoff of sediment and sediment-bound chemicals that flow from agricultural fields. However, expensive application rates limited early product adoption. In the 1990s, advanced technology and cost effective application strategies allowed for greater adoption. In studies conducted by Mitchell (1986) and Lentz et al. (1992), PAM's erosion controlling abilities and inexpensive application rates were reported. In 1995, PAM was introduced into the US market for its applications in soil erosion control, although earlier PAM product testing was already underway (Sojka and Lentz 1997). PAM was quickly accepted in the US and by 1999 approximately 400,000 irrigated ha were PAM-treated (http://www.naicc.org/Meeting/2001/UseofPAM.html). Globally, PAM is being used most extensively to stabilize soil in areas with arid and Mediterranean climates (Lentz and Sojka 1994; Lentz et al. 1998; Trout et al. 1995; Shainberg et al. 1991).

Commonly used as a soil additive, anionic high molecular weight, water soluble (non-crosslinked) PAM is often used for erosion and runoff control. To limit detachment of soil particles by rain or irrigation water, PAM stabilizes soil by bridging particles together through cation-bridging (Laird 1997) with Coulombic and Van der Waals forces (Orts et al. 1999; Orts et al. 2000). PAM's formulation, characterized by molecular weight, charge, and charge density, concurrent with soil properties such as texture and clay type, organic matter content, and ions present, further influence its effectiveness as a soil conserving product (Seybold 1994; Green et al. 2000). When particles become detached by raindrop impact or flowing water, PAM's high affinity for suspended sediment flocculates them from the water column and limits transport to downstream receiving systems. Thus, as a water-treatment product, PAM has the ability to flocculate particles suspended in the water column, including fine sediment particles that often have adsorbed contaminants. Lentz et al. (1998) reported that removing suspended sediment in runoff water was positively correlated with other contaminant removal, including phosphorous. Likewise, pesticides can also be found in association with detached sediment particles (Bahr and Stieber 1996). Studies on PAM's efficacy to promote flocculation of agricultural contaminants indicate its effectiveness as a sediment flocculant, resulting in reduced contaminants, such as nutrients, pesticides, and biological oxygen demand (BOD), in the water column (Agassi et al. 1995; Lentz et al. 1998; Lentz et al. 2001).

The objectives of this study were to (1) quantitatively determine product effectiveness at controlling contaminants associated with agricultural runoff, including suspended sediments and nutrients (phosphorus and nitrate) and (2) determine the potential aquatic toxicity of the PAM containing product.

Materials and Methods

In this study we used a storm water pellet containing high molecular weight (MW = 10–14E6 g mol^{-1}), anionic polyacrylamide (PAM SWP) produced by Ciba Specialty Chemicals Corporation (Sulfolk, VA). The product was evaluated with rainfall and irrigation runoff waters to assess its efficacy to flocculate sediment and reduce turbidity in runoff exiting the production field. Exposure studies were conducted at Judd Hill Cotton Plantation, a 1550-ha cotton production farm in Poinsett County, AR, USA. The sample collection sites were on Mhoon (Typic Fluvaquents) and Dundee (Aeric Ochraqualfs) silt loam soils. The sample collection sites were located on cotton fields that were sprinkler irrigated through center pivot irrigation. Water quality analyses and toxicity bioassays of untreated and treated runoff were analyzed at Arkansas State University Ecotoxicology Research Facility (ASU ERF). ASU ERF enforces good laboratory practices and quality assurance guidelines as set forth by the facility.

Treated and untreated water and sediment samples were collected from field locations following three irrigation- and three rain-induced runoff events. Testing consisted of PAM SWP placement in nylon mesh bags to ensure contact of runoff water and PAM SWP. Mesh bags were stabilized within the runoff flow by 1.2 m steel poles. A thin layer of PAM SWP was placed into each individual nylon mesh bag. The United States Natural Resource Conservation Service (NRCS) recommends dissolving 10 kg mL^{-1} (10 ppm) in initial inflow of furrow runoff water, followed by decreased concentration in later runoff episodes (anonymous 1995). The current study utilized enough PAM SWP to form a single layer in the nylon mesh bag since the PAM SWP does not require pre-dissolving. Approximately 50 pellets were evenly distributed within each bag to ensure contact of irrigation and rain-fed runoff water. Three liters of water was collected 1–2 m above treatment and 1–2 m downstream 5 min after exposure. Sediment was also collected from the upper 2–3 cm upstream and downstream of treatment. On-site measurements of irrigation and rain-fed runoff water included flow rate, dissolved oxygen (DO), pH, temperature, and conductivity (APHA 2005). Velocity was measured with a Marsh-McBirney flow meter (Marsh-McBirney, Inc. Frederick, MD).

