

# Environmental Degradation of Polyacrylamides

## II. Effects of Environmental (Outdoor) Exposure

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The environmental fate of a polyacrylamide thickening agent (PATA), formulated without and with a glyphosate-surfactant herbicide (GH), was examined under various environmental situations: formulation in surface water and ground water, volatility, and soil mobility. *Environmental Fate of PATA in Surface Water and Ground Water:* PATA was formulated at four concentrations in distilled-deionized water, three surface water samples, and two ground water samples, without and with a GH. Solutions were placed in glass bottles, covered with plastic wrap, and exposed to environmental (outdoor) conditions for 6 weeks. Acrylamide and ammonium concentration, pH, and bacterial and fungal populations were measured weekly. All solutions in this portion of the study had a homogeneous milky appearance but by the conclusions of the study were nearly transparent. The results of this study suggest that polyacrylamide can degrade to acrylamide under environmental conditions. Statistically, there was no linear correlation between the various parameters measured. *Volatility:* PATA was formulated without and with GH. Each solution plus an acrylamide standard (positive control) was placed in a glass beaker and exposed to environmental (outdoor) conditions for 6 days. Acrylamide concentration, ammonium concentration, pH, and solution volume were measured daily. Acrylamide and ammonium concentrations increased during the study in all formulations, except when solutions evaporated to dryness. pH did not change greatly over the course of the study for these samples. Those solutions containing PATA had a homogeneous milky appearance but by the conclusions of the study were nearly transparent. This suggests a physical structural change in the polymer. *Soil Mobility:* PATA formulated with GH was also applied to soil columns and soil boxes containing sand, Eudora sandy loam, Eudora sandy clay, and Kohola silt loam. Acrylamide could be detected by Day 2 in all soil columns. Acrylamide could not be detected in the runoff of any of the soil boxes. © 1997 Academic Press

## INTRODUCTION

Polyacrylamides (Fig. 1A) are water-soluble synthetic linear polymers made of acrylamide alone or the combination of

acrylamide and acrylic acid (Anonymous, 1991). Among their various uses, polyacrylamides are mixed with various organic solvents to form thickening agents that are then combined with herbicides (i.e., glyphosate) to increase the herbicides' surfactant capabilities (Bouse *et al.*, 1986). These polyacrylamide thickening agents (PATA) contain, on the average, 25 to 30% polyacrylamide (product label for Nalco-Trol II, Nalco Chemical Co., Naperville, IL; 38-F and 41-A, Sanag, Los Angeles, CA; DriftGard, Fresno, CA). Polyacrylamides for the most part are nontoxic. However, after polymerization these polymers can have a residue of acrylamide; a known peripheral nerve toxin (McMollister *et al.*, 1965) that can range between 0.05 and 5.0% of the final product (Croll *et al.*, 1974). Acrylamide (Fig. 1B) is highly water soluble and has been found to be absorbed by all routes of exposure (Windholz, 1989; Hashimoto and Aldridge, 1970; Druckery *et al.*, 1953). Once absorbed, acrylamide produces an ascending central/peripheral axonopathy in man and animals (Anonymous, 1985). The axonopathy is characterized as a disruption of sensory, motor, and autonomic function in the peripheral nerves and ascending tracts of the spinal cord (Miller and Spencer, 1985; Tilson, 1981; Spencer Schaumberg, 1974). Acrylamide axonopathy is reversible with time, but full recovery depends upon the severity of the intoxication (Miller and Spencer, 1985). All known cases of acrylamide toxicity are attributed to the handling of the monomer (Dyke *et al.*, 1975).

The contamination of surface water and/or ground water from agricultural chemicals (i.e., pesticides) and agricultural by-products (i.e., nitrates from fertilizers) is a concern to public health (Anonymous, 1990; Torstensson, 1990; Stratmann and Buhbauch, 1987; Visi *et al.*, 1986). Those chemicals and by-products which reach these aquifers share two common properties: (1) they are highly water soluble and (2) they do not interact with insoluble material. Such is the case of polyacrylamide and acrylamide in the environment. Polyacrylamides are capable of translocating through different soil types under various conditions (Spalding and Hyder, 1985; Lande *et al.*, 1979). For the most part, they are considered water soluble but under

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certain conditions can bind to particulate matter such as clay particles (Stutzmann and Siffert, 1977; Nabzar *et al.*, 1986). Once bound, polyacrylamides are suspected of biodegrading to CO<sub>2</sub> and NH<sub>3</sub>, although release of the acrylamide monomer has not been reported. Acrylamide, which can also translocate through various soil types and reach deep rock aquifers, is not absorbed by sediments or affected by water treatment (Lande *et al.*, 1979; Conway *et al.*, 1979; Brown *et al.*, 1980). Like polyacrylamide, acrylamide will biodegrade to CO<sub>2</sub> and NH<sub>3</sub> (Croll *et al.*, 1974; Cherry *et al.*, 1956).

In a previously reported study, the effects of various artificial temperature and light conditions on acrylamide concentrations in formulations of PATA alone decreased over time (Smith *et al.*, 1996). These concentrations decrease faster under conditions of high temperatures than of low temperatures and at 24-hr light than at 24-hr dark. In contrast, acrylamide concentrations for PATA solutions formulated with glyphosate-surfactant herbicide (GH) remained the same or increased throughout the study. These data suggest that GH either protected acrylamide and/or polyacrylamide from degradation or promoted degradation of polyacrylamide to acrylamide. Ammonium concentrations and pH were used as parameters for whether the monomer and/or polymer degraded. Ammonium concentrations typically increased as acrylamide concentrations decreased but leveled off when acrylamide concentrations could no longer be detected. pH indicated no statistically significant changes throughout the study. However, the addition of GH to a PATA solution immediately lowered the pH of the solution from approximately 7.7 to 4.9. Statistical analyses comparing acrylamide concentration, ammonium concentration, and pH with each other exhibited no strong linear relationship. Also, the addition of GH to a PATA solution changed the physical appearance of PATA from white suspended globules to a homogeneous milky solution. Once formulated, the physical appearance of these solutions remained unchanged throughout the course of the various studies. Adjusting the pH of PATA formulations did not promote the depolymerization of polyacrylamide. Finally, the ammonium ion concentration in dilute solutions of ammonium salts decreased as the amount of GH increased, suggesting that GH interacts with ammonium. Therefore, under artificial environmental conditions polyacrylamide may degrade to acrylamide.

In an agricultural setting, the degradation of polyacrylamide may create an environmental/health hazard. If polyacrylamide were to release acrylamide during the degradation process, acrylamide, because of its chemical properties, could adulterate surface water and/or ground water systems. Rural communities rely upon these types of water sources for livestock and personal use. Adulteration of these sources would impact not only the health but the economical welfare of those communities. Therefore, the objective of this study is to examine the environmental fate of a polyacrylamide thickening agent, alone and in combination with a herbicide, under environmental (outdoor) conditions.

## MATERIALS AND METHODS

### Chemicals

The polyacrylamide thickening agent used was NALCO-TROL II (Nalco Chemical Company, Naperville, IL). The glyphosate-surfactant herbicide used was RoundUp (Monsanto, St. Louis, MO). Distilled water was filtered through a NANOpure D1793 filtration system (Barnstead Sybron Corporation, Boston, MA). Acrylamide (>99.9% pure; standards) was obtained from the United States Biochemical Corporation (Cleveland, OH).

### *The Formulation of a Polyacrylamide Thickening Agent without and with Glyphosate-Surfactant Herbicide*

The PATA was formulated at four concentrations in distilled-deionized, ground, or surface water, without and with the GH. The formulations for PATA are as follows:

Code	PATA (liters)	Water (liters)
1X	1.2	378.5
2X	2.3	378.5
3X	3.5	378.5
4X	4.7	378.5

The concentration of the GH was reduced to 1/20th of its normal application concentrations because of masking effects observed during the chromatographic separation of acrylamide and glyphosate (see *Acrylamide Analyses*). The four PATA solutions were within the range recommended by the manufacturer for the GH.

### *Acrylamide Analyses*

Acrylamide analyses were performed by high-performance liquid chromatography (HPLC) using a Perkin-Elmer Series 2 liquid chromatograph (Norfolk, CT) with a Perkin-Elmer LC-55 B spectrophotometric detector, a Perkin-Elmer deuterium power supply for the LC-55 B spectrophotometric detector, and a Hewlett-Packard 3390A integrator. A 50- $\mu$ l injection loop was used to deliver samples onto a Hewlett-Packard 200  $\times$  2.1-mm reverse-phase ODS Hypersil 5- $\mu$ m C-18 column and 20  $\times$  2.1-mm 5- $\mu$ m guard column. A phosphate buffer was used for the mobile phase [0.84 g of KH<sub>2</sub>PO<sub>4</sub> (Merck) dissolved into 960 ml of deionized water and 40 ml of methanol (HPLC grade—Baker)]. Separation was at room temperature with a flow rate of 0.3 ml min and a wavelength of 208 nm. No filtration, extraction, or conjugation was performed on the samples. A four-point standard curve of acrylamide (0.1 to 2.0  $\mu$ g/ml) was run at the beginning and end of each sample period (Smith and Oehme, 1993).

