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Health Hazard Assessment for Native Americans Exposed to the Herbicide Fluridone via the Ingestion of Tules at Clear Lake, California, USA

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ABSTRACT

This study addresses concerns expressed by Native Americans regarding exposure via the consumption of aquatic vegetation to the herbicide fluridone (active ingredient) used by the California Department of Food and Agriculture (CDFA) Hydrilla Eradication Program in Clear Lake, California. In 2005, the Department monitored lakeshore vegetation, water, and sediment at four locations, before and after seasonal applications of fluridone. Subchronic and chronic exposures were evaluated, and hazard quotients calculated for a worst-case exposure (WCE) scenario. Ingestion rates and other exposure factors were developed in public meetings with tribal members. Environmental sampling found fluridone present at extremely low levels in tule vegetation, water, and sediment. Exposures were four times greater in subchronic timeframes than chronic timeframes; however, hazards were less due to the 25-fold larger reference dose (RfD) used for subchronic calculations: $RfD_{(subchronic)} = 2.0$ mg/kg-day, $RfD_{(chronic)} = 0.08$ mg/kg-day. Conservative, child, total daily ingestion (TDI) doses were calculated to be 8.3×10^{-5} mg/kg-day (subchronic) and 2.1×10^{-5} mg/kg-day (chronic). Hazard quotients (HQ) for subchronic and chronic exposures were on the order of 10^{-5} and 10^{-4}, respectively, indicating that at current application regimes, there is little to no hazard of adverse effects from fluridone exposure via ingesting Clear Lake tules.

Key Words: Native American, exposure, hazard, fluridone, tules.

INTRODUCTION

Aquatic herbicides are often the primary tool for the control and eradication of invasive aquatic weeds. Over the last 10 years, the California Department of Food and

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Agriculture (CDFA) *Hydrilla* Eradication Program has been increasingly successful in their efforts to eradicate *Hydrilla* from Clear Lake, California, by implementing a regime of survey followed by treatment with the fluridone-based aquatic herbicide Sonar©. Fluridone acts by inhibiting the formation of carotene in susceptible plants. Without carotene sunlight rapidly degrades chlorophyll, and the plant is unable to maintain photosynthesis (SePRO 2007). A fluridone-based herbicide was selected for use because it is extremely effective in controlling *Hydrilla* and is relatively nontoxic, quickly dropping out of the water column and adhering to sediment particles and organic material within sediment (McLaren Hart 1995).

Native American tribes around Clear Lake are concerned about exposure to aquatic herbicides because tribal members, young and old, eat emergent tule vegetation (*Schoenoplectus californicus* formerly *Scirpus californicus*) directly from the lake. Between April and June, tules are edible, and can be picked, peeled, and eaten raw. Many local tribal members carry salt shakers in their pockets to enhance this readily available snack. At times, tribal members will fill a trailer with large quantities of tules, then drive around the lake, stopping to share (consume) them with friends and family. In 2005 the CDFA began a study to investigate environmental concentrations of fluridone in tule vegetation, water, and sediment along tribal shoreline property, and conduct a human health hazard assessment for exposure to fluridone from the ingestion of tules.

Study Area

Clear Lake is the largest natural freshwater lake completely located within California. It rests between the inner and outer Coast Mountain Ranges of Northern California, at an elevation of 402 meters. Although the lake is large, comprising some 17,806 hectares of surface water, it is relatively shallow, with an average depth of 7.9 meters, and a maximum depth of 13.7 meters (Lake County 2003). The lake has 112 km of shoreline, and a storage capacity of approximately 386 million $m^3$of water. Fifty percent of the lake inflow is from creeks, the largest of which is Rodman Slough at the north end of the lake. Other water inputs include groundwater flows and rain. The area is volcanic in nature, with lake sediments largely comprised of fine alluvial sand and silt that washed down from the watershed into the lake, and organic matter from decomposing aquatic organisms.

Study Design

During public community scoping meetings, Native Americans from four Pomo Tribes (Big Valley, Robinson, Elem, and Upper Lake) were asked to describe their tule harvest and consumption practices. In attendance at these meetings were the manager of the Big Valley Rancheria Analytical Laboratory, the Assistant Environmental Program Director from Big Valley, 4 members of the Native American Basket Makers Association, and approximately 14 other tribal members. The information gathered during these meetings was used to select sampling sites and develop exposure factors for worst-case exposure calculations.