Following collection, water and sediment samples were packed on ice and transported to ASU ERF for analysis. Water was analyzed for alkalinity, hardness (as $CaCO_3$), orthophosphate, nitrite, nitrate, fecal coliform bacteria, turbidity, and total suspended solids (TSS). All water quality tests followed the American Public Health Association (APHA 2005) guidelines. Sediment particle size distribution (PSD) was determined by the hydrometer procedure as summarized by Gee and Bauder (1986). The cadmium reduction method, applying a 0.01 mg L^{-1} detection limit, was used to evaluate nitrate concentrations. Nitrite was assessed using the diazotization method with a 0.005 mg L^{-1} detection limit. Orthophosphate was determined using the ascorbic acid method. The evaluation of fecal coliform presence in water samples was accomplished by enumerating the number of blue colonies after a

24-h incubation period using rosalic acid as a growth medium.

Laboratory simulations were conducted using sediment samples collected at Judd Hill Cotton Plantation and reference sediment collected from the Black River, Lawrence County, AR, USA. To prepare PAM SWP sediment solutions for the laboratory simulation, untreated sediment samples were emulsified in moderately hard laboratory water (US EPA 2002). Sediment was added to target initial turbidities of 320–345 nephelometric turbidity units (NTU). Crushed PAM SWP was added at concentrations of 30 and 45 mg L^{-1} to prepared suspensions for Judd Hill Cotton Plantation and Black River sediment suspensions, respectively. In order to obtain targeted turbidity reduction, PAM SWP treatment concentrations varied between the study site and the reference sediment.

Aqueous and sediment laboratory bioassays were employed to determine potential in-stream effect of treatment residuals to receiving water bodies. In this current study, only field usage of the product was assessed for