### *Ammonium Analyses*

Ammonium analyses were performed colorimetrically using a Techicon Auto Analyzer (consisting of a sampler, manifold,

proportioning pump, and heating bath with double delay coil), a Techicon colorimeter (equipped with a 15-mm tubular flow cell and a 630- to 660-nm filter), and recorder. All samples were diluted 1:10 with distilled-deionized water before analysis. This method had a range of 0.01 to 2.0 mg  $\text{NH}_4$  as N/L. The samples reacted with alkaline phenol and hypochlorite to form indophenol blue that was proportional to the ammonia concentration. The blue color formed was intensified with sodium nitroprusside (Strickland and Parsons, 1972).

#### pH Readings

pH measurements were taken with an Orion Research digital pH meter, Model 701.

#### Environmental Exposure of Surface and Ground Water Samples

Three surface water samples (a pond, a creek, and a natural spring) and two ground water samples (well R and well V) were collected from three different sites within a 5-mile radius (Fig. 2). The two ground water samples are believed to be from different water tables, but exact conformation was not possible because of the fragile rock structure of the tables. Each solution was prepared in triplicate and placed in a 50-ml clear glass bottle with two layers of plastic wrap over the mouth. The plastic was held in place with a size 19 ( $3\frac{1}{2} \times \frac{1}{16}$ " ) rubber band. The solutions were placed outdoors and exposed to environmental temperature, radiation, and other weather conditions for 6 weeks (42 days). Each bottle was analyzed for acrylamide and ammonium concentrations, bacterial and fungal population, and pH on Days 0, 7, 14, 21, 28, 35, and 42.

#### Assaying Bacterial and Fungal Population

Solutions of PATA(1 $\times$ ) and PATA(4 $\times$ ) without and with GH were plated on PTYG agar and Sabourand Dextrose and Mycobiotic agar (SDM; Difco Laboratories, Detroit, MI). The PTYG and SDM plates were incubated at 25°C and observed

for bacterial and fungal populations, respectively. The PTYG plates were observed daily for 1 week. The SDM plates were observed weekly for 3–4 weeks.

#### Volatility

A volume of 100 ml of the following solutions: (1) 0.2  $\mu\text{g}$  acrylamide/ml distilled-deionized water, (2) PATA(4 $\times$ ), and (3) PATA(4 $\times$ )/GH, was placed in 150-ml beakers and exposed to environmental (outdoor) conditions for 1 week. The beakers were left open to the environment. The content of each beaker was analyzed in triplicate for acrylamide concentrations, ammonium concentrations, and pH on Days 0, 1, 2, 3, 4, 5, and 6.

#### Soil Tubes

PVC Schedule 80 tube (3" id) was cut to 2.5 feet in length and beveled on one end. Each tube was attached to a hydraulic press and driven into the ground to collect a core sample of undisturbed soil. The three soils selected were sand, Eudora sandy loam, and Kohola silt loam. Once filled, the beveled end of each tube was cut to a final length of 2 feet and capped with a  $\frac{3}{8}$ " plexiglass base plate containing a  $\frac{1}{4}$ " id opening for drainage. The soil within the tube was saturated from the bottom with 1 mM sodium sulfate to establish a saturated flow system. A solution of PATA(4 $\times$ ) with GH was made and the concentration of acrylamide in the solution determined. One hundred milliliters of this solution was added to each tube. Wash samples were taken every 8 hr for 1 week and analyzed for acrylamide.

#### Soil Boxes

Plastic tubs measuring 18  $\times$  12  $\times$  10" were used for soil boxes. The soil boxes were filled with Eudora sandy loam, Eudora sandy clay, or Kahola silt loam and exposed to outdoor conditions. The boxes underwent a 1-week conditioning phase (i.e., exposure to outdoor conditions). After the soil was conditioned, 1 liter of PATA(4 $\times$ ) with GH was sprayed over the surface of each box. Four days after the application of the PATA(4 $\times$ )/GH solution, the boxes were set at a 5° angle and washed with 1 mM sodium sulfate. The runoff (supernate) was collected and analyzed for acrylamide.

#### Statistical Analysis

With respect to PATA formulated in the various surface water and ground water samples, without and with GH, correlation coefficients were calculated for the following pairs of observations: acrylamide concentration and ammonium concentration, acrylamide concentration and pH, and ammonium concentration and pH (Ott, 1984).

## RESULTS

#### Environmental Conditions

The average high temperature was 33°C and the average low temperature was 20°C. Total hours of sunlight per day was from 14.1 to 14.6 hr.

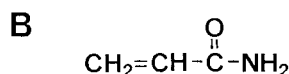
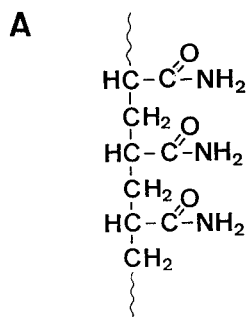


FIG. 1. Chemical structures of polyacrylamide (A) and acrylamide (B).

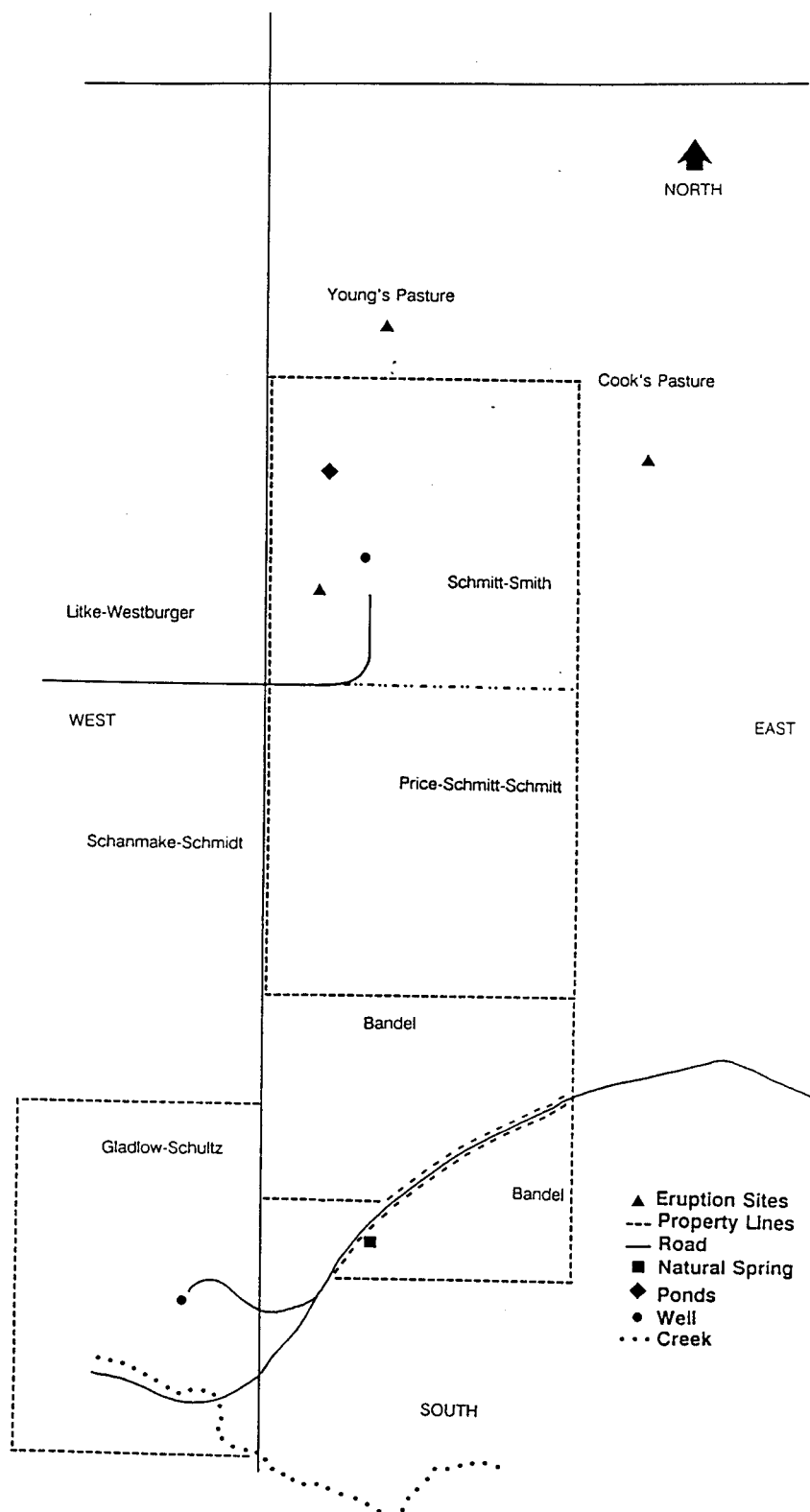


FIG. 2. A map of the sites where each of the surface water and ground water samples were collected.