In collaboration with the tribes, three sampling sites were selected for investigation on or adjacent to tribal property: Site 1 (Elem tribal property), Site 2 (Robinson Rancheria), and Site 3 (Big Valley Rancheria) (Figure 1). Although the ingestion

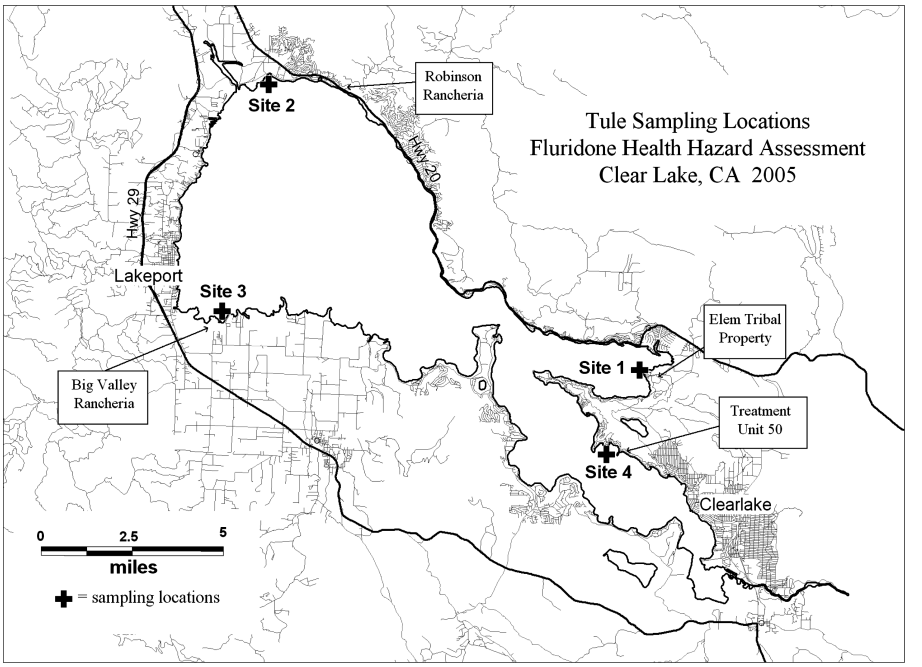

Figure 1. Environmental sampling locations at Clear Lake, California. Tules were collected from all four sites, with water and sediment collected from Site 3 and Site 4. Native American tribal shoreline properties are identified at or adjacent to the sampling sites. Unit 50 was the only site actively treated during this assessment.

exposure pathway was complete at these sites, the CDFA did not apply fluridone in either year 2004 or 2005 due to the lack of *Hydrilla* findings at these sites. Sampling Site 4 (Treatment Unit 50) was added to maximize the potential for tule exposure and uptake of fluridone from year 2005 herbicide treatments. Sampling events were scheduled before and after the first herbicide application of the season to provide baseline (pre-application) and post-treatment datasets.

Conservative, thorough, estimates of fluridone exposure were undertaken by sampling tule vegetation from two spots, at each of the four sampling locations, and sampling water and sediment from two spots at Sites 3 and Site 4 only. There were no "clean" sites to use as traditional reference sites, as the entire lake had been treated with fluridone for 10 years prior to the study.

A total of 2 trip blanks, 14 edible-tule, 16 whole-tule, 16 water, and 12 sediment samples were collected in the course of this investigation (Table 1). All sampling locations were mapped using geographic positioning system (GPS) technology (Figure 1).

Tules are edible only 3 months per year, from April through June. Because the harvest and consumption of tules is a cultural tradition, this exposure is likely to continue throughout a tribal member's lifetime. The ingestion exposure data were therefore evaluated in two ways: as a subchronic exposure (with intakes averaged

Table 1. Aggregated lake-wide environmental sample statistics for fluridone detection in tules, sediment, and water (recently treated and untreated areas combined).