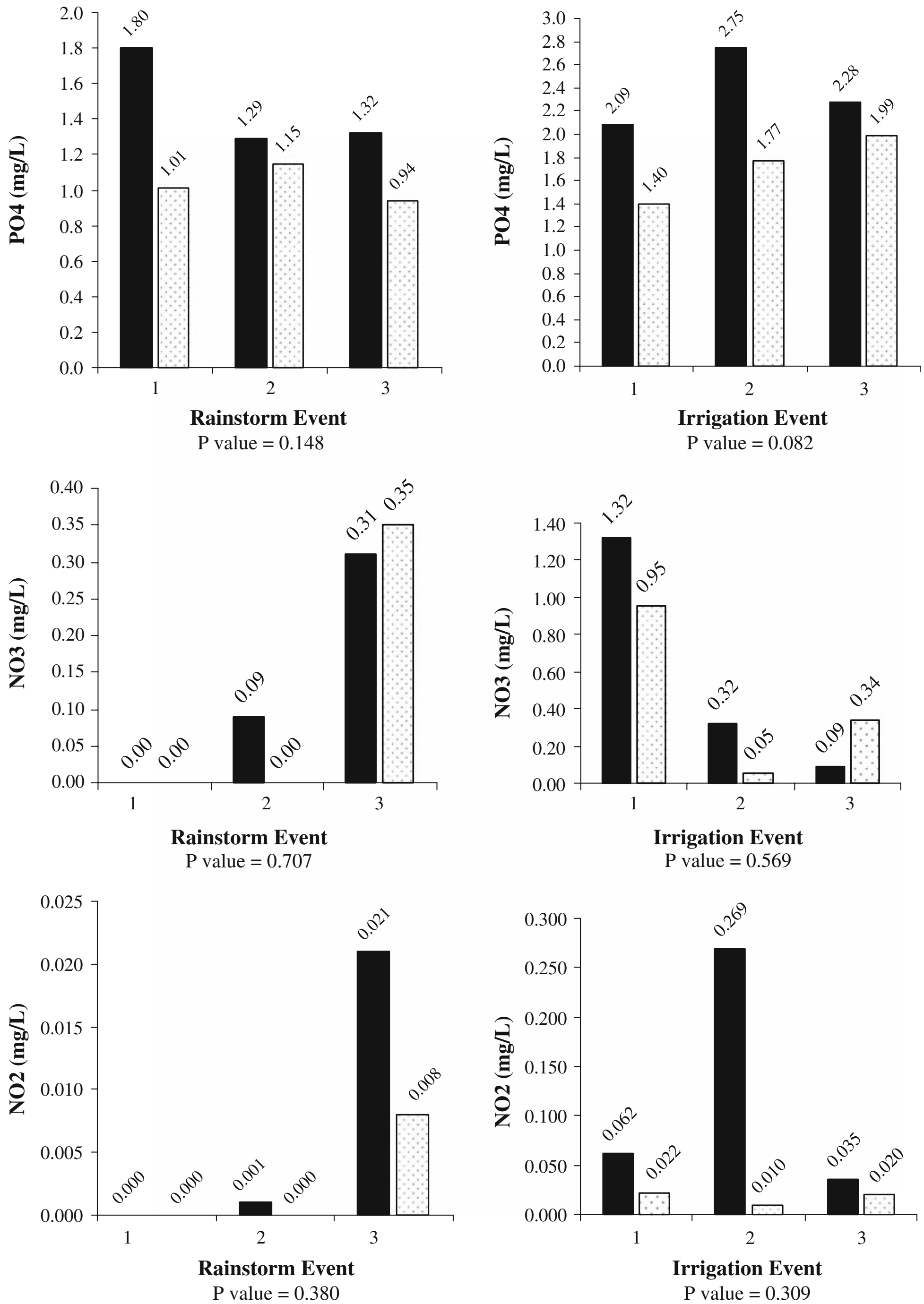

Fig. 1 Orthophosphate, nitrate and nitrite measured from water collected following rain- and irrigation-induced runoff events. Upstream indicates prior to PAM SWP treatment, indicated by solid bars; downstream collected below treatment indicated by patterned bars. *p* values as comparison of upstream/downstream provided for each parameter

potential toxicity. Acute 48-h aqueous toxicity was measured (indicated by % survival) by exposing fathead minnows (*Pimephales promelas*) and water fleas (*Ceriodaphnia dubia*) to water collected above and below PAM SWP field treatments (US EPA 2002). Organisms simultaneously exposed to moderately hard laboratory water were used as controls for statistical comparisons. Ten-day sediment toxicity tests were conducted using midge larvae (*Chironomus dilutus*) (US EPA 2000b) and were statistically compared to Black River reference sediment. Results from above and below field treatments were also statistically compared to determine a change in toxicological response following exposure to PAM SWP.

Aqueous and sediment toxicity results were calculated using ToxCalc™ (1996, Version 5.0.25, McKineyville, CA). Paired t-tests identified statistically significant differences between PAM SWP treated and untreated water and sediment samples using Minitab™ (2000, State College, PA). Data was tested for normality using Shapiro-Wilk's, Steel's Many-One Rank Test, and Dunnett's Test; alpha value was set at 0.05 for all analyses.

Results and Discussion

The effectiveness of PAM SWP as a water treatment product was based on its ability to reduce agricultural-related contaminants. By assessing water and sediment samples from irrigation and rain events, we measured reduced downstream orthophosphate concentrations as compared to upstream untreated water (Fig. 1). While nitrate concentrations were less affected by PAM SWP addition than orthophosphate, nitrite concentrations had greater reduction in irrigation than rain-induced runoff. Such reductions in downstream, treated samples may have resulted from differences in nutrient solubility; while orthophosphates are less soluble in water and often associated with suspended sediment particles, nitrates are more soluble and thus less affected by reduction of suspended sediments by anionic polymers.

Turbidity of PAM SWP-treated runoff water was decreased from upstream measurements in one rainstorm event and all three irrigation events with greatest turbidity reductions measured in irrigation events (Fig. 2). Although TSS were numerically lower in most treated samples compared to untreated samples, no statistically significant differences were detected between downstream and upstream samples (Fig. 3). The sampling events with the greatest flow rates (0.18, 0.28, and 0.30 m/s) (Table 1) resulted in greater TSS reduction than runoff events with lower flow; the same high flow runoff events also measured the greatest numerical reduction in orthophosphate and turbidity. This observation is consistent with applications by Lentz and Sojka (1999) who also reported more efficient performance of equal concentrations of PAM at greater flow rates. In the same study, Entry and Sojka (1999) also reported PAM's effectiveness at removing microorganisms, including bacteria and fungi, from runoff waters. However, in the current field study no statistical decrease in fecal coliforms were measured between PAM SWP-treated and untreated water samples.