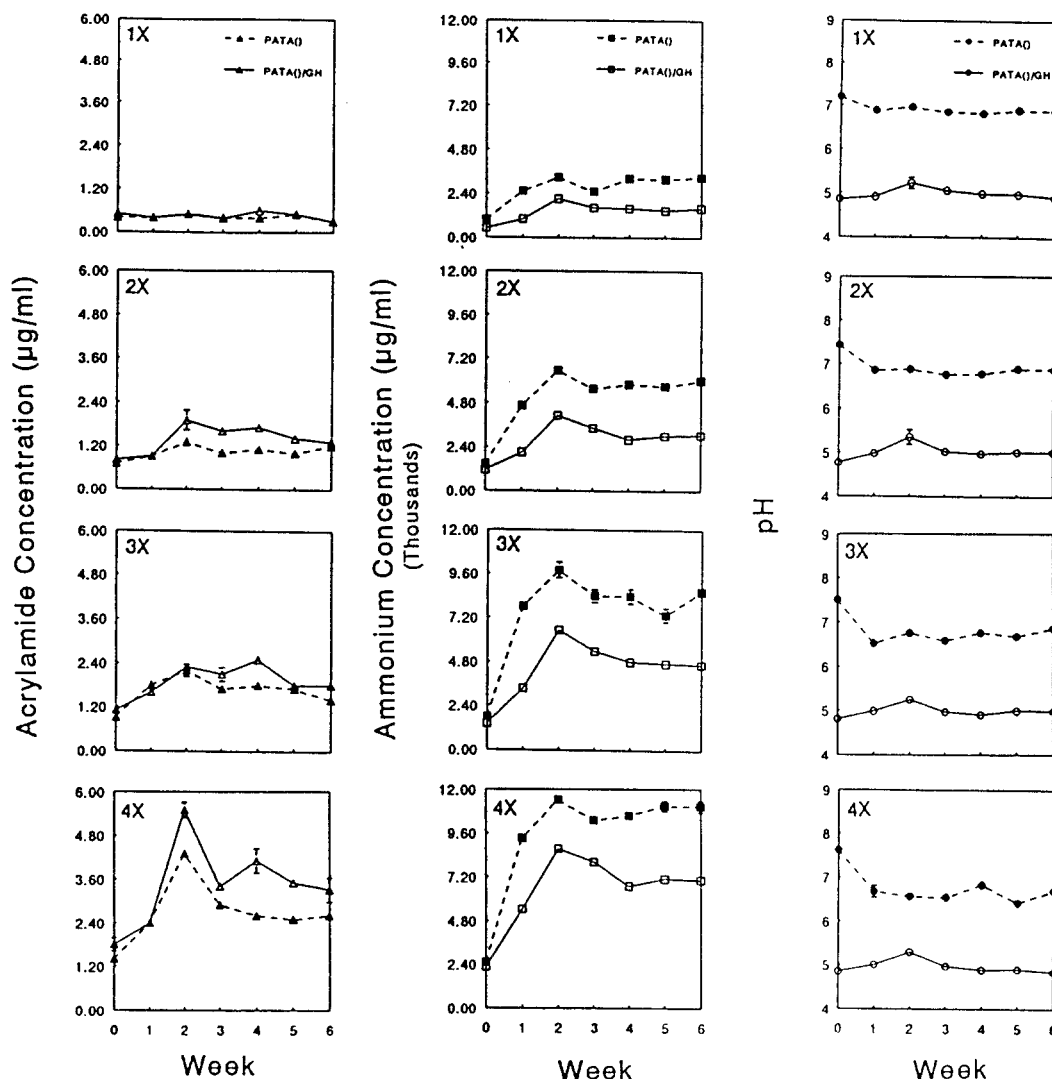


FIG. 3. Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in distilled-deionized water and exposed to environmental (outdoor) conditions for six weeks.

#### Distilled-Deionized Water

Acrylamide concentrations increased until Week 2 and then decreased throughout the remainder of the study. At Week 2, the acrylamide concentration for the various formulations was from 1.2 to 3.0 times the initial concentration (Fig. 3). The ammonium concentration in the various formulations increased until Week 2 and then fluctuated throughout the remainder of the study (Fig. 3). pH did not change greatly with respect to the individual groups (Fig. 3). The various formulations were not cultured for bacteria and fungi. Statistical analyses for the individual treatments revealed no strong linear correlation between the selected parameters (Table 1). All solutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation, and by the conclusion of the study all solutions were nearly transparent.

#### Creek

Acrylamide concentrations increased until Week 3 and then decreased throughout the remainder of the study. At Week 3,

the acrylamide concentration for the various formulations was from 3.2 to 6.2 times the initial concentration (Fig. 4). The ammonium concentration for the various formulations displayed a decreasing trend throughout the study (Fig. 4). The pH for the various formulations generally increased throughout the study (Fig. 4). There was no specific trend for the bacteria that cultured over the 6-week study period. There was no fungal growth over the 6-week study period. Statistical analyses for the individual treatments revealed no strong linear correlation between the selected parameters (Table 2). All solutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation, and by the conclusion of the study all solutions were nearly transparent.

#### Pond

Acrylamide concentrations increased until Week 5 and then decreased throughout the remainder of the study. At Week 5, the acrylamide concentration for the various formulations was from 2.5 to 9.5 times the initial concentration (Fig. 5). The

TABLE 1

Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Distilled Deionized Water without or with a Glyphosate Herbicide (PATA/GH)

Correlation pair	PATA( )				PATA( )/GH			
	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )
AC/ $\text{NH}_4$	0.28	0.88	0.78	0.70	0.12	0.77	0.80	0.75
AC/pH	0.03	-0.63	-0.83	-0.59	0.00	0.71	0.37	0.62
$\text{NH}_4$ /pH	-0.72	-0.85	-0.79	-0.89	0.60	0.70	0.63	0.49

Note. The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/ $\text{NH}_4$ , acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH;  $\text{NH}_4$ /pH, ammonium concentrations vs pH. PATA(1 $\times$ ), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2 $\times$ ), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3 $\times$ ), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4 $\times$ ), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1 $\times$ )/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2 $\times$ )/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3 $\times$ )/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4 $\times$ )/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.

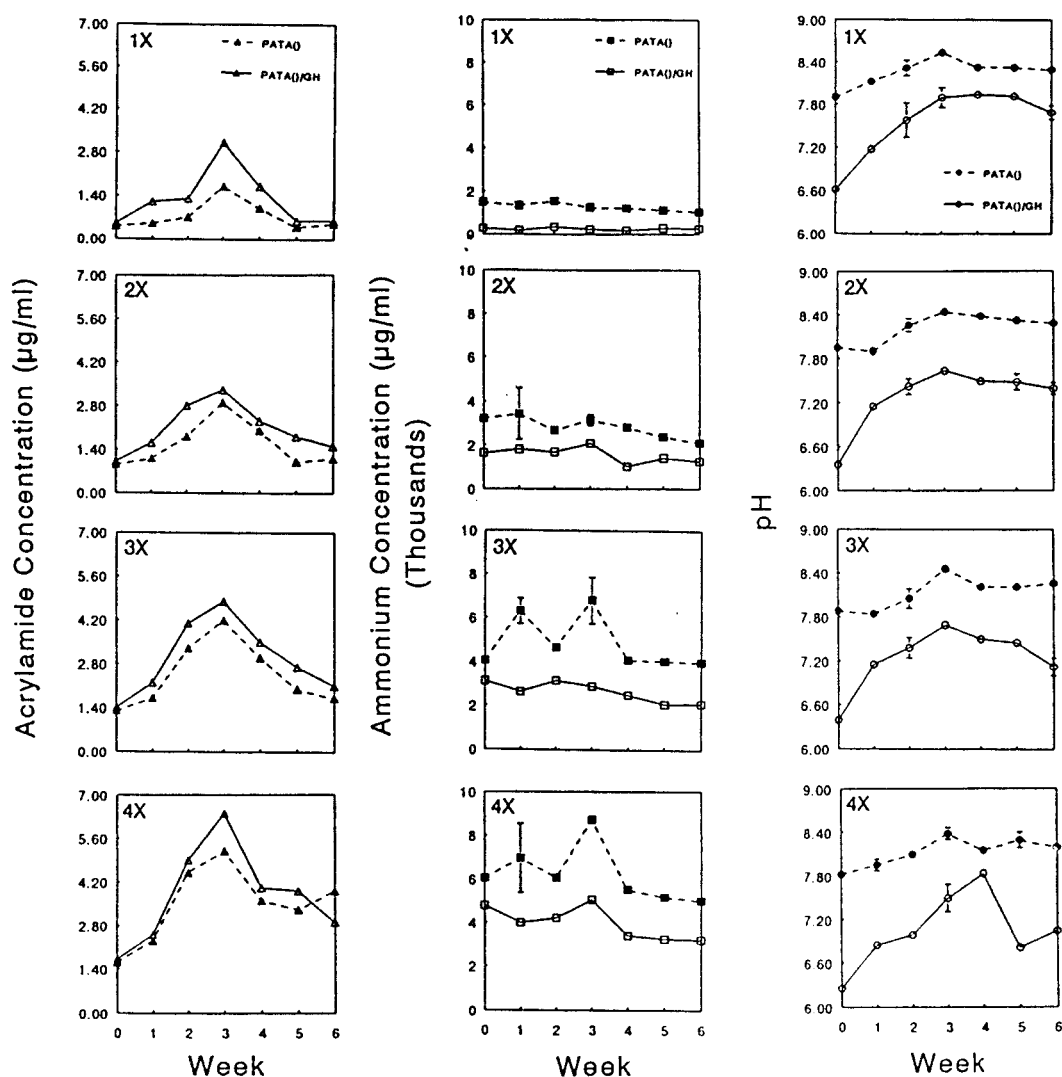


FIG. 4. Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in creek water and exposed to environmental (outdoor) conditions for 6 weeks.

