

## How Toxic Is Coal Ash? A Laboratory Toxicity Case Study

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### ABSTRACT

Under a consent agreement among the Environmental Protection Agency (EPA) and proponents both for and against stricter regulation, EPA is to issue a new coal ash disposal rule by the end of 2014. Laboratory toxicity investigations often yield conservative estimates of toxicity because many standard test species are more sensitive than resident species, thus could provide information useful to the rule-making. However, few laboratory studies of coal ash toxicity are available; most studies reported in the literature are based solely on field investigations. This brief communication describes a broad range of toxicity studies conducted for the Tennessee Valley Authority (TVA) Kingston ash spill, results of which help provide additional perspective on the toxicity of coal ash. *Integr Environ Assess Manag* 2015;11:5–9. © 2014 SETAC

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### INTRODUCTION

In 2010, the Environmental Protection Agency (EPA) proposed regulation of Coal Combustion Residuals (CCR) under the Resource Conservation and Recovery Act (RCRA), which may result in more stringent controls on the disposal of CCR, particularly fly ash, at coal-fired power plants (USEPA 2010). Clearly, there is a need for a comprehensive body of peer-reviewed scientific literature from which EPA can assess environmental impacts of CCR.

Rowe et al. (2002) provided a thorough review of research on the environmental effects of CCR disposal. A brief, independent review limited to studies that included aquatic and benthic organism exposures to ash or ash basin effluents identified only 13 published articles. Ten were field studies of benthic or fish biota in 1 lentic and 10 lotic habitats (Cherry et al. 1979; Reash et al. 1988; Lemly 1997; Lohner, Reash, Willet, Fletcher 2001; Lohner, Reash, Willet, Rose 2001; Lohner, Reash, Williams 2001; Smith 2003; Reash 2004, 2012; Otter et al. 2012); and 3 were laboratory studies (Stanley et al. 2013; Wang et al. 2013; Greeley et al. 2014a), all of which were investigations of the 2008 Tennessee Valley Authority (TVA) Kingston Fossil Plant ash spill. This imbalance between field- and laboratory-based studies is likely due to prevailing opinions that laboratory studies should predict toxic effects in the field (Lemly 1985). There is a widely held acceptance that laboratory studies are conservative (particularly in the use of test organisms that are usually more sensitive to toxicants than resident species) and provide for controlled exposure conditions that exclude the noise experienced in the natural environment (Chapman 2000; Wang et al. 2004).

Fly ash contains several potentially toxic constituents including As, Hg, and Se (Reash 2012). Selenium in particular

has received attention due to its propensity to bioaccumulate in receptor tissues and to impair reproduction; thus it has been the subject of much debate in the scientific community (Chapman 1999; Deforest et al. 1999; Fairbrother et al. 1999; Hamilton 1999; Lemly 1999; Ohlendorf 1999), and a Pellston Workshop (Chapman et al. 2010). Such attention to individual constituents of CCR often leads to the notion that if an individual metal or metalloid is toxic in some setting, then it must be toxic in fly ash and perhaps the only constituent of concern. Complex mixtures rarely “behave” as their individual constituents; therefore sediments, water, and tissues require examining multiple constituents before causation can be discerned (Rowe et al. 2002). In addition, concentrations and characteristics of contaminants in CCR vary considerably depending on the sources of coal and combustion conditions (Lemly 1985; Reash 2004, 2012). It follows that environmental assessments of CCR should include field studies in conjunction with laboratory toxicity tests, the latter of which are now widely accepted measures of potential adverse effects of complex mixtures and are required by regulatory authorities for spills.

The purpose of this communication is to enter into the scientific record a summary of previously unpublished aquatic and sediment laboratory toxicity studies performed in the aftermath of the 2008 TVA Kingston ash spill into the Emory and Clinch rivers. Toxicity testing began shortly after dredging operations commenced, proceeded through a postdredging residual ash assessment monitoring period, and continues in the long-term monitoring of the affected rivers. The findings from these laboratory studies 1) fill a void in the data available for potential effects of fly ash on aquatic biota, and 2) provide additional perspective for EPA and other regulatory agencies to use in deciding how CCR should be regulated.

### METHODS

Because of the many test methods and study designs used in the studies summarized here, we present only a broad account of the methods employed. These studies fall into 4 categories: 1) Dredging Period; 2) Postdredging Residual Ash Period; 3) Long-Term Monitoring; and 4) Oak Ridge National Laboratory

All Supplemental Data may be found in the online version of this article.

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(ORNL) Studies. Details are available in Supplemental Data; Stojak et al. (2015 this issue) provide more information and analysis of results for the Postdredging Residual Ash Period studies.

