International Journal of Soil, Sediment and Water

Volume 3 | Issue 2

Article 2

July 2010

ASSESSMENT OF STREAM FISH MORTALITY FROM SHORT-TERM EXPOSURE TO ILLITE CLAYS USED AS AN IN SITU METHOD FOR REMEDIATING 137CS CONTAMINATED WETLANDS

Bon-Jun Koo *California Baptist University,* bonjunkoo@calbaptist.edu

Dean E. Fletcher Savannah River Ecology Laboratory, dfletcher@srel.edu

Thomas G. Hinton thinton@uga.edu

Christopher D. Barton barton@uky.edu

Mark R. Matsumoto matsumot@engr.ucr.edu

Recommended Citation

Koo, Bon-Jun; Fletcher, Dean E.; Hinton, Thomas G.; Barton, Christopher D.; Matsumoto, Mark R.; Teune, Amanda M.; and Kaplan, Daniel I. (2010) "ASSESSMENT OF STREAM FISH MORTALITY FROM SHORT-TERM EXPOSURE TO ILLITE CLAYS USED AS AN IN SITU METHOD FOR REMEDIATING 137CS CONTAMINATED WETLANDS," *International Journal of Soil, Sediment and Water*: Vol. 3 : Iss. 2 , Article 2.

Available at: https://scholarworks.umass.edu/intljssw/vol3/iss2/2

This Article is brought to you for free and open access by ScholarWorks@UMass Amherst. It has been accepted for inclusion in International Journal of Soil, Sediment and Water by an authorized editor of ScholarWorks@UMass Amherst. For more information, please contact scholarworks@library.umass.edu.

See next page for additional authors

Follow this and additional works at: https://scholarworks.umass.edu/intljssw

ASSESSMENT OF STREAM FISH MORTALITY FROM SHORT-TERM EXPOSURE TO ILLITE CLAYS USED AS AN IN SITU METHOD FOR REMEDIATING 137CS CONTAMINATED WETLANDS

Cover Page Footnote

We are most appreciative of the technical assistance provided by Jamie Marsh (SREL), Yi Yi (SREL) and Michelle Standora (SREL). This research was supported by WSRS-SGCP and by the Environmental Remediation Sciences Division of the Office of Biological and Environmental Research, the U.S. Department of Energy, and through Financial Assistance Award No. DE-FC09-96-SR18546 to the University of Georgia Research Foundation. Westinghouse Savannah River Company is operated for the U.S. Department of Energy under contact DE-AC09-96SR18500.

Authors

Bon-Jun Koo, Dean E. Fletcher, Thomas G. Hinton, Christopher D. Barton, Mark R. Matsumoto, Amanda M. Teune, and Daniel I. Kaplan

ASSESSMENT OF STREAM FISH MORTALITY FROM SHORT-TERM EXPOSURE TO ILLITE CLAYS USED AS AN *IN SITU* METHOD FOR REMEDIATING ¹³⁷CS CONTAMINATED WETLANDS

Koo, Bon-Jun^{1§}, Fletcher, D.E.², Hinton, T.G.³, Barton, C.D.⁴, Matsumoto, M.R.⁵, Teune, A.M.⁶, D.I. Kaplan⁷

¹Department of Natural and Mathematical Sciences, California Baptist University, Riverside, CA, USA, 92504

² Savannah River Ecology Laboratory, University of Georgia, Aiken, SC, USA, 29802

³ Savannah River Ecology Laboratory, University of Georgia, Aiken, SC, USA, 29802

⁴Department of Forestry, University of Kentucky, Lexington, KY, USA, 40545

⁵Department of Chemical and Environmental Engineering, University of California, Riverside, CA,USA, 92521

⁶ Department of Natural and Mathematical Sciences, California Baptist University, Riverside, CA, USA, 92504

⁷Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, SC, USA, 29808

ABSTRACT

Due to their physical properties, illite clays can sorb cesium-137 almost irreversibly, and therefore sequester contamination from the environment. However, applying large amounts of clay to natural aquatic habitats for *in situ* remediation purposes may create conditions of high turbidity and sedimentation. To evaluate potential effects of turbidity from illite application on survivorship of stream fish, yellowfin shiners (Notropis *lutipinnis*) and tessellated darters (*Etheostoma olmstedi*) were subjected to treatment with two different types of clay in flow-through simulated stream raceways. Turbidity and fish mortality were subsequently monitored for seven days. At 2-m downstream from the application point, mean turbidity peaked during clay application at 525 and 72 nephelometric turbidity units (NTU) in the air-floated illite and semi-dry illite treatments, respectively. Turbidity returned to levels similar to that of the controls (4-6 NTU) after four hours in the air-floated illite raceways and one hour in the semi-dry illite raceways. Although the majority of the suspended clay was quickly flushed from the system and the remaining settled to the bottom, turbidity did continue to fluctuate because of fish movements and sediment resuspension. Fish mortality did not significantly differ among control and illite treated raceways.

Keywords: Cesium-137; Illite clay; Wetlands; Environmental impact; Turbidity, Fish

[§] Corresponding Author: Bon-Jun Koo, 8432 Magnolia Ave., Department of Natural and Mathematical Sciences, California Baptist University, Riverside, CA, USA, 92504. Tel.: 951-343-4621; Email:bonjunkoo@calbaptist.edu

1. INTRODUCTION

Cesium-137, a fission product, is a radionuclide of primary concern in radioactive waste It is a significant soil and sediment contaminant at various U.S. management. Department of Energy (DOE) locations, such as the Hanford Site in Washington and Oak Ridge in Tennessee (Brisbin et al., 1974; Sharitz et al., 1975; Liu et al., 2003). Over 1,200 hectares of wetlands on the DOE's Savannah River Site (SRS; Aiken, SC, USA), encompassing both lentic and lotic habitats, are contaminated with a variety of radionuclides including ¹³⁷Cs, ⁶⁰Sr, and isotopes of Pu. Cesium-137 is often the radionuclide of primary concern in these systems because of its biological mobility, moderately long half-life (30.2 years), and detrimental effects. One estimate of the 137 Cs contamination level within the aquatic systems of the SRS is 21 TBq (564 Ci; Carlton et al., 1992). The problem of contaminated aquatic systems and their associated risks are heightened because ¹³⁷Cs is particularly bioavailable. Plants are able to uptake cesium from the soil into tissues at a concentration ratio of 14.9 ± 2.3 Bq kg⁻¹ dry plant / Bq kg⁻¹ dry soil (Hinton et al., 1999). This value is well above the default ration level of 0.1 recommended for risk calculations by the National Council on Radiological Protection and Measurement (Hinton et al., 2002).