Sample type	No. samples lake-wide	Number non-detect	No. fluridone detections	Fluridone min. detection (ppb)	Fluridone max. detection (ppb)
Vegetation edible	14	13	1	2.2	2.2
Vegetation whole	16	14	2	2.9	3.4
Water	18	2	16	0.18	0.29
Sediment (wet wt)	11	0	11	2.3	65.0

over a 91-day period), and as a chronic, lifetime exposure (with intakes averaged over 365 days/year for 70 years).

HUMAN HEALTH HAZARD ASSESSMENT

Health hazard assessments are used to determine if a particular chemical poses significant hazards to human health and, if so, under what circumstances. This study used the U.S. Environmental Protection Agency's (USEPA's) *Risk Assessment Guidance for Superfund (RAGS), Volume I, Human Health Evaluation Manual, Part D* (USEPA 1989) process. The contribution of fluridone from the ingestion of tules, and incidental amounts of sediment and water are evaluated by this hazard assessment. Also in keeping with international hazard evaluation methods (Cheng and Gobas 2007; Caldas and Souza 2004; WHO 1997), this hazard assessment compared total daily intakes (TDI) of fluridone with an identified acceptable daily intake (USEPA 1984).

A highly conservative "worst-case" exposure scenario (WCE) was evaluated for each time frame (subchronic and chronic). WCE calculations used the largest ingestion rates estimated by tribal members, the highest environmental concentrations detected, and the highest estimated exposure frequencies to calculate an average daily dose (ADD) for child and adult receptors.

HAZARD ASSESSMENT

Fluridone has been identified as the chemical of concern for this study. Fluridone is not known to be a carcinogen. Mutagenicity and cancer studies reviewed by the USEPA *Tolerance Registration Eligibility Decision* (USEPA 2004) found fluridone to be negative for inducing mutations or increasing tumor incidence in all guideline studies. Key research studies with rats have identified the following noncarcinogenic effects: Glomerulonephritis, atrophic testes, eye keratitis; decreased body weight and organ weights (USEPA 1987; Elanco 1980). Rat metabolism studies showed no bioaccumulation of fluridone, however, fluridone residues were found to be present in exposed livestock tissue (USEPA 2004).

The Medical Toxicology Branch of the CALEPA, Department of Pesticide Regulation (CALEPA 2000), evaluated all fluridone studies conducted by the Lilly Research

Laboratory, (Greenfield, IN), and found no adverse effects in chronic, reproduction, or teratology studies (CALEPA 2000). A possible adverse effect of increased skin fibrosarcomas was detected in mouse oncogenicity studies. In a review of fluridone exposure studies, (CALEPA 2000) found no adverse effects in chronic dog toxicity studies, rat reproduction studies, rat teratology studies, rabbit teratology studies, gene mutation studies, chromosome effects studies, or DNA damage studies, but did identify a possible increase in fibrosarcomas occurrence in high-dose female mice.

EXPOSURE ASSESSMENT

Methodology

The ingestion of emergent vegetation and the incidental ingestion of associated sediment and lake water were evaluated in the exposure assessment. The concentration of fluridone in these items, the duration of exposure, and the pattern of exposure were defined. Average daily doses (ADD) were calculated for each item separately, and collectively as a total daily ingestion dose (TDI). Dermal and inhalation exposures were not evaluated.

Edible new shoots and root bulbs emerge in late spring in some but not all tules. Both an edible portion and a homogenized whole-tule sample were collected from two spots at each of the four sampling locations. Edible-tule samples were prepared by peeling away outer reeds until a starchy white inner shoot was revealed. Fifty to 100 grams of edible shoots and small edible tubers (that pulled free with the stalk) were placed into new 1-quart Mason jars and stored in coolers on blue ice until transport to the CDFA laboratory for analysis. Whole-tule samples were prepared by cutting tules into 2.5 cm sections. Fifty to 100 grams of whole-tule pieces were placed in 1-gallon Ziplock bags along side edible samples. Tribal members were present during tule sampling to ensure that edible samples were representative of what the Native Americans ate. Vegetation and sediment samples were analyzed for fluridone by the GC/MS and LC/MS methods developed by the CDFA Center for Analytical Chemistry (CDFA laboratory) (CDFA 2006).