### Sample collection and processing

During the dredging period, all sediment samples were collected by VibeCore™ in areas with deep deposits of ash from the failed storage cell, homogenized on-site, and shipped refrigerated to the laboratories in 5 gallon plastic containers. In the other 2 sampling and testing periods, sediment samples were collected by Ponar dredges, homogenized on-site, and shipped refrigerated to the laboratories in container sizes appropriate for the test methods. All sediment toxicity tests were conducted with overlying water collected from unaffected upstream river locations. For all dredge plume test samples, plumes were visually located by boat personnel, a Hydrolab was lowered to determine the depth of greatest turbidity, and samples were collected via peristaltic pumps at the most turbid depth. A dewatering ditch was constructed on-site at the Kingston Fossil Plant to receive dredge spoil. The effluent from this ditch entered the active ash pond, which in turn spilled over into a stilling pond that discharges to the plant's intake channel. ISCO™ samplers were used to collect samples from the stilling pond outfall.

### Study design

Toxicity study designs from TVA were approved by all state and federal stakeholders before commencement of investigations. Studies were performed by independent commercial laboratories or researchers at the Department of Energy's ORNL in accordance with the EPA, the American Society for Testing and Materials (ASTM), or the US Army Corps of Engineers (USACE) guidance documents (USEPA 1998, 2000, 2002a, 2002b; ASTM 2000). For all studies presented in this communication, toxicity is defined as a statistically-significant difference in response relative to upstream reference control samples.

### Dredging period studies

The goals of dredge period testing were to 1) assess potential effects from the dredging (elutriate and plume samples) and ash dewatering activities (stilling pond samples) to water column biota (*Ceriodaphnia dubia*, *Pimephales promelas*) and benthic biota (*Lampsilis siliquoidea* and *Lampsilis cardium*), and 2) to characterize the toxicity of ash deposits in the river system (whole ash samples) to benthic biota (*Hyalella azteca*, *L. siliquoidea*, *L. cardium*, *Lumbriculus variegatus*, *Corbicula fluminea*).

### Postdredging residual ash period studies

The primary purpose of postdredging studies was to assess potential toxicity of the ash remaining in the Emory and Clinch rivers after dredging operations ceased. These were long-term exposures of aquatic (*C. dubia*) and benthic organisms, including the epibenthic sentinel species *H. azteca* and infaunal species *Chironomus dilutus*, to ash-contaminated sediments and upstream controls. A total of 8 sediment samples were collected from impacted areas in each of the Emory and Clinch rivers. Three-brood *C. dubia* chronic tests were performed, and 10-day screening tests with *H. azteca* and *C. dilutus* were conducted to determine which 3 samples from each river exhibited the greatest toxicity. These, along with 1 sample from

each river that exhibited no toxicity were used in 28-day *H. azteca* survival and growth studies and in partial life-cycle *C. dilutus* survival, growth, and emergence studies.

### Long-term monitoring studies

The goal of long-term monitoring toxicity studies is to assess the potential toxicity for benthic organisms as part of the overall Monitored Natural Recovery removal action. The 10-day survival and growth test with *H. azteca* is being used in the ongoing monitoring. The first round of testing was conducted in November 2013, and the next round is tentatively scheduled in the fall of 2017.

### Oak Ridge National Laboratory studies

Oak Ridge National Laboratory designed and conducted a research investigation to examine the potential effects of direct long-term contact and water-borne coal ash exposures on fish survival and reproduction, and on early development of offspring using *P. promelas*. Adult fish were exposed to ash from the Kingston spill or reference sediment for 120 days in replicate 20 gallon flow-through aquaria, with cumulative survival and egg production of adults as endpoints, as well as the survival and frequency of developmental abnormalities during 7-day embryo-larval tests conducted with offspring produced after the completion of the long-term parental exposures (Greeley et al. 2014b). A related combination field and laboratory study conducted in the spring of 2011 further evaluated the potential for fish affected by the ash release to produce offspring with developmental deformities. That study used a 2-way (crossover) ANOVA experimental design site involving the in vitro spawning in the laboratory of redear sunfish (*Lepomis microlophus*) collected from exposed or reference sites, followed by 7-day embryo-larval tests in both site and reference water (Greeley et al. 2014c).

## RESULTS

Summary information (study description, test media, toxicity observed) for each toxicity study sponsored by TVA following the 2008 Kingston fly ash spill is presented in Supplemental Data. During this period, laboratory toxicity tests have used 8 sentinel species. From the total of 215 studies performed to date, 173 (80%) resulted in no statistically significant toxicity relative to upstream water and sediment controls. The Supplemental Data includes laboratory toxicity reports of these studies.