Two main reasons for the enhanced bioavailability of ¹³⁷Cs in these aquatic systems are the low percentage of clays in SRS soils and the fact that the existing clays are dominated by kaolinite and iron oxides. Kaolinite, which has a 1:1 layer structure in which tetrahedral (T) and octahedral (O) sheets are arranged in the sequence T-O, forms weak bonds with ¹³⁷Cs. In contrast, 2:1 phyllosilicate minerals have a 2:1 layer in the sequence T-O-T, such as illites and micas, and form very strong bonds with ¹³⁷Cs. Illite minerals can sorb ¹³⁷Cs almost irreversibly so that once ¹³⁷Cs sorbs to the mineral it is contained and will not readily desorb. Interestingly, SRS sediments have some mica-like minerals (Ruhe and Matney, 1980), but they are largely ineffective in forming strong bonds with ¹³⁷Cs (Seaman et al., 2001). This is presumably the result of lower particle unit charge, due to extensive weathering.

Environmentally sensitive wetlands pose a significant remediation challenge. Current cleanup technologies for aquatic systems are generally destructive to the ecosystem, and may increase exposure of contaminants to workers (Whicker et al., 2004). Typical cleanup protocols entail draining the wetland and either immobilizing the sediments, where 95% of the radionuclides reside (Whicker et al., 1990), or hauling the sediments off to be buried. New technologies are needed to avoid this type of environmentally destructive remediation. Hinton et al. (2001, 2002, and 2006) previously showed that naturally occurring illite minerals with a high complexing capability for ¹³⁷Cs can sequester ¹³⁷Cs and reduce its bioavailability when applied to ¹³⁷Cs-contaminated wetlands on the SRS. Over the course of a 42-week treatment period, illite minerals applied *in situ* reduced ¹³⁷Cs concentrations in water 25- to 30-fold, in aquatic plants 3- to 5-fold, and in fish 2- to 3-fold.

However, the application of large amounts of clay to aquatic ecosystems could create conditions of high turbidity and sedimentation that can have environmental impacts of their own. High levels of turbidity are a common by-product of activities related to urban development, inadequate land management practices, deforestation, mining, road construction, and intensive agriculture (Cohen et al., 1993; Henley et al., 2000; Iwata et al., 2003). Each source of suspended sediment may have different effects on nearby aquatic habitats based upon the length of exposure time, soil particle size, the concentration of suspended sediment, the organisms that inhabit the area, and other contaminants that the sediment might contain (Newcombe and MacDonald, 1991; Waters, 1995; Jowett and Boustead, 2001). Thus it is important to examine specifically the impacts of illite remediation of ¹³⁷Cs contaminated wetlands.

Illite application as an *in situ* technique for remediation of ¹³⁷Cs-contaminated streams would be a temporary source of sediment pollution, with many potential shortand long-term environmental impacts from increased turbidity and sedimentation. Initial steps in assessing of this technique in streams included evaluation of settling rates of different types of illite clay, and observing fish mortality after clay application to flowing water in artificial streams. Fish mortality was found to be no greater in treated waters than in controls.

2. MATERIALS AND METHODS

2.1 Characteristics of illites

Numerous minerals were tested in the laboratory and Todd Light Illites were found to enhance adsorption and sequestration of ¹³⁷Cs, even in the presence of competing ions such as ammonium (Hinton et al., 1999). Todd Light illites are available from a commercial supplier (Kentucky-Tennessee Clay Company in Nashville, TN) in five different particle sizes with different moisture contents. Particle size and moisture content influence mineral settling rates and thus could be particularly important to a stream application. Therefore, three Todd Light illites of different particle size and moisture content were purchased for this study (air-floated, semi-dry and crude with moisture contents of 1-3%, 8-12%, and 20-22%, respectively).

In an effort to select the most appropriate type of illite for remediation, the settling rates and flocculation behavior of the three Todd Light Illites within beakers of static water (3 g of each clay; 1.5 L water; 24 hrs settling duration) were compared. After 19 hours, the water was vigorously stirred with a glass rod for 10 seconds and the resuspension and flocculation in the water were qualitatively noted.

The Todd Light Illites were also examined for settling characteristics using a batch beaker test. This study was performed in the laboratory by adding the illites to tap water in beakers. This experiment was conducted in a similar manner (20 g illites in 3.5 L water) to experiments described above except that the water was continuously mixed for 70 h by a Stir-pak[®] Laboratory Mixer (overhead stirrer, Cole-Parmer Instrument Company, Vernon Hills, IL) at a speed of 875 rpm. Suspension and flocculation of illites in the water were observed. The experiment was replicated three times.

Physicochemical properties of the different types of Todd Light illites were determined following methods of the Natural Resources Conservation Service (NRCS, 1996). Analyses were performed on air-dried colloid samples that had been crushed and passed through a 0.23-mm sieve. All analyses were conducted in triplicate. Cation exchange capacities (CEC) were determined by summation of exchangeable cations (Ca, Mg, Na, K, and Al) extracted with three overnight extractions of 0.05 M BaCl₂. Elemental compositions of the BaCl₂ and dithionite-reduced extracts were determined by inductively coupled plasma – optical emission spectroscopy (ICP-OES). Organic carbon (OC) was determined using a Leco Carbon Analyzer, Model CR-12 (Leco Corp., St. Joseph, MI). Total sand, silt, and clay concentrations were determined by the micropipette method (Miller and Miller, 1987) in which soil particles were dispersed in a 0.5% (w/v) sodium metaphosphate and 0.1 M NaOH solution. Clay pH was measured in a 1:1 clay-water suspension.