Water samples were collected by holding 250-ml plastic HPLC sample bottles upside down, 15 cm below surface before righting the container. Containers were capped and wrapped with parafilm. The CDFA laboratory analyzed water samples for fluridone using an enzyme-linked immuno-assay (ELISA) method, brand name FasTEST™ (SEPRO 2006).

Fluridone confirmation analysis by GC/MS method was not performed specifically for this study, but relied on positive GC/MS results from 4 years of previous *Hydrilla* Eradication Program sampling at Clear Lake. Water sampling protocols are described in the CDFA 2004 Sample Analysis Plan (SAP).

The top 8 to 10 cm of sediment was collected by hand pressing new 15-cm acetate sleeves into lakebed sediments immediately adjacent to vegetation sample points at each of two spots, at Site 3 (Big Valley Rancheria) and Site 4 (Treatment Unit 50). In cases where vegetation formed a mat too dense to penetrate, sediment was collected within a 0.6-meter radius of the vegetation sample point, or a soil sampling hammer was used. 50–100 grams of sediment per sample was transferred from the acetate

sleeves into new factory clean Mason jars. Sediment samples were also analyzed for fluridone by methods developed by the Center for Analytic Chemistry (CDFA-CAC 2006).

QA/QC

Quality assurance trip blanks were prepared for each of two sampling events in the office prior to field activities. Laboratory grade HPLC water was poured into triple-rinsed factory sterilized sample bottles, capped, and wrapped with parafilm. Trip blanks were place in sample collection coolers, and loaded into the field truck. At Site 3, duplicate sediment and water samples were collected during pre-treatment sampling. Due to the difficulty of obtaining enough edible-tule material to analyze, no duplicate vegetation samples were collected.

Appropriate laboratory QA/QC including instrument calibration, and use of reference toxicants and spikes was conducted by the Center for Analytical Chemistry (CAC) in accordance with good laboratory practices (GLP). The "Limit of Detection" for fluridone in vegetation samples, using a CAC developed LC/MS method was 2 ppb; and the "Limit of Quantitation" was 5 ppb (CDFA-CAC 2006). The "Limit of Detection" for fluridone in water samples using the ELISA method was 0.1 μg/liter (ppb); the "Limit of Quantitation" was 0.3 ppb (CDFA 2005). The "Limit of Detection" for fluridone in sediment samples analyzed by the LC/MS method was 0.5 ppb. The "Limit of Quantitation" was 1.7 ppb.

Exposure Factors

Exposure factors used in the calculation of average daily dose were: food, water, or sediment concentration (FdC, WC, and SC), ingestion rate (IR), exposure frequency (EF), exposure duration (ED), body weight (BW), and averaging time (AT) (Table 2).

For this worst-case exposure assessment, maximum detected fluridone concentrations from environmental samples of tule vegetation, water, and sediment were used. Other exposure factors were derived from the updated USEPA *Exposure Factors Handbook* (EFH) (USEPA 1997), the USEPA *Risk Assessment Supplemental Guidance for "Standard Default Exposure Factors"* (USEPA 1991), interviews with tribal members, and consultations with California Department of Health Services risk analysts (Table 2).

Although fluridone was detected in an edible-tule sample at 2.2 ppb, the larger, maximum detected concentration from whole-tule samples of 3.4 ppb was used as the concentration in food for "worst-case" exposure calculations. Similarly, the value used for water fluridone concentration was 0.294 ppb, the highest value detected in 16 water samples. The value used for sediment fluridone concentration was 65 ppb, the highest detected value of 11 sediment samples (Tables 1 and 3).

Estimates of the amount of tules ingested daily varied widely among tribal members. The *Exposure Factors Handbook* (USEPA 1997) found a realistic mean for total intake of vegetable produce (normalized to body weight) to be 373 g/day for the adult and 49 g/day for the child. The "worst-case" exposures evaluated in this study used a high, interview derived ingestion rate of 710 g/day for the adult, and 355 g/day (half the adult amount), for the child. Conservative, incidental water ingestion rates of 50 ml/day, and 25 ml/day for the adult and child, respectively, and

Table 2. Exposure factors used in calculating average daily dose (ADD), and hazard quotients (HQ) for worst-case exposure scenario receptors.