TABLE 2

**Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Creek Water without or with a Glyphosate Herbicide (PATA/GH)**

Correlation pair	PATA( )				PATA( )/GH			
	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )
AC/NH <sub>4</sub>	-0.06	0.09	0.37	0.39	-0.13	0.32	0.21	0.24
AC/pH	0.66	0.56	0.60	0.43	0.43	0.70	0.79	0.62
NH <sub>4</sub> /pH	-0.31	-0.09	0.17	0.04	0.15	-0.04	-0.22	-0.19
AC/BA	0.68	—	—	-0.31	-0.28	—	—	-0.24
NH <sub>4</sub> /BA	-0.39	—	—	-0.32	0.24	—	—	-0.32
pH/BA	0.70	—	—	0.11	0.16	—	—	-0.27

*Note.* The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/NH<sub>4</sub>, acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH; NH<sub>4</sub>/pH, ammonium concentrations vs pH; AC/BA, acrylamide concentrations vs bacterial activity; NH<sub>4</sub>/BA, ammonium concentrations vs bacterial activity; pH/BA, pH vs bacterial activity. PATA(1 $\times$ ), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2 $\times$ ), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3 $\times$ ), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4 $\times$ ), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1 $\times$ )/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2 $\times$ )/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3 $\times$ )/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4 $\times$ )/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.

ammonium concentration for the various formulations displayed a decreasing trend throughout the study (Fig. 5). The pH generally did not change greatly with respect to treatment groups (Fig. 5). There was no specific trend for the bacteria that cultured over the 6-week study period. There was no fungal growth over the 6-week study period. Statistical analyses for the individual treatments revealed no strong linear correlation between the selected parameters (Table 3). All solutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation, and by the conclusion of the study all solutions were nearly transparent.

#### Natural Spring

Acrylamide concentrations increased until Week 4 and then remained elevated throughout the study. At Week 4, the acrylamide concentration for the various formulations was from 1.5 to 3.7 times the initial concentration (Fig. 6). The ammonium concentration for the various formulations displayed either a fluctuating or a decreasing trend throughout the study (Fig. 6). The pH for the various formulations was increased by Week 6 for all groups except for PATA(3 $\times$ )/GH (Fig. 6). There was no specific trend for the bacteria that cultured over the 6-week study period. There was no fungal growth over the 6-week study period. Statistical analyses for the individual treatments revealed no strong linear correlation between the selected parameters (Table 4). All solutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation, and by the conclusion of the study all solutions were nearly transparent.

#### Well R

Acrylamide concentrations increased until Week 4 and then remained elevated throughout the study. At Week 4, the acrylamide concentration for the various formulations was from

19.2 to 46.7 times the initial concentration (Fig. 7). The ammonium concentration for the various formulations displayed either a fluctuating or a decreasing trend throughout the study (Fig. 7). The pH for the various formulations was increased by Week 6 for all groups (Fig. 7). There was no specific trend for the bacteria that cultured over the 6-week study period. There was no fungal growth over the 6-week study period. Statistical analyses for the individual treatments revealed no strong linear correlation between the selected parameters (Table 5). All solutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation, and by the conclusion of the study all solutions were nearly transparent.

#### Well V

Acrylamide concentrations were not detected until Week 1 or 2 (Fig. 8). Acrylamide concentrations for PATA(1 $\times$ ) and PATA(2 $\times$ ) without and with GH were detected only on Weeks 2 and 3 with an increase from 0 to 2 times the original concentrations. The acrylamide concentration for PATA(3 $\times$ ) and PATA(4 $\times$ ) without and with GH was from 9 to 20 times the formulated concentration at Week 0. The ammonium concentration for the various formulations displayed a decreasing trend throughout the study (Fig. 8). Similar patterns were noted for pH between all groups (Fig. 8). There was no specific trend for the bacteria that cultured over the 6-week study period. There were no fungi that cultured over the 6-week study period. Correlation coefficients for the individual treatments revealed no strong linear correlation for the investigated parameters (Table 6). Upon formulation, the PATA solutions, regardless of formulation, had a homogeneous milky appearance, but after 6 weeks the solutions were nearly transparent.

#### Volatility

*Study 1.* The total amount of acrylamide in solutions of PATA(4 $\times$ ) and PATA(4 $\times$ )/GH increased by Day 6 of the

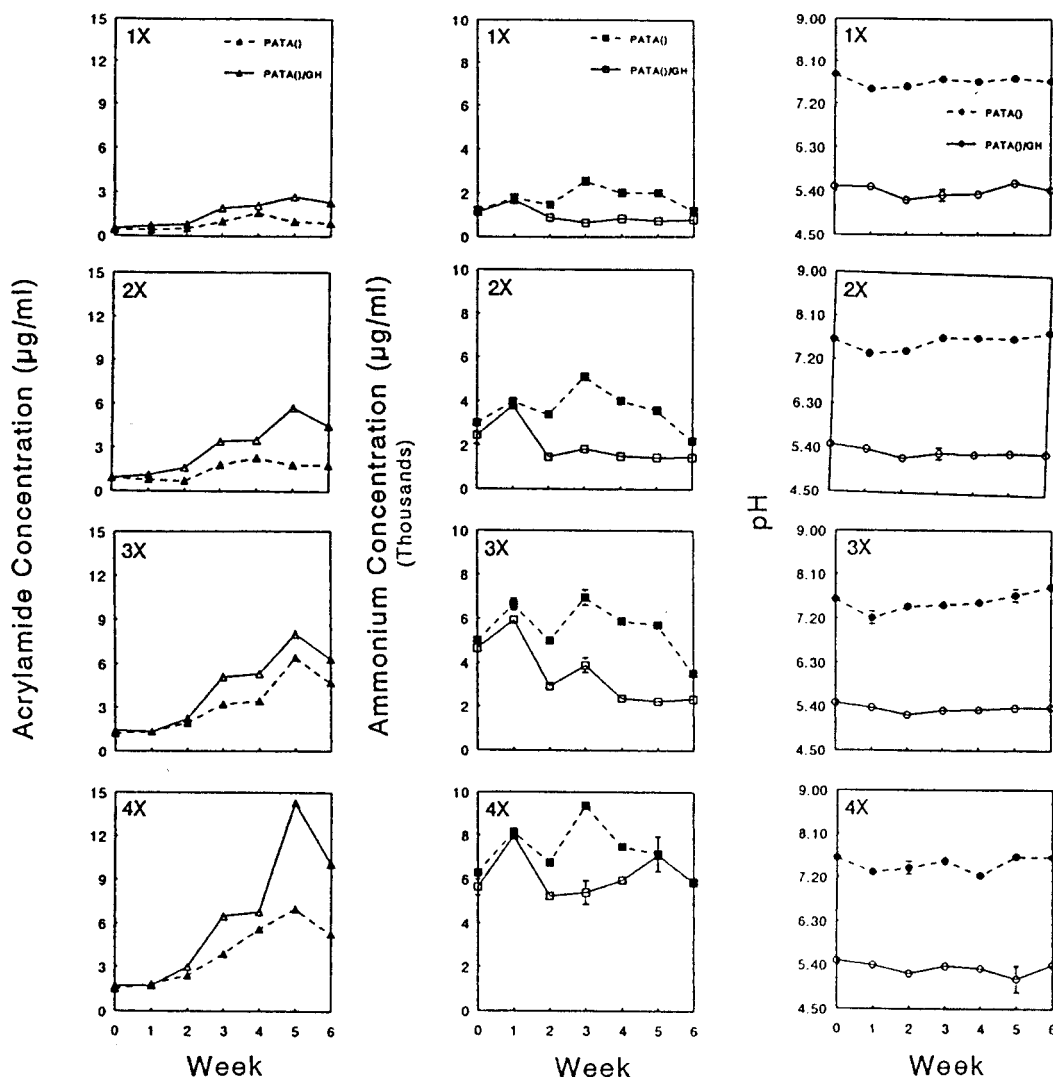


FIG. 5. Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in pond water and exposed to environmental (outdoor) conditions for 6 weeks.