2.2 Study organisms

To evaluate lethal effects of illite application on stream fish, two locally common species found in the middle Savannah River Basin were used. Yellowfin shiners (*Notropis lutipinnis*) are usually less than 65-mm standard length (SL). These shiners are often abundant in small to intermediate size tributary streams where they inhabit the middle of the water column. Yellowfin shiners feed on terrestrial insects and a variety of aquatic invertebrates and are an important invertivorous predator (Sheldon and Meffe, 1993).

The tessellated darter (*Etheostoma olmstedi*) also rarely exceeds 70-mm SL. Tessellated darters are benthic feeders preying on a variety of aquatic insects, especially chironomid larvae (Sheldon and Meffe, 1993). Darters (Percidae, Etheostomatinae) are generally sensitive to environmental disturbance due to their benthic habits (Page, 1983) and the number of native insectivorous cyprinid species present at a site is often incorporated into stream environmental assessments (Karr et al., 1986; Paller et al., 1996).

2.3 Fish collection

Using backpack electrofishers and a 3-mm mesh seine, fish were collected on 22-23 July, 14-15 August, and 3-5 September 2003. Approximately 80 tessellated darters and 200 yellowfin shiners were obtained during each collection period from Fourmile Branch, a tributary of the Savannah River located entirely within the SRS (Figure 1). Fourmile Branch, originating near the center of the SRS, runs approximately 24 km to the Savannah River (WSRC, 1998). It is characterized as a blackwater, sandy bottom stream with a channel that alternates between swift runs and pools. Due to effects from thermal effluents released from the DOE nuclear production reactors prior to 1986, much of Fourmile Branch has early successional riparian vegetation.

2.4 Clay application and fish exposure

Three indoor continuous flow-through raceways (U shape; 35 cm width, 33 cm depth, with 10 m and 8 m lengths for outside and inside banks, respectively) were used for this study. Stream water was pumped from Upper Three Runs, a relatively undisturbed tributary of the Savannah River, into a reservoir tank and subsequently released into the raceways. A filter system was employed to remove large debris. The filter system was a two-stage process with a 5-mm screen (20 cm x 20 cm) attached to the outlet pipe of the reservoir tank and a 3-mm mesh plastic screen installed in the raceways. Turbidity prior to clay application was examined for background levels in stream water. Turbidity samples were collected in triplicate at 2, 3, 4, 5, 6, and 7 meters of the raceways. Turbidity values decreased exponentially with time and stabilized between 7-8 nephelometric turbidity units (NTU) twelve hours after the system was turned on. Based on this pretreatment characterization of the raceways, the system was started 12 hours prior to introducing fish. Water depths were maintained at 13-14 cm at the beginning of the raceways and 14-15 cm at the lower end, with an average velocity of 9 cm sec⁻¹ in each raceway. Water temperatures did not vary among raceways, but water temperatures did vary slightly among replicates due to seasonal temperature changes: 22-23 ⁰C in July and August, and 20-21 ^oC in September.

Upon collection, fish were held in a 660-L continuous flow-through holding tank, which used the same water as raceways until stocked into the experimental raceways. During the first replicate, fish were weighed in a tared beaker of water before being introduced into the experimental raceways. This added holding time and handling proved too stressful without a much longer acclimation period, particularly to the yellowfin shiners that suffered high mortality. Therefore, the weighing procedure was discontinued for the remaining replicates and tessellated darters and yellowfin shiners were held for approximately 24 hours and one to several hours, respectively, prior to stocking in replicates two and three.

Following the holding period, fish were randomly placed into one of the three designated treatment raceways: untreated control, treated with air-floated illite, or treated with semi-dry illite. For each of the three replicates of the experiment, treatment condition was rotated among raceways. Each raceway was stocked with 50 yellowfin shiners and 20 tessellated darters. Stocked fish were allowed to stabilize in the raceway systems for three days prior to beginning the experiment. Any fish that died in the raceways during this stabilization period was replaced with a fish from the holding tank. The fish mortality was less than 5 percent and replacement was done at once prior to the treatment addition. After the three-day acclimation period, 9.8 kg of moisturized airfloated and semi-dry illites were applied at the head of the raceways. In this manner, illites were flowed readily into the system and dispersed evenly. This amount of illite was determined to be equal to that which can cover the sediment with 0.25 cm. Hinton et al. (2001, 2002, and 2006) indicated that this quantity was previously used to successfully reduce ¹³⁷Cs concentrations in water and fish in field trials using lentic limnocorrals.



Figure 1. Map of the study area and three fish collection locations on Fourmile Branch of the Savannah River within the Savannah River Site in South Carolina.

Each replicate of the experiment was maintained for seven days. Thus, nine trials were conducted (2 types of illites and 1 control x 3 replicates). Turbidity was measured at hours 1, 2, 4, 8 and 24 on the day clay was applied, and then on the 2^{nd} , 4^{th} and 7^{th} day after application. Turbidity samples were collected in triplicate at 2, 3, 4, 5, 6, and 7 meters of the raceways. Turbidity values in NTU were measured using a LaMotte 2020 Turbidimeter (LaMotte Co., Chestertown, MD). Fish survival was assessed on the 7^{th} day.