Receptor	[Conc] (ppb)	Ingestion rate (g/day)	Exposure frequency (days/yr)	Exposure duration (yrs)	Body weight (kg)	Averaging time (days)	Reference dose (mg/kg-day)	Average daily dose (mg/kg-day)	Hazard quotient (unitless)
				TULE					
SUB-CHRONIC									
Child WCE	3.4	355	91	1	15	91	2.0	8.0E-05	4.0E-05
Adult WCE	3.4	710	91	1	70	91	2.0	3.4E-05	1.7E-05
CHRONIC									
Child WCE	3.4	355	91	6	15	2,190	0.08	2.0E-05	2.5E-04
Adult WCE	3.4	710	91	70	70	25,550	0.08	8.6E-06	1.1E-04
				SEDIMENT					
SUB-CHRONIC									
Child WCE	65	0.5	91	1	15	91	2.0	2.2E-06	1.1E-06
Adult WCE	65	1.0	91	1	70	91	2.0	9.3E-07	4.6E-07
CHRONIC									
Child WCE	65	0.5	91	6	15	2,190	0.08	5.4E-07	6.8E-06
Adult WCE	65	1.0	91	70	70	25,550	0.08	2.3E-07	2.9E-06
				WATER					
SUB-CHRONIC									
Child WCE	0.3	25	91	1	15	91	2.0	4.9E-07	2.5E-07
Adult WCE	0.3	50	91	1	70	91	2.0	2.1E-07	1.1E-07
CHRONIC									
Child WCE	0.29	25	91	6	15	2,190	0.08	1.2E-07	1.5E-06
Adult WCE	0.29	50	91	70	70	25,550	0.08	5.3E-08	6.6E-07

Table 3. Maximum detected concentrations of fluridone in tule vegetation, sediment, and lake water samples, for each sampling location, pre-, and post-herbicide treatment.

Location	Spot	Edible-tule max.value detected (ppb)		Whole-tule max. value detected (ppb)		Sediment max. value detected (wet wt) (ppb)		Water max. value detected (ppb)	
Treatment		*Pre-	**Post-	Pre-	Post-	Pre-	Post-	Pre-	Post-
Elem (Site 1)	1	ND	ND	ND	ND	—	—	—	—
	2	ND	ND	ND	ND	—	—	—	—
Robinson (Site 2)	1	—	ND	ND	2.9	—	—	—	—
	2	ND	ND	ND	ND	—	—	—	—
Big Valley (Site 3)	1	—	ND	ND	ND	35.2	65	0.23	0.23
	2	ND	ND	ND	ND	2.7	2.9	0.24	0.21
Unit 50 (Site 4)	1	ND	ND	ND	ND	5.6	3.7	0.22	0.24
	2	ND	2.2	ND	3.4	4.8	11.7	0.21	0.29

*Pre-treatment, **Post-treatment.

incidental sediment ingestion rates of 1.0 g/day and 0.5 g/day for the adult and child, respectively, were arrived at through professional judgment, and informal consultations with California Department of Health Services (CDHS) risk assessors (Table 2). Hawley (1985) estimated a realistic adult soil ingestion rate of 0.48 g/day based on ingesting a layer of soil from one hand after yard work. We used a lower value because tules are rinsed in lake water before consumption, and are not expected to carry much sediment.

Opportunities for ingestion exposure occur during spring and summer when tules are edible. At Clear Lake, this period lasts 3 months (April 1st to June 30th). A conservative estimate of one large meal of tules per day throughout the 3-month harvest period was used to derive a 91-day exposure frequency.

Subchronic exposure calculations used an exposure duration of 1 year. Chronic exposure calculations used a lifetime exposure duration of 70 years for the adult, and 6 years for the child. Standard USEPA default values of 70 kg and 15 kg were used for adult and child body weights, respectively (USEPA 1997). A subchronic averaging time of 91 days was used to calculate a short, 1-time, 3-month exposure. Chronic averaging time was calculated by multiplying the 70-year (or 6-year), exposure duration by 365 days/year, resulting in an averaging time of 25,550 days for the adult, and 2,190 days for the child (Table 2).