Although statistical comparisons of upstream/downstream nutrient measurements resulted in p values > 0.05, most upstream values were numerically greater. This suggests no statistically significant differences but indicates a degree of nutrient reduction following treatment in most runoff events (Fig. 1). Measured TSS following rain-induced runoff had statistically significance differences between upstream/downstream of PAM SWP treatment (p value $= 0.006$). Failure of many of these water quality parameters to produce statistical significance may be due to the large disparity in data among the various runoff events. Numerical differences between PAM SWP-treated and untreated samples, however, reveal trends that indicate the

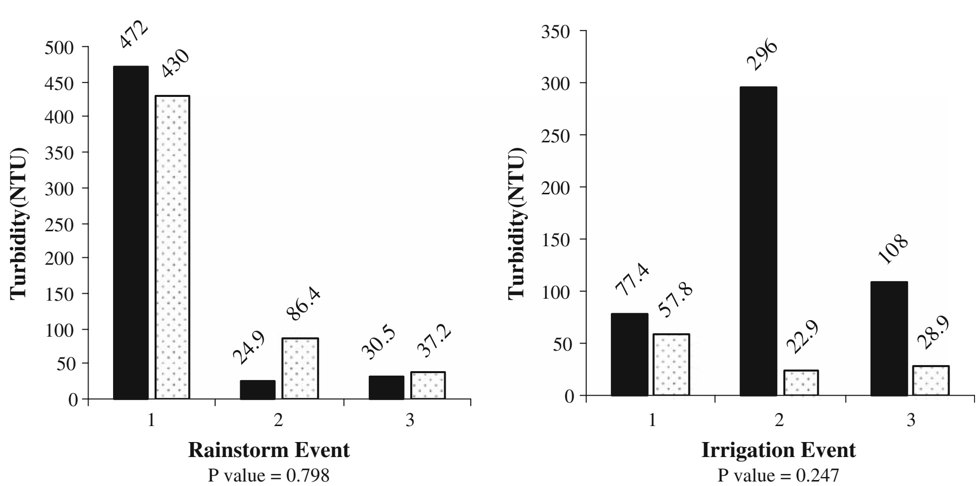

Fig. 2 Turbidity measured from water collected following rain- and irrigation-induced runoff events. Upstream indicates prior to PAM SWP treatment, indicated by solid bars; downstream collected below treatment indicated by patterned bars. p values as comparison of upstream/downstream provided for each parameter

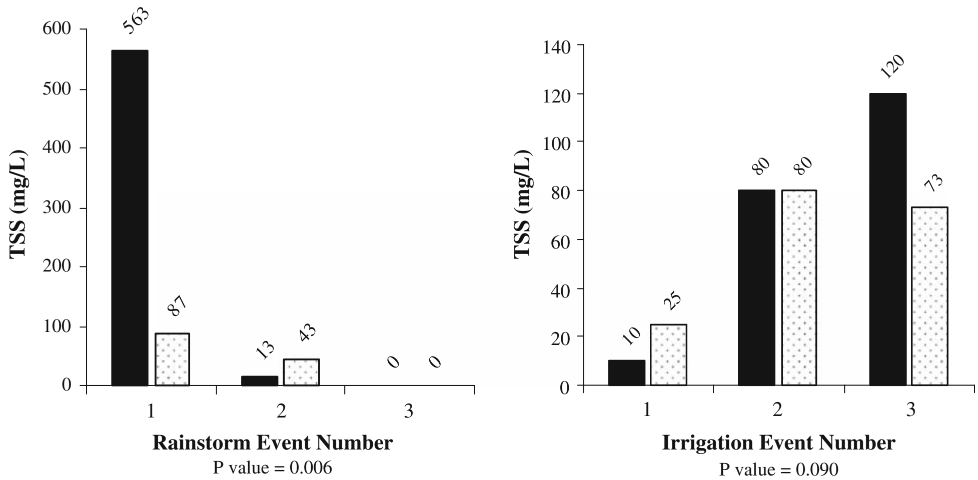

Fig. 3 Total suspended solids (TSS) measured from water collected following rain- and irrigation-induced runoff events. Upstream indicates prior to PAM SWP treatment, indicated by solid bars; downstream collected below treatment indicated by patterned bars. *p* values as comparison of upstream/downstream provided for each parameter