To minimize disturbance of fish in the experimental raceways and consequent artificial resuspension of the clay, dark curtains were hung around each raceway. Habituation to feeding area was avoided by reaching through the curtain and pouring the food into the first half of the raceway, at locations that varied each day. Raceway fish were fed an average of 4.4 g per day of a 50% mixture of commercially available frozen brine shrimp and bloodworms (Chironomid larvae) (San Francisco Bay Brand, Newark, CA), that was approximately three percent of the combined body weight of all fish in the raceway.

The experimental units in this research were the replicated raceways in the artificial stream experiments. Thus, sample sizes for statistical analyses were the number of raceways, not the total number of fish and turbidity samples taken. Differences between treatments (control, air-floated, and semi-dry clay applications) were compared using Analysis of Variance (ANOVA) and General Linear Models (GLMs) in SAS (SAS, 1999).

3. **RESULTS AND DISCUSSION**

3.1 Physicochemical characteristics and settling behavior of illites

Analyses of the three Todd Light illites indicated that surface area, OC, pH, texture, and Fe concentration were similar (p > 0.05). However, concentrations of Al, Ca, K, and Mg occurring in the illites decreased as clay particle size increased, as did CEC (Table 1). Na concentration was lower in the largest particle clay, but did not significantly differ between the other two types of illite.

Turbidity was measured after the batch beaker laboratory settling experiment and yielded readings of 360 ± 2.65 , 208 ± 7.09 , and 140 ± 1.53 NTU (mean \pm Stdev.) for airfloated, semi-dry, and crude illite treatments, respectively. As anticipated, the turbidity measurements were significantly different and the crude illite produced the lowest turbidity because it had the largest aggregation size (average size > 2 cm in diameter) and highest moisture content. Semi-dry and crude illites immediately flocculated and gradually re-suspended into the water column from the sediment when stirred (as the large particles were broken down). The lower CEC of the semi-dry and crude illites may have induced instability by weakening the repulsive forces between particles (Goldberg et al., 1990), allowing colloid flocculation to occur. With both illite types, after suspension, the water throughout the experimental period, about 10 days.

3.2 Clay associated turbidity

Background turbidity values measured before clay application were low, with averages between 3.9-5.9 NTU for all three raceways across all three replicates. Air-floated illite produced the highest average turbidity values, measuring between 232-525 NTU at application. Because the air-floated illite was very fine, some remained suspended in a film on the water surface and did not mix into the water column. Semi-dry illite did not cloud the water to the same degree as the air-floated because of the higher moisture content and larger particle size. The semi-dry illite only increased the turbidity of the water to 50-81 NTU. At the application site near the head of the raceway, a small submerged pile of the semi-dry clay could be seen throughout the seven day experiment. This pile of clay was slowly eroded down and did cause some variation between turbidity measures in the semi-dry treatment. This was not observed in the air-floated treatment.

| Property ^a | Type of Illite | | | |
|---|--------------------|------------------|------------------|--|
| | Air-floated | Semi-dry | Crude | |
| pH | $4.2 \pm 0.2a^{b}$ | $4.7 \pm 0.2a$ | $4.4 \pm 0.1a$ | |
| OC (%) | $1.09 \pm 0.01a$ | $1.07 \pm 0.02a$ | $1.06 \pm 0.02a$ | |
| CEC (meq 100g ⁻¹) | $7.68 \pm 0.03a$ | $6.16 \pm 0.07b$ | $4.92 \pm 0.07c$ | |
| EC (μ S cm ⁻¹) | $155.0 \pm 2.1a$ | 111.0 ± 1.3a | $119.1 \pm 2.4a$ | |
| Surface Area $(m^2 g^{-1})$ | $31.3 \pm 0.5a$ | $28.6 \pm 0.6a$ | $32.2 \pm 1.2a$ | |
| Clay (%) | $55.6 \pm 0.1a$ | 57.4 ± 4.3a | $56.3 \pm 2.0a$ | |
| Sand (%) | $34.8 \pm 0.2a$ | $30.7 \pm 4.6a$ | $35.2 \pm 2.9a$ | |
| Silt (%) | $9.6 \pm 0.1a$ | $11.9 \pm 0.3a$ | $8.5 \pm 0.9a$ | |
| $Ca (mg kg^{-1})$ | 599 ± 6.1a | 489 ± 6.4b | 389 ± 6.2c | |
| $K (mg kg^{-1})$ | $36.7 \pm 0.5a$ | $18.8 \pm 0.1b$ | $11.0 \pm 0.5c$ | |
| $Mg (mg kg^{-1})$ | $182 \pm 5.8a$ | $146 \pm 5.4b$ | $128 \pm 1.4c$ | |
| Na $(mg kg^{-1})$ | $23.1 \pm 2.5a$ | $13.4 \pm 0.3a$ | $9.7 \pm 0.3b$ | |
| Al (mg kg ⁻¹) | $40.8 \pm 0.3a$ | $32.9 \pm 3.0b$ | $29.2 \pm 1.1c$ | |
| $\operatorname{Fe}\left(\operatorname{mg}\operatorname{kg}^{-1}\right)$ | $18.9 \pm 0.3a$ | $17.5 \pm 0.4a$ | $25.8 \pm 0.9a$ | |

Table 1. Selected physicochemical properties of illites used in the study (mean \pm Stdev; n = 3)

 ^{a}OC = organic carbon, CEC = cation exchange capacity, EC = electrical conductivity ^bWithin a property, means among the three types of illites followed by the same letter are not significantly different (p > 0.05).

Because of the continual flow of new water into the raceways, the initial turbidity cleared within several hours as suspended clay was flushed out of the system. The same phenomenon would likely occur in a natural stream with a single application of illite. Turbidity decreased to levels close to those of the controls (Figure 2) within four hours after the application of air-floated treated raceways and one hour after for semi-dry treated raceways. However, there was substantial resuspension of clay in both treatment conditions throughout the course of the experimental period, due to fish movement across the bottom of the raceways. As fish moved they disturbed the settled sediment, and thus temporarily elevated turbidity measurements at certain distances along the raceways and produced larger standard deviations within replicates.