Exposure Dose Calculation

Human exposure via the ingestion pathway was calculated according to the USEPA *Risk Assessment Guidance for Superfund* (USEPA 1989) by using the following formula:

$$\mathrm{ADD(mg/kg-day)} = [\mathrm{CFd} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}]/[\mathrm{BW} \times \mathrm{AT}]$$

where ADD is the average daily dose of fluridone residue from each ingested item (tules, sediment, or water); CFd is the fluridone concentration in each ingested

item (mg/kg); IR is daily ingestion rate (IR) (g/day); EF is the expected exposure frequency (days/year); ED is the years of exposure duration; BW is the receptor's body weight (BW) (kg); and AT is the averaging time (AT) (days) (Table 2).

The resulting residue intake estimate (ADD) for tules was summed with the residue intake estimates for incidentally ingested water and sediment, to arrive at a total daily intake (TDI) estimate for each receptor, in each scenario (Table 4).

EFFECTS ASSESSMENT (DOSE RESPONSE)

The amount of fluridone that would cause varying degrees of health effects was investigated in studies of mice, rats, rabbits, and dogs. Critical effects from exposure to fluridone observed in 2-year chronic rat feeding studies were glomerulonephritis, atrophic testes, eye keratitis; decreased body weight and organ weights (Elanco 1980). These effects were observed at exposure doses of 650 ppm (25 mg/kg/day). The chronic no observable effects level (NOEL) for the rat study (Elanco 1980) was 200 ppm (or 8 mg/kg/day). The USEPA IRIS database was used in this study to develop a chronic reference dose (RfD) of 0.08mg/kg-day for fluridone. Effects of fluridone exposure in a 1-year chronic dog feeding study showed weight loss, increased liver weight and alkaline phosphastase in test subjects (Elanco 1981). The NOEL for the 1-year dog study was 75 mg/kg-day. The systemic lower exposure limit (LEL) = 150 mg/kg-day. Another 3-month (subchronic) dog feeding study (Elanco 1978b) found no adverse health effects. Increased liver weight was observed in a 3-month (subchronic) rat feeding study (Elanco 1978a).

HAZARD ANALYSIS—METHODS

Hazard quotients (HQ) were calculated by comparing the ADD estimates derived in the Exposure Assessment, with a non-carcinogenic reference dose (RfD) established by the USEPA. An RfD of 2.0 mg/kg-day (Elanco 1978b) was used to derive subchronic HQs. An RfD of 0.08 mg/kg-day (Elanco 1980) was used to derive chronic HQs (USEPA Integrated Risk Information System—IRIS). An uncertainty factor of 100 (10 for human variability and 10 for extrapolation from animal study to human receptor) was incorporated into the NOEL of each reference study to derive the RfD. The lack of adverse effects found in the reproductive and developmental studies reviewed (CALEPA 2000) indicates that children, as a group, are not particularly sensitive to fluridone. For this reason, no additional safety factors were applied to the child receptor (NRC 1993).

HQs for the ingestion of tules, sediment, and water, for both subchronic and chronic scenarios, and adult and child receptors are presented in Table 2. Comprehensive HQs for each scenario and receptor were also evaluated using a total daily intake (TDI) exposure value in hazard calculations (Table 4).

RESULTS

Even after 10 years of herbicide treatment, environmental sampling showed very little fluridone in the lake environment (Table 1). At Sites 1 and 2, no fluridone was

Table 4. Total daily intake (TDI) doses for fluridone, and hazard quotients (HQ) associated with a "worst-case" exposure scenario.

Receptor	Tule IR[a] (g/day)	ADD[b]$_{Tule}$ (mg/kg-day)	Water IR[a] (ml/day)	ADD[b]$_{Water}$ (mg/kg-day)	Sed.[e] IR[a] (g/day)	ADD[b]$_{Sed}$ (mg/kg-day)	Total daily intake (TDI) (mg/kg-day)	RfD[c] (mg/kg-day)	HQ[d] (unitless)
SUB-CHRONIC									
Child ADD	355	8.0E-05	25	4.9E-07	0.50	2.2E-06	8.3E-05	2.0	4.2E-05
Adult ADD	710	3.4E-05	50	2.1E-07	1.00	9.3E-07	3.6E-05	2.0	1.8E-05
CHRONIC									
Child ADD	355	2.0E-05	25	1.2E-07	0.50	5.4E-07	2.1E-05	0.08	2.6E-04
Adult ADD	710	8.6E-06	50	5.3E-08	1.00	2.3E-07	8.9E-06	0.08	1.1E-04