product's capability of improving water quality. Overall TSS reduction was most significant, and results also indicate that the PAM SWP product reduced nutrient and turbidity levels more efficiently following irrigation events. Irrigation runoff had significantly greater water hardness than water sampled during rainfall events indicating that cations present in groundwater used for irrigation increased PAM SWP efficacy (Table 1) (Laird 1997; Shainberg et al. 1990).

Sediment deposited in Judd Hill Plantation agricultural ditches is composed primarily of silt and sand (Table 2) and would be expected as the predominant soil type of this study's field locations is silt loam. With Fincastle silt loam (71% silt) soils in Indiana, the molecular weight (MW) of PAM was the determining factor affecting soil slaking and low MW PAM (6 Mg mol^{-1}) was reported to be the most effective soil stabilizer (Green et al. 2004). Thus the high molecular weight of the PAM SWP in the current study may have limited optimum field results.

Paired *t*-tests of laboratory simulations utilizing crushed PAM SWP resulted in significant reductions in turbidity and fecal coliforms, but reduction of TSS was not significant at the preset alpha value of 0.05 (p value = 0.095)

Table 1 Measured physicochemical parameters from upstream and downstream of PAM SWP amendments following runoff events

	pH[a]	DO (mg L^{-1})[a]	Cond (mS cm^{-1})[a]	Temp (°C)[a]	Flow rate (m s^{-1})[a]	Alkalinity (mg L^{-1})	Hardness (mg L^{-1})	Fecal coliforms (CFU 100 mL^{-1})
Rainstorm event #1								
Upstream	6.61	7.5	41	25.4	0.3	8	10	280
Downstream	6.72	7.5	29	25.0	0.3	8	10	330
Rainstorm event #2								
Upstream	6.76	7.8	46	23.5	0.04	20	30	7,200
Downstream	6.68	8.2	56	23.5	0.04	18	20	6,700
Rainstorm event #3								
Upstream	6.45	6.7	37	23.3	0.02	16	20	77,200
Downstream	6.25	6.6	62	23.4	0.02	14	10	79,200
Irrigation Event #1								
Upstream	7.85	6.6	452	30.9	NT	200	210	9,920
Downstream	7.70	7.3	463	30.7	NT	188	220	10,560
Irrigation event #2								
Upstream	8.12	8.1	410	26.0	0.28	178	220	29,640
Downstream	8.05	8.8	418	25.4	0.28	172	200	27,630
Irrigation event #3								
Upstream	8.10	8.1	415	25.8	0.18	186	200	10,920
Downstream	8.12	8.0	423	25.9	0.18	182	210	9,440

[a] Measurements taken on site

NT = Not tested

Table 2 Particle size composition (PSC) of sediment collected from upstream and downstream of PAM SWP amendments following runoff events

	% Sand	% Clay	% Silt
Rainstorm event #1			
Upstream	33.28	0.11	66.62
Downstream	29.49	0.02	70.49
Rainstorm event #2			
Upstream	11.10	0.05	88.85
Downstream	6.66	0.01	93.33
Rainstorm event #3			
Upstream	47.16	0.06	52.78
Downstream	22.19	0.00	77.83
Irrigation event #1			
Upstream	47.61	0.00	52.40
Downstream	49.10	0.09	50.81
Irrigation event #2			
Upstream	71.67	0.00	28.53
Downstream	67.78	0.00	32.44
Irrigation event #3			
Upstream	63.64	0.00	36.36
Downstream	58.03	0.00	42.23

(data not shown). Presence of calcium and magnesium, concurrent with increased surface area of the crushed pellets, most likely enhanced product effectiveness for turbidity and fecal coliforms in these laboratory simulations. Laboratory water utilized in these exposures has measured hardness of 90–110 mg L^{-1} thus providing sufficient divalent cations for efficient product bridging.