3.3 Fish mortality

There were 150 yellowfin shiners and 60 tessellated darters used for each replicate, 450 and 180 fish total, respectively, over the course of the study. The control suffered the most mortality, with the loss of seven yellowfin shiners. In the subsequent two replicate experiments; mortality in controls was reduced to one and two individuals, respectively. The higher standard deviation for the control treatment (Table 2) was due to the high mortality in the first replicate.

No more than three yellowfin shiners were lost from any clay treated replicate. Total mortality of yellowfin shiners over the duration of all replicates of the experiment was 10 in the control, 6 in semi-dry, and 6 in air-floated. Average mortality across replicates was $4.0 \pm 2\%$ for air-floated and $4.0 \pm 2\%$ for semi-dry illite treated raceways. Yellowfin shiner mortality in control raceways was $6.7 \pm 3\%$. These variations in yellowfin shiner

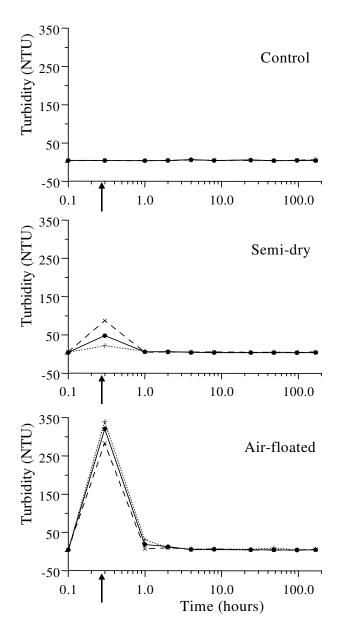


Figure 2. Turbidity measurements taken at three meters in control, semi-dry and air-floated illite treatments for replicate 1 (\bullet), replicate 2 (x), and replicate 3 (+). Point of illite application is indicated by an arrow. Before treatment and during treatment turbidity measures are represented by 0.1 and 0.3 hours, respectively.

Table 2. Mortality of yellowfin shiners (*Notropis lutipinnis*) and tessellated darters (*Etheostoma olmstedi*) in different clay treatments. Each raceway was stocked with 50 yellowfin shiners and 20 tessellated darters at the beginning of the seven-day replicates. Actual numbers of individuals dead are presented, with percentages in parentheses

| Fish/Treatment | Replicate 1 | Replicate 2 | Replicate 3 | Average | SD |
|--|-------------|-------------|-------------|-----------|-----|
| Yellowfin shiner (Notropis lutipinnis) | | | | | |
| Control | 7 (14) | 2 (4) | 1 (2) | 3.3 (6.4) | 3.2 |
| Semi-dry | 1 (2) | 2 (4) | 3 (6) | 2 (4.0) | 1.0 |
| Air-floated | 2 (4) | 1 (2) | 3 (6) | 2 (4.0) | 1.0 |
| Tessellated darter (<i>Etheostoma olmstedi</i>) | • | - | - · · · · | - | |
| Control | 0 (0) | 0 (0) | 0 (0) | 0 (0) | 0.0 |
| Semi-dry | 0 (0) | 0 (0) | 0 (0) | 0 (0) | 0.0 |
| Air-floated | 0 (0) | 0 (0) | 0 (0) | 0 (0) | 0.0 |

mortality among the control and illite treated raceways were not statistically significant (df = 2,6; $R^2 = 0.13$; p > 0.60) (Table 2).

There was no mortality of tessellated darters in any of the three treatments or in any of the replicates.

3.4 Discussion

Analyses of particle settling and flocculation rates among three commercially available illites that differed in particle size and moisture content revealed flexibility of the clay application method and versatility of the illites to different remediation needs. Air-floated Todd Light Illites were found to disperse quickly throughout the water column and would be ideal for remediating lakes, or possibly long reaches of a contaminated stream. If, however, small localized contaminated hot spots within streams are the target of remediation, then more rapid settling characteristics of the crude Todd Light Illite would allow one to place the mineral specifically where needed, rather than broadly dispersing it within the stream.

As noted, the objective of this study was to assess the impact of turbid water resulting from clay application on fish mortality. Based on the study results, turbidity from the application of clay to the raceways at the optimum concentration previously determined in a ¹³⁷Cs-contaminated wetland (Hinton et al., 1999) had no lethal effect on the species tested. It should be noted however, that the optimum clay concentration required to reduce ¹³⁷Cs bioavailability was determined in lentic habitats. Field testing will be required to corroborate its effectiveness in a stream. When illite was applied to the artificial flow-through streams, water clarity returned within a matter of hours. Both concentration and exposure time of suspended sediments are important determinants of

their effects on aquatic systems (Newcombe and MacDonald, 1991). Such a rapid reduction in the turbidity will diminish initial impacts to organisms that would be affected by the high concentration of suspended sediments. These results are in agreement with those of others who have used clean, uncontaminated sediment to test for effects of short-term turbidity and found no mortality (Lake and Hinch, 1999; Au et al., 2004).

Variables that may contribute to the background level of fish mortality include stress of handling, life stage, physiological condition, water temperature, and disease (Henley et al., 2000). The yellowfin shiner mortality rate in the control treatment was inflated slightly by higher mortality in the first replicate when fish in this treatment became stressed due to the added handling stress involved with the weighing process employed in the first replicate. In the subsequent replicates there were no noticeable mortality problems in any treatment raceway. Impacts of the clay application on aquatic biota and habitat quality will be affected by stream characteristics such as water velocity. Turbidity will be flushed through the system at a reduced rate in slower moving waters. This would be most extreme in lentic waters that do not have the advantage of flowing water to carry and disperse the clay. Hinton et al. (2004) predicted up to 115 days would be required for water to clear in a pond habitat treated with illite clays.