[a]IR = Ingestion Rate.
[b]ADD = Average Daily Dose.
[c]RfD = Reference Dose.
[d]HQ = Hazard Quotient.
[e]Sed. = Sediment.

detected in any of seven edible-tule samples, and was detected in only one of eight whole-tule samples, at 2.9 ppb (Table 3). At Site 3, fluridone was undetectable in any of the four whole-tule or three edible-tule samples collected (Table 3). At Site 4 no fluridone was detected in any pre-treatment samples, with post-treatment detections of 2.2 ppb and 3.4 ppb in edible- and whole-tule samples from Spot 2 only (Table 3).

Duplicate pre-treatment sediment samples from Site 3, Spot 1 (Big Valley Rancheria at the swimming pier), found relatively high fluridone concentrations, 26.07 ppb and 35.21 ppb (wet weight), with 65 ppb (w.w.) detected in the one post-application sample. The large difference in duplicate sample values indicates the non-homogeneous nature of fluridone distribution in sediments and is likely an artifact of slow-release herbicide pellet distribution. One hundred meters down the shoreline at Site 3, Spot 2, pre- and post-application fluridone concentrations in sediment were significantly lower, less than 2.9 ppb (Table 3).

Fluridone concentrations in water samples from Sites 3 and 4 were consistently low, ranging from 0.184 ppb to 0.294 ppb. No fluridone was detected in either of the two QA/QC trip blanks prepared for this study. Fluridone concentrations in duplicate water samples from Site 3 varied by only 0.02 ppb. Differences between duplicate sediment samples from Site 3 ranged from 9% to 53%, due to one large outlying value.

Exposure Assessment Results

The average daily doses (ADD) of fluridone calculated in this "worst-case" exposure evaluation, for the ingestion of tule vegetation, and incidental amounts of sediment and water are presented in Table 2. The ADD for tule ingestion ranged from a high of 8.0×10^{-5} mg/kg-day in subchronic child calculations to a low of 8.6×10^{-6} mg/kg-day in chronic adult calculations (Table 2). Contributions from the incidental ingestion of lake water and sediment were even smaller, on the order of $10^{-7}$to 10^{-8} mg/kg-day for both child and adult receptors. Comprehensive total daily intake (TDI) calculations yielded subchronic and chronic child TDI exposures of 8.3×10^{-5} mg/kg-day, and 2.1×10^{-5} mg/kg-day, respectively. Adult exposures were slightly lower at 3.6×10^{-5} mg/kg-day, and 8.9×10^{-6} mg/kg-day, respectively (Table 4).

Hazard Quotients Results

Hazard quotients (HQ) for adult subchronic and chronic tule exposures were 1.7×10^{-5} and 1.1×10^{-4}, respectively (Table 2). HQs for the adult total daily intake (TDI) subchronic and chronic exposures were virtually the same: 1.8×10^{-5} and 1.1×10^{-4}, respectively (Table 4). HQs for the child TDI subchronic and chronic exposures were slightly greater, 4.2×10^{-5} and 2.6×10^{-4}, respectively (Table 4).

DISCUSSION

Environmental field sampling was designed to maximize the detection of fluridone in material that Native Americans are ingesting out of Clear Lake. The effective tule sample size became very small when fluridone was detected in only 1 of 14 edible-tule, and 2 of 16 whole- tule samples (Table 1). The problem with having such a small

number of positive detections is that in order to calculate a mean, non-detect samples values default to half the limit of detection (LOD), which grossly over estimates actual fluridone concentrations, skewing the mean. For this reason, no statistical figures are presented in Table 1.