TABLE 3

Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Pond Water without or with a Glyphosate Herbicide (PATA/GH)

Correlation pair	PATA()				PATA()/GH			
	(1x)	(2x)	(3x)	(4x)	(1x)	(2x)	(3x)	(4x)
AC/ $\text{NH}_4$	-0.52	-0.53	-0.74	0.06	0.37	-0.04	-0.14	-0.09
AC/pH	0.25	0.66	0.53	0.14	0.13	-0.09	-0.07	-0.32
$\text{NH}_4$ /pH	-0.21	-0.48	-0.58	-0.04	-0.08	-0.13	-0.12	0.13
AC/BA	-0.15	—	—	0.73	-0.53	—	—	-0.43
$\text{NH}_4$ /BA	-0.24	—	—	-0.15	-0.44	—	—	-0.33
pH/BA	0.15	—	—	0.20	0.23	—	—	0.29

Note. The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/ $\text{NH}_4$ , acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH;  $\text{NH}_4$ /pH, ammonium concentrations vs pH; AC/BA, acrylamide concentrations vs bacterial activity;  $\text{NH}_4$ /BA, ammonium concentrations vs bacterial activity; pH/BA, pH vs bacterial activity. PATA(1x), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2x), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3x), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4x), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1x)/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2x)/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3x)/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4x)/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.



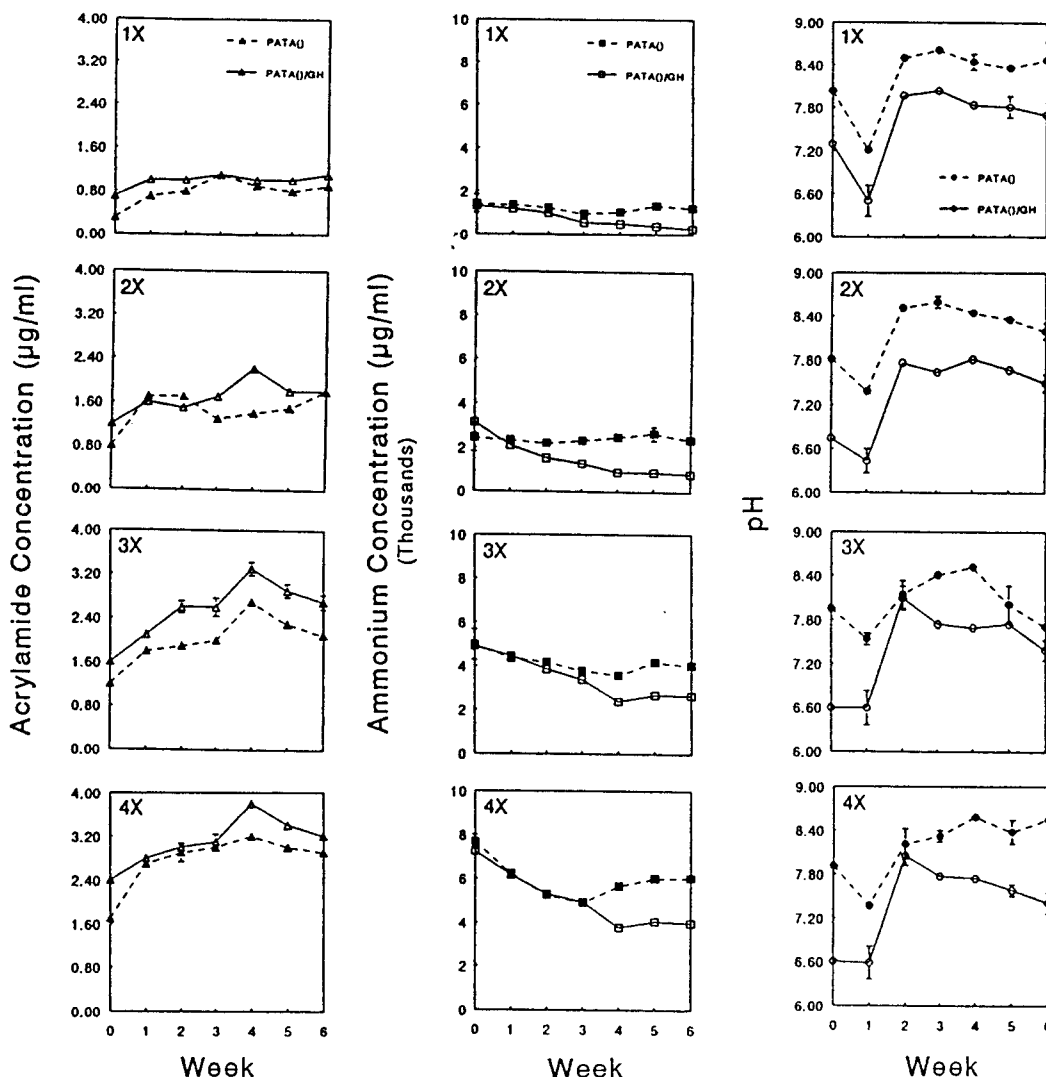


FIG. 6. Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in natural spring water and exposed to environmental (outdoor) conditions for 6 weeks.

study, while the total amount of acrylamide in solutions of acrylamide alone (control) decreased (Fig. 9). The total amount of ammonium was increased by Day 6 for solutions containing acrylamide alone (control) and PATA(4×)/GH, while solutions containing PATA(4×) fluctuated throughout the study (Fig. 9). The pH for the three solutions was lower by Day 6 (Fig. 9). (Note: Heavy rains occurred in the evening of Days 1 and 2, adding approximately 50 ml to each beaker over the 2-day period). None of the solutions reached dryness by the end of the 6-day study period. The volume within the beakers ranged from 100 to 33 ml.

**Study 2.** The total amount of acrylamide decreased in solutions of acrylamide alone (control) and PATA(4×) when dryness was reached (Day 5). In comparison, solutions of PATA(4×)/GH acrylamide concentration decreased until Day 4 and then rose to the original concentrations on Day 5 (Fig. 9).

The total amount of ammonium decreased through Day 5, when dryness was reached (Fig. 9). The pH for solutions of acrylamide alone (control) and PATA(4×)/GH showed no great change by Day 5, while the pH for solutions of PATA(4×) decreased until dryness was reached (Fig. 9). A light drizzle occurred in the evening of Day 1, but did not increase the volume in the beakers. Volume ranged from 100 to 0 ml.

#### Soil Columns

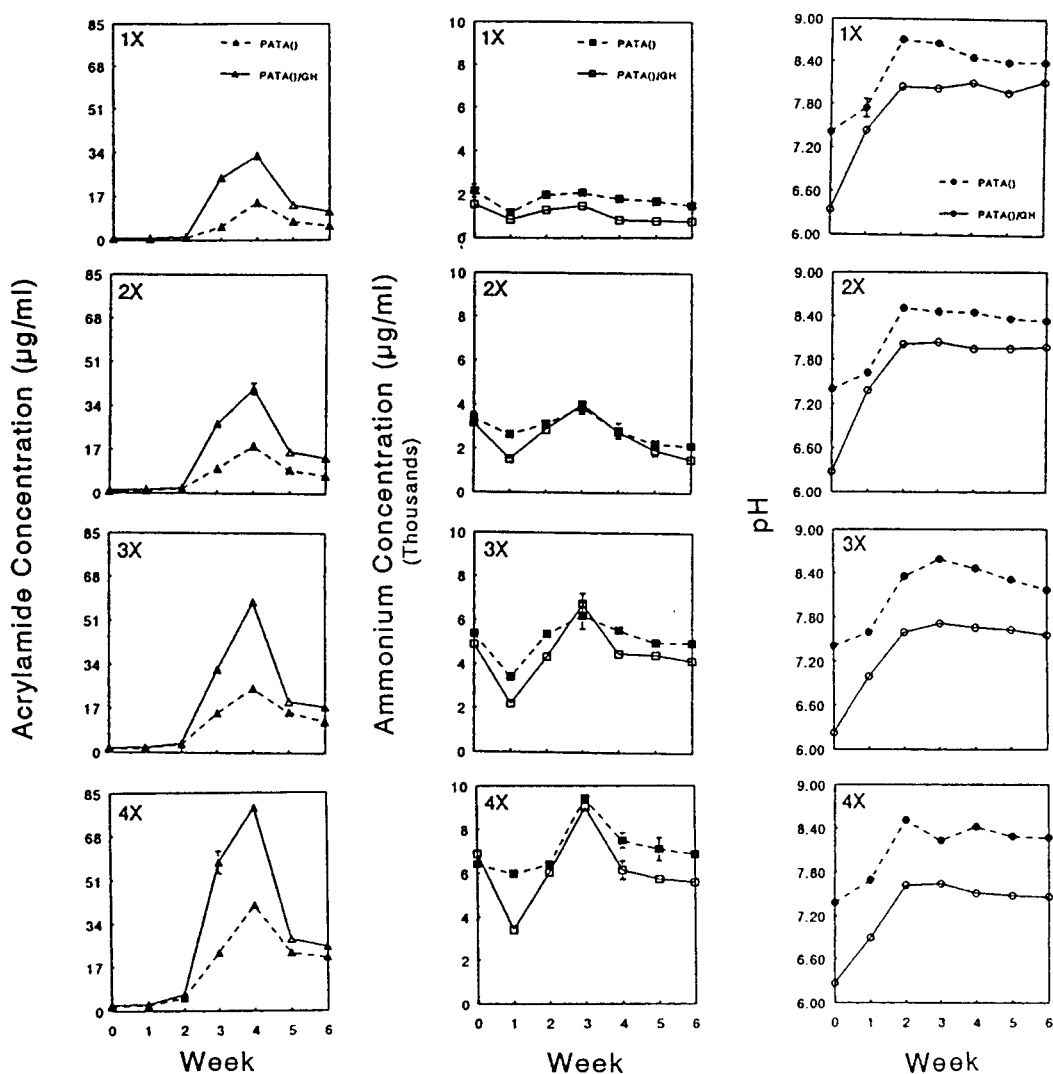
The results of the soil column study are given in Table 7. Acrylamide migrated the fastest through the columns filled with sand (97% recovery), then the Eudora sandy loam (73% recovery), and last Kahola silt loam (43% recovery). Acrylamide was detected in the sand columns after 2 days of continuous washing and appeared as a single peak. Acrylamide