Also, substrates and water quality of habitats that have already been disturbed by contaminants may be less impacted by illite treatment than more pristine waters. Additionally, some fish that permanently inhabit waters with high turbidity can become adapted and may not experience as many negative sublethal effects with additional turbidity (Bunt et al., 2004).

Although sublethal effects were not assessed, it is worthy to note that after clay application both species of fish tended to congregate in areas of clearer water and less sediment deposition—darters were rarely observed resting on the clay sediments. In fact, because of observations during an experimental trial clay application, fish excluding screens had to be placed immediately downstream of the clay introduction location to prevent fish from escaping effects of the clay. Similar observations of behavioral avoidance to suspended sediment have been reported (Bisson and Bilby, 1982; Gradall and Swenson, 1982; Boubee et al., 1997). In a remediation of a real stream, many fish will likely avoid the clay treatment turbidity and sedimentation by migrating up or downstream away from the turbidity or into any available side channels. This migration could have consequences at the individual, population and the community levels.

Fish may be influenced by direct exposure to suspended solids or indirectly by impacts to habitat or prey organisms. Increased suspended sediments have been shown to cause behavioral, sublethal, and lethal effects in many species of fish. Three categories of effects were defined by Newcombe and MacDonald (1991): (1) Lethal effects that increase mortality, reduce populations, or damage the ecosystem's ability to produce fish; (2) Sublethal effects that damage tissues or physiology of an organism enough to disrupt function, but not cause death; and (3) Behavioral effects that alter normal activity patterns

and behavior from that typical of an undisturbed environment. While not independent of each other, these categories are useful in discussing effects of turbidity on fishes.

The level of turbidity detrimental to a fish is species specific, but behavioral changes include adjustments in feeding and predator/prey interactions and have been documented at turbidities as low as 5-40 NTU (Barrett et al., 1992; De Robertis et al., 2003; Sweka and Hartman, 2003). Activity levels have increased at sediment concentrations of 10-15 mg L⁻¹ (1 NTU equals to 0.13 Silica Unit, in mg SiO₂ L⁻¹) likely produced by an alarm reaction or attempts to avoid the disturbance (Chiasson, 1993a,b). Social hierarchies and territorial behavior have been disrupted at a turbidity over 20 NTU (Berg and Northcote, 1985). Physiological effects that may represent sublethal costs include effects on respiration and gill function (Servizi and Martens, 1992; Shaw and Richardson, 2001) and increased stress response, e.g. depressed leukocrit (Lake and Hinch, 1999).

Effects of suspended sediments on fishes have been found to differ appreciably due to species, fish size, life stage, and physiological condition, or of habitat characteristics such as water temperature, dissolved oxygen concentration, particle size, and chemical composition of the sediments and presence of other contaminants (Newcombe and MacDonald, 1991; Waters, 1995). The effects of exposure are governed by both concentration and exposure time (Newcombe and MacDonald, 1991). Direct lethality from exposure to suspended sediment is often caused by the gills becoming coated or damaged (Au et al., 2004). This is often indicated by increased coughing rates (Servizi and Martens, 1992).

 LC_{50} 's reported in the literature have a wide range. In some cases, species that share similar tolerance levels for suspended sediments have been noted to have the same habitat preference and maintain similar positions in the water column (Wilber and Clark, 2001). *Anadromous salmonids* are the most frequently studied (Waters, 1995) and for juveniles in 96 hr exposures LC_{50} 's ranged from 22.7 – 164.5 g L⁻¹ (Servizi and Gordon, 1990; Servizi and Martens, 1991; Lake and Hinch, 1999). In adult salmonids across multiple studies, mortality was only found in unnaturally high turbidities over 100 g L⁻¹ as summarized in Wilber and Clark (2001). In contrast, other results suggest thresholds as low as 1.4 g L⁻¹ over six weeks for green grouper (*Epinephelus coidoides*) (Au et al., 2004).

4. CONCLUSIONS

Tradeoffs will always exist between minimizing the threat presented by ¹³⁷Cs contamination and the environmental disturbance and risks resulting from remediation efforts. Illite treatment provides an alternative remediation strategy that may cause less damage to sensitive aquatic habitats than other ¹³⁷Cs remediation strategies in which the wetland is completely drained and the contaminated sediments are dredged up and relocated. Knowing the environmental risks of remediation is essential for striking the best balance between reducing ¹³⁷Cs risks and minimizing wetland disturbance.

This study represents a first step in assessing the effects of an illite application process by documenting the rapid clearing of the water and adult fish mortality comparable to controls. However, younger life stages are generally more susceptible to suspended sediments than adults (Servizi and Martens, 1991). Susceptibility of eggs and larvae has been well established (Isono et al., 1998; Wilber and Clark, 2001). This study addresses the acute short-term exposure of adults to turbidity, but the longer-term effects of sediments that will develop when large amounts of clay are introduced into a system remain to be examined. Such sediments may affect habitat quality and many aspects of aquatic communities including fish (Newcombe and MacDonald, 1991; Jowett and Boustead, 2001), invertebrates (Lawrence and Ward, 1982; Alin et al., 1999), and primary producers (Grobbelaar, 1985; Holz et al., 1997). While these results are encouraging and suggest that the illite *in situ* remediation method is far less destructive than draining the wetland and dredging up sediments, future field studies of the fate of the illite clay in a stream following treatment and the corresponding response of the entire community of aquatic organisms will provide much needed guidance in the implementation of this promising 137 Cs remediation technique.