The Code of Federal Regulations (CFR 40) pesticide residue tolerance for fluridone is 0.15 ppm for grass and legume forage, and irrigation water. The detection of fluridone in environmental samples from Clear Lake was well below this level. Fluridone was detected in tule vegetation at only two of eight sample spots: the active treatment area, Site 4—Spot 2 (2.2 ppb and 3.4 ppb), and at the north end of the lake, Site 2—Spot 1 (2.9 ppb) (Table 3). Environmental samples showed fluridone to be ubiquitously present throughout lake water at extremely low levels, less than 0.3 ppb. This could explain the presence of fluridone at low levels in areas where the herbicide had not been applied in preceding years, such as Site 2. Curiously, fluridone was not detected in vegetation at Site 3, which showed high sediment concentrations (maximum 65 ppb) compared to the rest of the lake (Table 3). Sediment samples from Site 4, where fluridone was applied during this investigation and in years preceding, showed only low levels of fluridone in both pre- and post-treatment samples (ranging from 2.27 ppb to 11.74 ppb). The unexpectedly high sediment concentrations at Site 3 (Spot 1), in both pre- and post-treatment samples, suggest a recent, undocumented application of fluridone by a private party to this area, as the CDFA had not applied fluridone to the site for 2 years preceding this study, and the county did not issued any citizen permits to apply fluridone in 2005.

The adult ingestion rate of 710 g/day (or approximately three cups) of tules was derived from consumption estimates by tribal members. Estimates varied widely from person to person and lacked quantitative values or consensus. The highest intake estimate was used in this hazard assessment to err on the side of conservatism, and protect public health. The use of realistic ingestion rates based on the consumption of homegrown fruits and vegetables (USEPA 1997) yielded smaller exposures and hazards than presented here.

Subchronic exposures were expectedly higher than the chronic exposures, as the same intake amounts were averaged over a 3-month period instead of a year. Tule exposures (ADD) for the child ranged from 8.0×10^{-5}mg/kg-day (subchronic) to 2.0×10^{-5}mg/kg-day (chronic). Similarly, adult tule exposures ranged from 3.4×10^{-5}mg/kg-day (subchronic) to 8.6×10^{-6}mg/kg-day (chronic) (Table 4).

Calculations of additional fluridone exposure caused by the incidental ingestion of lake water and sediment were negligible even when maximum detected environmental concentrations were used in "worst-case" exposure calculations. Water fluridone concentrations were relatively constant throughout the lake, averaging 0.22 ppb. Use of the highest detected water concentration, 0.29 ppb in incidental ingestion calculations did not significantly affect total daily intake (TDI) estimates, adding only 2.1×10^{-7} mg/kg-day to adult subchronic exposures (Table 4). Similarly, even the use of an extremely high, outlying value of 65 ppb for sediment fluridone concentrations (approximately 5 times the 14.0 ppb average of all sediment samples together), added only 9.3×10^{-7} mg/kg-day to adult subchronic exposures (Table 4). The exposure and hazard values calculated from using this extremely conservative sediment value, erred toward over estimating exposure and health hazards.

The USEPA *Health and Environmental Effects Profile for Fluridone* identified an acceptable daily intake (ADI) of 0.09 mg/kg-bw/day (10^{-1}) for fluridone via oral exposure (USEPA 1984). This is calculated to be the amount of fluridone to which humans can be exposed on a daily basis over a lifetime without suffering deleterious effects. TDI exposures in the CDFA study were five orders of magnitude lower (10^{-6}mg/kg-day) than the acceptable daily intake parameters outlined by the USEPA (Table 4).

Although exposures (ADD) for subchronic scenarios were higher than chronic scenarios, the hazards (HQ) were lower. This occurred because the reference dose (RfD) used in subchronic exposure calculations (2.0 mg/kg-day) was 25 times greater than the RfD used in chronic exposure calculations (0.08 mg/kg-day). HQs calculated for fluridone, in all scenarios evaluated, were small enough to be negligible. Subchronic hazards were on the order of 10^{-5}, with chronic hazards on the order of 10^{-4} (Table 4).

CONCLUSION

Native Americans living around Clear Lake, California, are exposed to fluridone not only through traditional handcrafts such as boat and basket making, but via the seasonal consumption of tules. Interestingly, the higher exposures found for the shorter (subchronic) time-frame did not translate to higher health hazards. The highest health hazards came from chronic scenarios where exposures doses were lower than in subchronic scenarios. In summary, neither the hazards calculated in subchronic nor chronic exposures indicated levels of health concern. Under current fluridone application regiments, effectively no exposure hazard was found for eating tules from Clear Lake.

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