TABLE 4

**Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Natural Spring Water without or with a Glyphosate Herbicide (PATA/GH)**

Correlation pair	PATA( )				PATA( )/GH			
	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )
AC/ $\text{NH}_4$	-0.33	0.01	-0.49	-0.67	-0.51	-0.76	-0.86	-0.85
AC/pH	0.40	0.10	0.43	0.30	0.25	0.55	0.57	0.56
$\text{NH}_4$ /pH	-0.28	-0.06	-0.27	-0.41	-0.56	-0.88	-0.59	-0.67
AC/BA	0.27	—	—	0.12	0.33	—	—	0.30
$\text{NH}_4$ /BA	0.04	—	—	0.50	-0.27	—	—	0.13
pH/BA	-0.33	—	—	-0.45	-0.42	—	—	-0.30

*Note.* The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/ $\text{NH}_4$ , acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH;  $\text{NH}_4$ /pH, ammonium concentrations vs pH; AC/BA, acrylamide concentrations vs bacterial activity;  $\text{NH}_4$ /BA, ammonium concentrations vs bacterial activity; pH/BA, pH vs bacterial activity. PATA(1 $\times$ ), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2 $\times$ ), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3 $\times$ ), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4 $\times$ ), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1 $\times$ )/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2 $\times$ )/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3 $\times$ )/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4 $\times$ )/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.



**FIG. 7.** Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in ground water (well R) and exposed to environmental (outdoor) conditions for 6 weeks.

TABLE 5

**Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Ground Water (Well R) without or with a Glyphosate Herbicide (PATA/GH)**

Correlation pair	PATA( )				PATA( )/GH			
	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )
AC/NH <sub>4</sub>	0.01	-0.07	0.41	0.53	-0.39	0.09	0.24	0.33
AC/pH	0.44	0.60	0.64	0.62	0.54	0.52	0.54	0.50
NH <sub>4</sub> /pH	0.14	-0.06	0.48	0.34	-0.57	-0.20	0.16	0.21
AC/BA	0.44	—	—	0.42	-0.48	—	—	-0.39
NH <sub>4</sub> /BA	-0.23	—	—	-0.20	-0.08	—	—	-0.14
pH/BA	0.46	—	—	0.64	0.12	—	—	-0.66

*Note.* The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/NH<sub>4</sub>, acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH; NH<sub>4</sub>/pH, ammonium concentrations vs pH; AC/BA, acrylamide concentrations vs bacterial activity; NH<sub>4</sub>/BA, ammonium concentrations vs bacterial activity; pH/BA, pH vs bacterial activity. PATA(1 $\times$ ), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2 $\times$ ), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3 $\times$ ), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4 $\times$ ), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1 $\times$ )/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2 $\times$ )/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3 $\times$ )/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4 $\times$ )/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.

was detected in the Eudora sandy loam and Kahola silt loam columns after 2.5 and 3 days of continuous washing, respectively. The concentration of acrylamide in the wash for these columns was lower than that observed in the wash of the sand columns and remained steady for the duration of the study.

#### Soil Boxes

Acrylamide was not detected in the runoff water from any of the soil boxes after 4 days of outdoor environmental exposure.

### DISCUSSION

This study examined three major questions: (1) Would polyacrylamide degrade to acrylamide under actual environmental conditions and would water quality (distilled-deionized, surface, or ground) accelerate or inhibit the degradation? (2) Would acrylamide concentrate as solutions evaporate? (3) Would polyacrylamide degrade to acrylamide when in contact with soil and would the acrylamide percolate through the soil or travel across the surface in the runoff? The results of this study indicated that acrylamide concentrations increased when the various formulations of PATA without and with GH were exposed to actual environmental conditions. These results were independent of the water source used to formulate the various solutions (i.e., distilled-deionized water, surface water, ground water). During the observation period, acrylamide traditionally reached its highest concentration between Week 3 and Week 5 for the various formulations. These data suggest that polyacrylamide degrades under environmental conditions, releasing acrylamide.

A major factor in environmental degradation in photolytic effects or energies. The energy from light can break chemical bonds, thereby, degrading polymers (Decker, 1989; Rabek, 1987; Grassie and Scott, 1985). Polyacrylamide is primarily

composed of C–C, C–H, and C–N bonds. The bond strengths of C–C, C–H, and C–N are approximately 340, 420, and 414 kJ/mol, respectively, and can be broken by wavelengths of 325, 250, and 288 nm, respectively (Crosby, 1976). The spectral radiation from the sun which reaches the earth's surface extends from the infrared (>700 nm), through the visible spectrum (400–700 nm), into the ultraviolet (4–400 nm). However, ozone absorbs all solar radiation below 286–300 nm (Crosby, 1976). Therefore, sunlight is capable of breaking a C–C bond but has minimal or no effect upon the C–H or C–N bond (Fig. 10).

Designing an exposure system to evaluate photolytic effects on PATA formulations and not volatility effects posed a challenge. Placing the various solutions in glass bottles with screw-top caps was not an appropriate exposure system. Glass produces a decrease in wavelength transmission by 350 nm and a wavelength cutoff at 300 nm. Therefore, glass bottles will only allow radiation energy capable of breaking a C–C bond to pass, but not energies capable of breaking a C–H or C–N bond. To correct this limitation, plastic wrap was placed over the mouth of each bottle. Plastic wrap has a decrease in wavelength transmission by 350 nm, but a wavelength cutoff of <250 nm. The use of plastic wrap increased the spectrum of radiation that could reach the PATA formulations. There was also no significant loss of solution volume with the use of plastic wrap as a cap for the 6-week period. Therefore, photolytic effects caused the polyacrylamide thickening agent, without or with the glyphosate-surfactant herbicide, to degrade to acrylamide.

The observed fluctuations in ammonium, pH, or bacterial growth may be the result of interactions between the three parameters within the test system. Ammonium was analyzed as a degradation by-product of acrylamide and polyacrylamide. The ammonium concentration increased in solutions formu-