5. REFERENCES

- Alin, S., Cohen, A.S., Bills, R., Gashagaza, M.M., Michel, E., Tiercelin, J.J., Martens, K., Coveliers, P., Mboko, S., West, K., Soreghan, M., Kimbadi, S., and Ntakimazi, G. 1999. Effects of landscape disturbance on animal communities in Lake Tanganyika, East Africa. Conserv. Biol.13, 1017-1033.
- Au, D.W.T., Pollino, C.A., Wu, R.S.S., Shin, P.K.S., Lau, S.T.F., and Tang, J.Y.M. 2004. Chronic effects of suspended solids on gill structure, osmoregulation, growth, and triiodothyronine in juvenile green grouper *Epinephelus coioides*. Mar. Ecol. Prog. Ser. 266, 255-264.
- Barrett, J.C., Grossman, G.D., and Rosenfeld, J. 1992. Turbidity-induced changes in reactive distance of rainbow trout. Trans. Amer. Fish Soc. 121, 437-443.
- Berg, L., and Northcote, T.G. 1985. Changes in territorial, gill-flaring, and feeding behavior in juvenile coho salmon (*Oncorhynchus kisutch*) following short-term pulses of suspended sediment. Can. J. Fish Aquat. Sci. 42, 1410-1417.
- Bisson, P.A., and Bilby, R.E. 1982. Avoidance of suspended sediment by juvenile coho salmon. North Am. J. Fish Manage. 2, 371-374.
- Brisbin, Jr. I.L., Beyers, R.J., Dapson, R.W., Geiger, R.A., Gentry, J.B., Gibbons, J.W., Smith, M.H., and Woods, S.K. 1974. Patterns of radiocesium in the sediments of a stream channel contaminated by production reactor effluents. Health Phys. 27, 19-27.
- Boubee, J.A., Dean, T.L., West, D.W., and Barrier, R.F.G. 1997. Avoidance of suspended sediment by the juvenile migratory stage of six New Zealand native fish species. NZ J. Mar. Freshwat. Res. 31, 61-69.
- Bunt, C.M., Cooke, S.J., Schreer, J.F., and Philipp, D.P. 2004. Effects of incremental increases in silt load on the cardiovascular performance of riverine and lacustrine rock bass, *Ambloplites rupestris*. Env. Poll. 128, 437-444.
- Carlton, W.H., Bauer, L.R., Evans, A.G., Geary, L.A., Murphy, Jr. C.E., Pinder, III J.E., and Strom, R.N. 1992. Cesium in the Savannah River Site environment. Westinghouse Savannah River Company, Aiken, SC 29808. Report No. WSRC-RP-92-250. Final Report, 1992.
- Chiasson, A. 1993a. The effect of suspended sediments on ninespine stickleback, *Pungitius pungitius*, and golden shiner, *Notemigonus crysoleucas*, in a current of varying velocity. Environ. Biol. Fish. 37, 283-295.
- Chiasson, A. 1993b. The effects of suspended sediment on rainbow smelt (Osmerus mordax): a laboratory investigation. Can. J. Zool. 71, 2419-2424.
- Cohen, A.S., Bills, R., Cocquyt, C., and Caljon, A. 1993. The impact of sediment pollution on the biodiversity of Lake Tanganyika. Conserv. Biol. 7, 667-677.
- De Robertis, A., Ryer, C.H., Veloza, A., and Brodeur, R.D. 2003. Differential effects of turbidity on prey consumption of piscivorous and planktivorous fish. Can. J. Fish Aquat. Sci. 60, 1517-1526.

- Goldberg, S., Kapoor, B.S., and Rhoades, J.D. 1990. Effect of aluminum and iron oxides and organic matter on flocculation and dispersion of arid zone soils. Soil Sci. 150, 588-593.
- Gradall, K.S., and Swenson, W.A. 1982. Responses of brook trout and creek chubs to turbidity. Trans. Am. Fish Soc. 111, 392-395.

Grobbelaar, J. 1985. Phytoplankton productivity in turbid waters. J. Plankton Res. 7, 653-663.

- Henley, W.F., Patterson, M.A., Neves, R.J., and Lemly, A.D. 2000. Effects of sedimentation and turbidity on lotic food webs: a concise review for natural resource managers. Rev. Fish Sci. 8, 125-139.
- Hinton, T.G., Kaplan, D.I., Knox, A.C., and Serkiz, S.M. 1999. *In situ* remediation of ¹³⁷Cs contaminated wetlands using naturally occurring minerals. Westinghouse Savannah River Company, Savannah River Site, Aiken, SC. 29808. Report No. WSRC-TR-99-00229. Final Report, 1999.
- Hinton, T.G., Knox, A., Kaplan, D.I., and Serkiz, S.M. 2001. An *in situ* method for remediating ¹³⁷Cs-contaminated wetlands using naturally occurring minerals. J. Radioanal. Nucl. Chem. 249, 197-202.
- Hinton, T.G., Knox, A., and Kaplan, D.I. 2002. Field deployment of illite clay as an *in situ* method for remediation ¹³⁷Cs-contaminated wetlands (U). Westinghouse Savannah River Company, Savannah River Site, Aiken, SC. 29808. Report No. WSRC-TR-2002-00516 and SREL-69 (UC-66E). Final Report, 2002.
- Hinton, T.G., Koo, B-J., Watson, S.I., Fletcher, D.E., Coughlin, D.P., and Standora, M.M. 2004. Short-term environmental impacts of illite clays when used as an *in situ* method for remediation ¹³⁷Cs-contaminated wetlands (U). Westinghouse Savannah River Company, Savannah River Site, Aiken, SC. 29808. Report No. WSRC-TR-2004-00125 and SREL-73. UC-66ee. Final Report, 2004.
- Hinton, T.G., Kaplan, D.I., Knox, A., Coughlin, D.P., Nascimento, R.V., Watson, S.I., Fletcher, D.E., and Bon-Jun Koo. 2006. Use of illite clay for in situ remediation of ¹³⁷Cs-contaminated water bodies: field demonstration of reduced biological uptake. Environ. Sci. Tech. 40, 4500-4505.
- Holz, J.C., Hoagland, K.D., Spawn, R.L., Popp, A., and Andersen, J.L. 1997. Phytoplankton community response to reservoir aging 1968-92. Hydrobiologia. 346, 183-192.
- Isono, R.S., Kita, J., and Setoguma, T. 1998. Acute effects of kaolinite suspension on eggs and larvae of some marine teleosts. Comp. Biochem. Physi. Part C. 120, 449-455.
- Iwata, T., Nakano, S., and Inoue, M. 2003. Impact of past riparian deforestation on stream communities in a tropical rain forest in Borneo. Ecol. Applic. 13, 461-473.
- Jowett, I.G., and Boustead, N.C. 2001. Effects of substrate and sedimentation on the abundance of upland bullies (*Gobiomorphus breviceps*). NZ J. Mar. Freshwat. Res. 35, 605-613.
- Karr, J.R., Fausch, K.D., Angermeier, P.L., Yant, P.R., and Schlosser, I.J. 1986. Assessing biological integrity in running waters: a method and its rationale. Illinois Natural History Survey Special Publication 5.
- Lake, R.G., and Hinch, S.G. 1999. Acute effects of suspended sediment angularity on juvenile coho salmon (*Oncorhynchus kisutch*). Can. J. Fish Aquat. Sci. 56, 862-867.
- Lawrence, J.G., and Ward, J.V. 1982. Effects of sediment releases from a reservoir on stream macroinvertebrates. Hydrobiologia. 96, 177-184.
- Liu, C.X., Zachara, J.M., Smith, S.C., Mckinley, J.P., and Ainsworth, C.C. 2003. Desorption kinetics of radiocesium from subsurface sediments at Hanford Site, USA. Geochim. Cosmochim. Acta. 67, 2893-2912.
- Miller, W.P., and Miller, D.M. 1987. A micro-pipette method for soil mechanical analysis. Commun. Soil Sci. Plant Anal. 18, 1-15.
- Newcombe, C.P., and MacDonald, D.D. 1991. Effects of suspended sediments on aquatic ecosystems. North Am. J. Fish Manag. 11, 72-82.
- NRCS (Natural Resources Conservation Service). 1996. Soil Survey Laboratory Methods Manual. Soil Survey Investigations. United States Department of Agriculture, Washington, DC 20250. Report No. 42. Final Report, 1996.