Aqueous test organisms exposed to PAM SWP-treated and untreated waters from rain and irrigation events did not elicit significantly decreased survival after 48 h when compared to laboratory controls in moderately hard water (Table 3). There was also no significant differences between aqueous test organism survival exposed to PAM SWP-treated and untreated runoff waters. These results indicate that exposure to PAM SWP product does not elicit an acute toxic response to aquatic organisms. Previous aqueous toxicity studies with anionic PAM revealed high LC50 (i.e. low toxicity) values in fish studies with no adverse effect measured in 90-day tests using anionic PAM at >100 ppm (Buckholz 1992; Barvenik 1994) and protection of downstream aquatic biota has also been reported (Goodrich et al. 1991; Buchholz 1992).

When compared to laboratory controls, decreased survival of *C. dilutus* was measured in sediment from all rainstorm events (Table 3). Although there was no significant difference between survival of test organisms exposed to PAM SWP-treated and untreated sediments collected following rainstorm events 1 and 2, organisms exposed to downstream sediment collected from rainstorm event 3 had significant increased survival as compared to untreated (upstream) samples. Decreased survival and growth (as compared to laboratory control sediment) was measured in PAM SWP-treated sediment following irrigation event 1, but no significant difference was measured

Table 3 Measured endpoints from acute of water and sediment samples collected following PAM SWP treatments at Judd Hill Plantation

	C. dubia	*P. promelas*	*C. dilutus*	
	% survival	% survival	% survival	Growth (mg)
Rainstorm event #1				
Upstream	NT	100 ± 0.0	70 ± 13.2[a]	1.70 ± 0.36[a]
Downstream	NT	80 ± 32.3	47.5 ± 14.5[a]	1.35 ± 0.41[a]
Rainstorm event #2				
Upstream	85 ± 16.0	85 ± 18.1	22.5 ± 10.4[a]	1.38 ± 0.76[a]
Downstream	100 ± 0.0	100 ± 0.0	15 ± 16.0[a]	1.14 ± 0.0[a]
Rainstorm event #3				
Upstream	100 ± 0.0	97.5 ± 5.7	0.0 ± 0.0[a]	NT
Downstream	85 ± 16.0	95 ± 6.7	65 ± 0.0[a]	2.90 ± 0.56
Irrigation event #1				
Upstream	90 ± 16.8	100 ± 0.0	85 ± 12.2	2.81 ± 0.81
Downstream	100 ± 0.0	90 ± 8.9	72.5 ± 12.5[a]	1.80 ± 0.59[a]
Irrigation event #2				
Upstream	90 ± 10.8	87.5 ± 15.3	95 ± 6.7	3.48 ± 0.38
Downstream	100 ± 0.0	95 ± 6.7	97.5 ± 5.7	3.60 ± 0.26
Irrigation event #3				
Upstream	100 ± 0.0	100 ± 0.0	93.3 ± 12.5	3.44 ± 0.17
Downstream	100 ± 0.0	100 ± 0.0	100 ± 0.0	3.28 ± 0.69

Mean 48-h survival ± SD given for *Ceriodaphnia dubia* and *Pimephales promelas*. Mean 10-day survival and growth ± SD given for *Chironomus dilutus*

[a] Denotes significant difference from control ($\alpha = 0.05$)

NT = not tested

between this sediment and the untreated sediment from this same event. Since no aqueous toxicity was measured in samples from rainstorm event 3, increased survival of PAM SWP treated sediments following this runoff event could be attributed to a decreased bioavailability of sediment-bound contaminants.

Use of PAM SWP can effectively mitigate agricultural-associated contamination. These field tests measured lower phosphorous, turbidity, TSS, and in one event, decreased toxicity following PAM SWP treatment. Laboratory simulations showed greater efficacy for reducing turbidity, TSS, and fecal coliform counts when compared to field testing of the product. Since the presence of divalent cations is essential to the efficiency of anionic PAM products, the presence of calcium and magnesium in laboratory and irrigation waters increased product efficacy compared to rainwater applications. Laboratory simulations also suggest that applying PAM SWP in granular form may be an effective application method to consider. While further research is underway to thoroughly assess the product's potential in the agricultural industry, extended field testing may include multiple application types (i.e. liquid emulsions, granules), variations in time intervals, flow rates, and distances from the point of application.

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