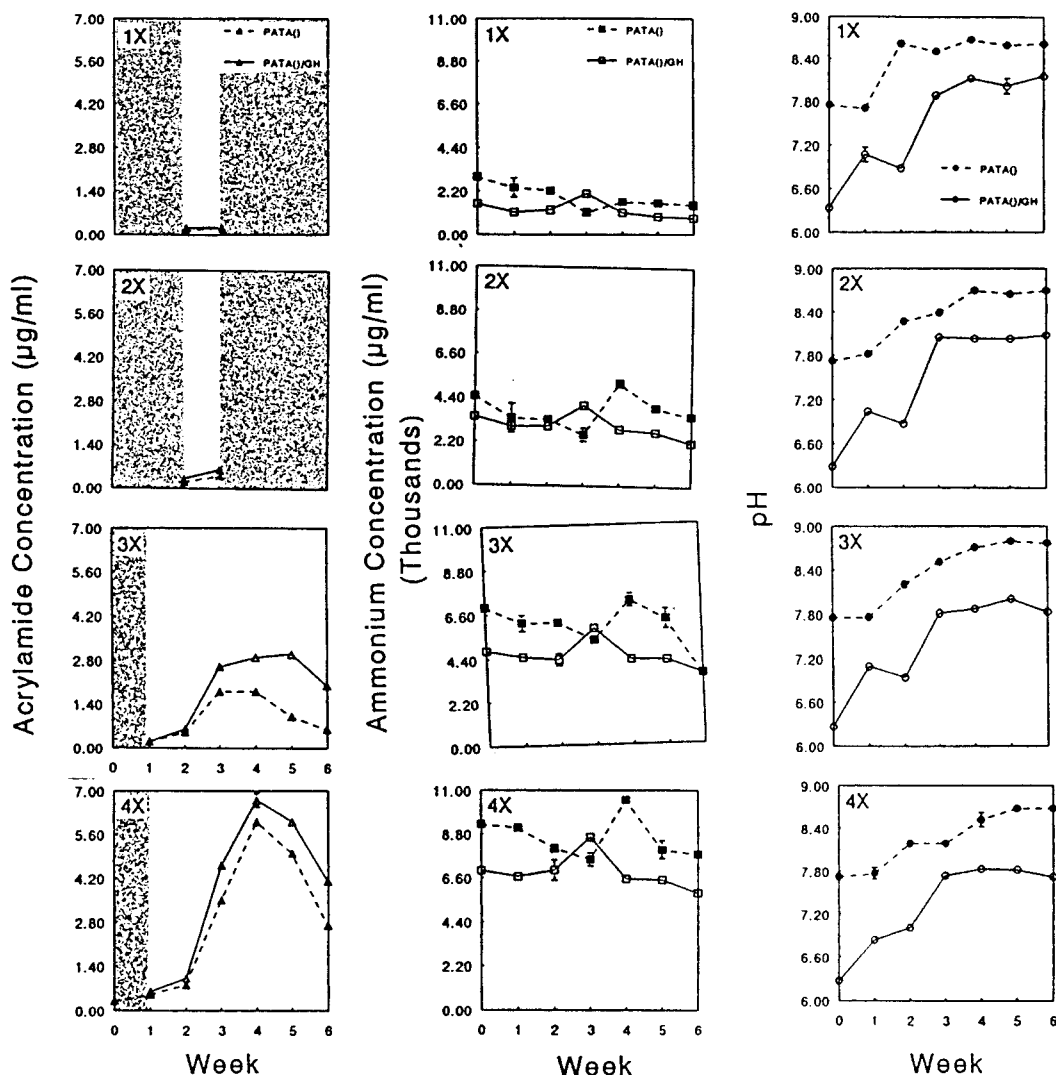


FIG. 8. Acrylamide concentrations, ammonium concentrations, and pH for four concentrations of a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in ground water (well V) and exposed to environmental (outdoor) conditions for 6 weeks.

lated with distilled-deionized water but fluctuated or decreased for those solutions formulated with surface water or ground water. pH was measured as an indicator of acrylamide and polyacrylamide degradation, based upon the potential release of ammonium or hydrogen. A statistical comparison, however, indicated no observable pattern between the concentration of ammonium and the corresponding pH for the solution. In addition, solutions were cultured for bacteria and fungus, to observe whether they might metabolically contribute to the degradation of the polymer or monomer. Bacteria produce ammonia as a by-product from the degradation of organic matter (Hobbie and Fletcher, 1988). This process is most efficient under neutral conditions (Alexander, 1961). Bacteria also utilize ammonia during protein synthesis (Willett, 1988). Therefore, bacterial degradation of organic matter to ammonia and its reuptake into bacterial protein could establish a steady-state environment based upon the nitrogen needs of the bacteria.

Essential to bacterial growth is a usable carbon source. High-molecular-weight surfactants are poorly degraded and create an inhibitory effect on local bacterial populations (Swisher, 1987). In this study, bacterial populations were observed one week and then absent the next. This absence may be due to the availability of a usable carbon source. Bacterial growth would be dependent upon the effects of photolysis to break down the polymer to a size that can be used.

Analysis of the two volatility studies indicate similar but different results. The objective of the study was to examine the environmental fate of acrylamide in solution as the water evaporated, similar to that of a pond or creek bed drying up during the summer months. Heavy rains occurring during the night of Days 1 and 2 of the study required the study to be repeated in order to acquire samples that mimicked dry summer conditions. The solutions from both studies were analyzed for 7 days, the main difference between the two studies being that

TABLE 6

**Pearson Correlation Coefficients ( $N = 21$ ) for Various Combinations of Acrylamide Concentrations ( $\mu\text{g/ml}$ ), Ammonium Concentrations ( $\mu\text{g/ml}$ ), pH, and Bacterial Activity of a Polyacrylamide Thickening Agent (PATA) Formulated in Ground Water (Well V) without or with a Glyphosate Herbicide (PATA/GH)**

Correlation pair	PATA( )				PATA( )/GH			
	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )	(1 $\times$ )	(2 $\times$ )	(3 $\times$ )	(4 $\times$ )
AC/ $\text{NH}_4$	-0.35	-0.57	-0.16	0.15	0.66	0.73	0.00	-0.04
AC/pH	0.28	0.05	0.69	0.76	-0.05	0.18	0.91	0.93
$\text{NH}_4$ /pH	-0.59	0.06	-0.12	-0.23	-0.27	-0.25	-0.16	-0.01
AC/BA	-0.24	—	—	0.12	-0.47	—	—	0.13
$\text{NH}_4$ /BA	-0.19	—	—	-0.57	-0.47	—	—	-0.48
pH/BA	0.25	—	—	0.30	0.31	—	—	0.28

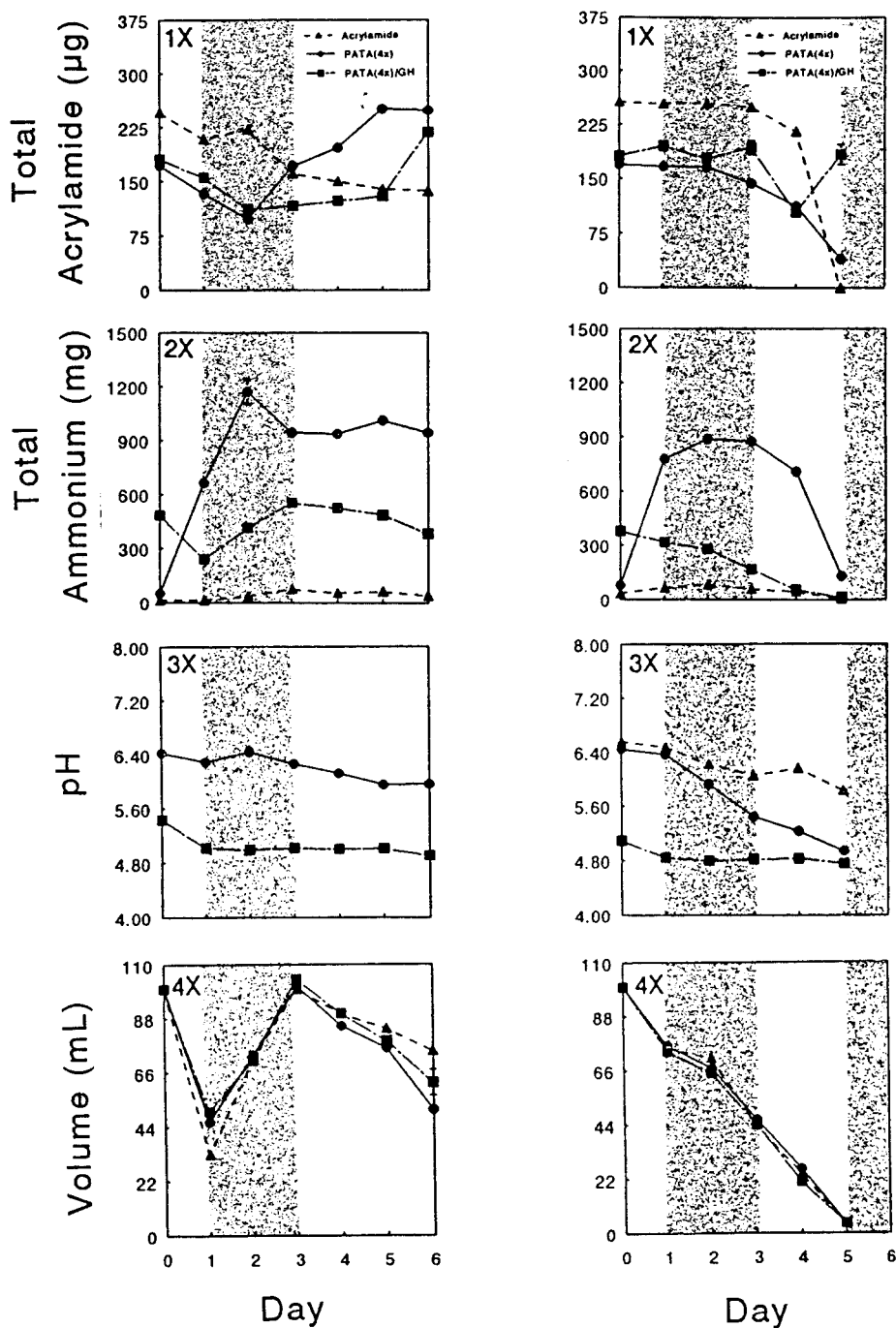
*Note.* The formulations were placed in glass bottles with plastic wrap covering the mouth of the bottles and exposed to the environment for 6 weeks. AC/ $\text{NH}_4$ , acrylamide concentrations vs ammonium concentrations; AC/pH, acrylamide concentrations vs pH;  $\text{NH}_4$ /pH, ammonium concentrations vs pH; AC/BA, acrylamide concentrations vs bacterial activity;  $\text{NH}_4$ /BA, ammonium concentrations vs bacterial activity; pH/BA, pH vs bacterial activity. PATA(1 $\times$ ), 1.167 liters polyacrylamide thickening agent/378.5 liters water; PATA(2 $\times$ ), 2.334 liters polyacrylamide thickening agent/378.5 liters water; PATA(3 $\times$ ), 3.500 liters polyacrylamide thickening agent/378.5 liters water; PATA(4 $\times$ ), 4.667 liters polyacrylamide thickening agent/378.5 liters water; PATA(1 $\times$ )/GH, 1.167 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(2 $\times$ )/GH, 2.334 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(3 $\times$ )/GH, 3.500 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water; PATA(4 $\times$ )/GH, 4.667 liters polyacrylamide thickening agent/378.5 liters water + 45.4 g isopropylamine salt of glyphosate/378.5 liters water.

the solutions in Study 1 did not evaporate to dryness by the end of the study period while those in Study 2 did. The volatility of acrylamide in solution is low (Windholz, 1989). This can be observed throughout Study 1 and the majority of Study 2. However, in Study 2 when the solutions had evaporated to <25% their original volume, the concentration of acrylamide dropped to nondetectable levels with the exception of PATA/GH, which rose. Therefore, acrylamide does not concentrate as the volume of the solution decreases but either volatilizes or degrades (i.e., ammonium). Ammonium was detected throughout Study 1 and most of Study 2, but not at the conclusion of Study 2, when the solutions had nearly evaporated. There was no correlation between the concentration of acrylamide or ammonium and the pH of the solution.