Page, L.M. 1983. Handbook of Darters. T. F. H. Publications Inc., Neptune City, NJ.

- Paller, M.H., Reichert, M.J.M, and Dean, J.M. 1996. The use of fish communities to assess environmental impacts in South Carolina coastal plain streams. Trans. Am. Fish Soc. 125, 633-644.
- Reid, S.M., Isaac, G., Metikosh, S., and Evans, J. 2003. Physiological response of rainbow trout to sediment released during open-cut pipeline water crossing construction. Water Qual. Res. J. Canada. 38, 473-481.
- Ruhe, R.V., and Matney, E.A 1980. Clay mineralogy of selected sediments and soils. Report prepared by the Savannah River Laboratory. E. I. dePont de Nemours and Company, Aiken, SC. 1980.
- SAS (SAS Procedure Guide). 1999. Version 8.01. SAS Inst. Inc. Cary, NC.
- Seaman, J.C., Meehan, T., and Bertsch, P.M. 2001. Immobilization of cesium-137 and uranium in contaminated sediments using soil amendments. J. Environ. Qual. 30, 1206-1213.
- Servizi, J.A., and Gordon, R.W. 1990. Acute lethal toxicity of ammonia and SS mixtures to Chinook salmon (*Oncorhynchus tshawytscha*). Bull. Environ. Contam. Toxicol. 44, 65-656.
- Servizi, J.A., and Martens, D.W. 1991. Effect of temperature, season, and fish size on acute lethality of suspended sediments to coho salmon, *Oncorhynchus kisutch*. Can. J. Fish Aquat. Sci. 48, 493-497.

- Servizi, J.A., and Martens, D.W. 1992. Sublethal responses of coho salmon (Oncorhynchus kisutch) to suspended sediments. Can. J. Fish Aquat. Sci. 49, 1389-1395.
- Sharitz, R.R., Scott, S.L., Pinder, III J.E., and Woods, S.K. 1975. Uptake of radiocesium from contaminated floodplain sediments by herbaceous plants. Health Phys. 28, 23-28.
- Shaw, E.A., and Richardson, J.S. 2001. Direct and indirect effects of sediment pulse duration on stream invertebrate assemblages and rainbow trout (*Oncorhynchus mykiss*) growth and survival. Can. J. Fish Aquat. Sci. 58, 2213-2221.
- Sheldon, A.L., and Meffe, G.K. 1993. Multivariate analysis of feeding relationships of fishes in blackwater streams. Environ. Biol. Fish. 37, 161-171.
- Sweka, J.A., and Hartman, K.J. 2003. Reduction of reactive distance and foraging success in smallmouth bass, *Micropterus dolomieu*, exposed to elevated turbidity levels. Env. Bio. Fish. 67, 341-347.
- Waters, T.F. 1995. Sediment in streams: sources, biological effects, and control. Am. Fish. Soc. Monogr. 7.
- Whicker, F.W., Pinder, III J.E., Bowling, J.W., Alberts, J.J., and Brisbin, I.L. 1990. Distribution of long-lived radionuclides in an abandoned reactor cooling reservoir. Ecol. Monogr. 60, 471-496.
- Whicker, F.W., Hinton, T.G., MacDonnell, M.M., Pinder, III J.E., and Habegger, L.J. 2004. Avoiding destructive remediation at DOE sites. Science. 303, 1615-1616.
- Wilber, D.H., and Clark, D.G. 2001. Biological effects of suspended sediments: a review of suspended sediment impacts on fish and shellfish with relation to dredging activities in estuaries. N. Amer. J. Fish. Manag. 21, 855-875.
- WSRC (Westinghouse Savannah River Company). 1998. Integrator operable unit work plan for the Fourmile Branch (U). Westinghouse Savannah River Company, Savannah River Site, Aiken, SC. 29808. Report No. WSRC-RP-97-448, Rev.0. Final Report, 1998.