The results of the soil mobility study demonstrated that acrylamide was able to migrate through the three selected soil types. Acrylamide had its highest mobility through sand, followed by Eudora sandy loam and Kahola silt loam. These results agreed with those of Lande *et al.* (1979). The mobility of a compound through soil depends on (1) the physical and chemical characteristics of the compound, (2) the physical and chemical characteristics of the soil, and (3) application factors (Anonymous, 1986). Physical and chemical characteristics of the compound include solubility, volatility, soil absorption, and soil dissipation. Acrylamide is highly soluble in water, 212 g/100 ml water, and has low volatility in aqueous solution (Windholz, 1989). Soil adsorption of acrylamide is unknown. Acrylamide has a half-life of 18 to 45 hr at 22°C for soils containing 25 mg acrylamide/kg (Lande *et al.*, 1979). However, these half-lives depend upon climatic conditions and temperature. Physical and chemical characteristics of the soil include clay and organic carbon content, soil texture, soil structure, porosity, soil moisture, and depth to ground water. Clay

minerals/clay content increase the cation exchange capacity and surface area of soil, thereby increasing the adsorption capacity. Eudora sandy loam has 9% clay and Kahola silt loam has 37% clay (Rice and Rogers, 1986). Soils high in organic carbon absorb chemicals and, therefore, inhibit their movement into ground water. Eudora sandy loam has 2.74% organic carbon and Kahola silt loam has 37% organic carbon (Rice and Rogers, 1986). Leaching is more rapid and deeper in coarse or light-textured sandy soils than in fine or heavy-textured clayed soils (Anonymous, 1986). Soil structure is affected by its texture and percentage organic carbon. Kahola silt loam would allow the chemical to seep unimpeded through the seams between the aggregates (Anonymous, 1986). Porosity is not known for these two soils. The effect soil moisture (level of wetness) has on the mobility of acrylamide was only evaluated on columns at 100% saturation. The mobility of acrylamide with respect to depth to ground water was evaluated from the surface to 4 feet below the surface. Although application factors for chemicals include climatic conditions, rate of application, timing and method of application, irrigation and cultivation practices, spillage/disposal, and nonagricultural application, these parameters were not investigated because of their wide variability.

The same principles that define the mobility/leachability of a chemical through a particular soil type are also applicable for predicting the probability of the chemical in surface runoff. The results of the soil runoff study indicated that acrylamide was not detected in the runoff. The lack of acrylamide in the runoff may be due to polyacrylamide binding to insoluble particles, which protected the PATA from environmental degradation. Another explanation may be that the combination of soil and sunlight could have degraded the polyacrylamide to by-products other than acrylamide.



**FIG. 9.** Volatility of acrylamide concentrations, ammonium concentrations, pH, and volume for an acrylamide standard (control) and a polyacrylamide thickening agent without and with a glyphosate-surfactant herbicide formulated in distilled-deionized water and exposed to outdoor conditions for 6 days. The shaded area denotes nights in which it had rained.

## CONCLUSION

It has been demonstrated that under artificial environmental conditions, polyacrylamide may degrade to acrylamide. In an agricultural setting, the degradation of polyacrylamide to acrylamide creates a potential environmental/health hazard (i.e., contamination of surface water and/or ground water systems).

Under environmental (outdoor) conditions acrylamide levels in solutions of PATA increased when formulated in distilled-deionized water, surface water, or ground water samples without and with GH, suggesting that polyacrylamide degraded to acrylamide. Ammonium concentrations were increased in distilled-deionized water but varied or decreased in the surface water or ground water samples. pH was not affected. All so-

TABLE 7

Acrylamide Concentration ( $\mu\text{g/ml}$ ) from Soil Columns Containing Three Different Types of Soil after the Application of a 100-ml Polyacrylamide Thickening Agent Formulated at 4.667/378.5 Liters Water Plus a Glyphosate Surfactant Herbicide at 1.4 kg Isopropylamide Salt of Glyphosate and Leached with 1 mM Sodium Sulfate for 1 Week

	Sand		Sandy loam		Silt loam	
Column 1 <sup>a</sup>	0.315 $\pm$ 0.003		0.064 $\pm$ 0.006		0.041 $\pm$ 0.001	
Column 2 <sup>a</sup>	0.253 $\pm$ 0.026		0.058 $\pm$ 0.005		0.048 $\pm$ 0.005	
Average	0.273 $\pm$ 0.021		0.061 $\pm$ 0.004		0.046 $\pm$ 0.001	
	C1	C2	C1	C2	C1	C2
Total volume of wash (ml) through each column <sup>b</sup>	964	998	1623	3325	1279	2602
Total acrylamide ( $\mu\text{g}$ ) recovered from each column	157.4	140.5	73.2	151.8	39.3	93.2
% Recovery	102.7	91.7	47.7	99.1	25.6	60.8

<sup>a</sup> The average acrylamide concentration was based on the daily acrylamide concentration, starting with the first day acrylamide is detected.  
<sup>b</sup> No acrylamide was detected in the first 400–550 ml of sodium sulfate wash.

lutions had a homogeneous milky appearance at the beginning of the study period, independent of formulation (water or chemical) and by the conclusion of the study, all solutions were nearly transparent. The volatility of acrylamide was very low, independent of formulation, except when solutions were near dryness. Trends for the ammonium ion concentrations and pH resembled those observed in artificial and outdoor studies. Acrylamide leached the fastest from columns containing sand only, followed by columns containing Eudora sandy loam, then Kahola silt loam. A single acrylamide peak after 2 days of washing was observed from the sand columns. Acrylamide

concentrations were detected in the other two soil types after 2 days of washing, wherein it was leached off at a constant rate over the next 5 days. Acrylamide could not be detected in the runoff water from any of the soil boxes after 4 days of outdoor environmental exposure.

ACKNOWLEDGMENT

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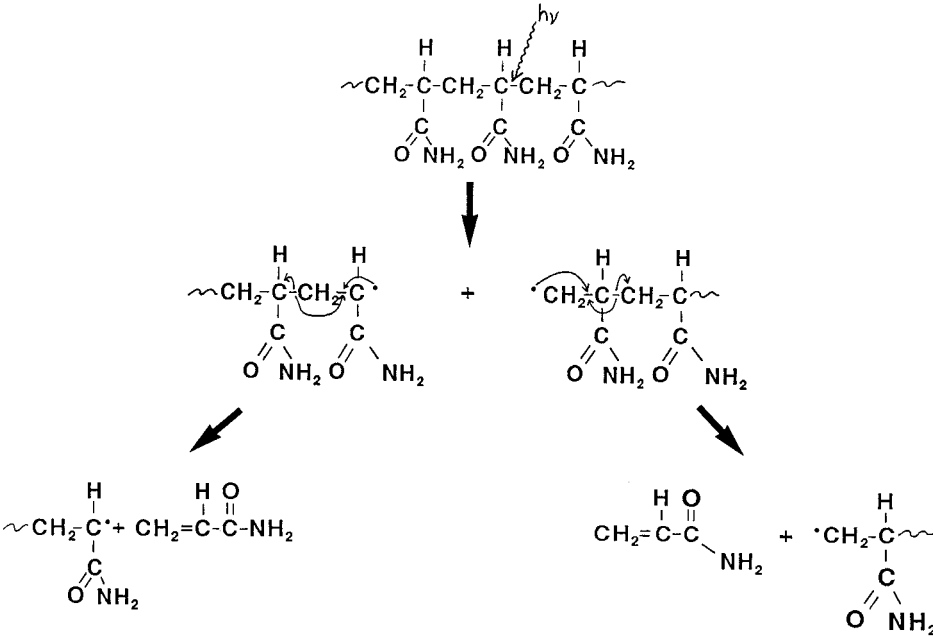


FIG. 10. Photolysis of polyacrylamide.

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