### Dredging period studies

The dredge core samples collected in the Emory River consisted almost entirely of fly ash released from the failed dredge cell (>80% ash). However, only tests with *H. azteca* exposed to whole ash resulted in statistically-significant toxicity for all 4 core samples. Follow-up bioavailability studies with *H. azteca* exposures to whole ash and porewater treated with an anionic exchange resin indicated that reduced survival was due to physical properties of fly ash, whereas reduced growth was due to metal or metalloid constituents (Table 1; Supplemental Data). The ash sample used in that study was 86% fly ash, consisting of 77.9% solids with a specific gravity of 2.650; the grain size distribution was 7.5% sand, 68.4% silt, and 24.1% clay. This ash had a total organic carbon (TOC) content of 540 mg/kg and no detectable acid volatile sulfides.

Exposures of newly-transformed freshwater juvenile mussels to whole ash and ash elutriates resulted in significant toxicity in

**Table 1.** Summary of unpublished laboratory toxicity test results in the aftermath of TVA's Kingston Fossil Plant fly ash release of December, 2009

	Study description	Test media	Nr of studies	Toxicity observed? <sup>a</sup>	
				No	Yes
Dredging period	96 h <i>Ceriodaphnia dubia</i> elutriate toxicity	Emory ash cores	8	7	1
	96 h <i>Pimephales promelas</i> elutriate toxicity	Emory ash cores	8	6	2
	10 d <i>Lampisilis spp.</i> elutriate toxicity	Emory ash cores	8	4	4
	10 d <i>Hyalella azteca</i> survival and growth	Emory ash cores	4	0	4
	5 d juvenile <i>Lampisilis siliquoidea</i> survival	Emory ash cores	2	2	0
	5 d juvenile <i>Lampisilis cardium</i> survival	Emory ash cores	2	1	1
	10 d Juvenile <i>Lampisilis siliquoidea</i> survival	Emory ash cores	2	1	1
	10 d Juvenile <i>Lampisilis cardium</i> survival	Emory ash cores	2	1	1
	4 d <i>Lumbriculus variegatus</i> (prebioaccumulation)	Emory ash cores	4	4	0
	28 d <i>Corbicula fluminea</i> bioaccumulation	Emory ash cores	2	2	0
	3 Brood <i>Ceriodaphnia dubia</i> survival and reproduction	Dredge plume	3	3	0
	7 d <i>Pimephales promelas</i> survival and growth	Dredge plume	1	1	0
	3 Brood <i>Ceriodaphnia dubia</i> survival and reproduction	Stilling pond effluent	3	3	0
	7 d <i>Pimephales promelas</i> survival and growth	Stilling pond effluent	1	1	0
	96 h <i>Ceriodaphnia dubia</i> survival	Dredge plume	24	24	0
	96 h <i>Pimephales promelas</i> survival	Dredge plume	24	24	0
	96 h <i>Ceriodaphnia dubia</i> survival	Stilling pond effluent	23	22	1
	96 h <i>Pimephales promelas</i> survival	Stilling pond effluent	23	23	0
	10 d <i>Hyalella azteca</i> bioavailability study with ash and porewater	Bulk ash	1	0	1
	Postdredging residual ash period	3 Brood <i>Ceriodaphnia dubia</i> survival and reproduction	Clinch River sediment	8	8
3 Brood <i>Ceriodaphnia dubia</i> survival and reproduction		Emory River sediment	8	8	0
10 d <i>Hyalella azteca</i> survival and growth		Clinch River sediment	8	7	1
10 d <i>Chironomus dilutus</i> survival and growth		Clinch River sediment	8	6	2
10 d <i>Hyalella azteca</i> survival and growth		Emory River sediment	8	3	5
10 d <i>Chironomus dilutus</i> survival and growth		Emory River sediment	8	1	7
28 d <i>Hyalella azteca</i> survival and growth		Clinch River sediment	4	2	2
Partial life cycle <i>Chironomus dilutus</i> survival, growth, emergence		Clinch River sediment	4	3	1
28 d <i>Hyalella azteca</i> survival and growth		Emory River sediment	4	0	4
Partial life cycle <i>Chironomus dilutus</i> survival, growth, emergence		Emory River sediment	4	0	4
LTM period	10 d <i>Hyalella azteca</i> survival and growth	Clinch River sediment	2	2	0
	10 d <i>Hyalella azteca</i> survival and growth	Emory River sediment	2	2	0
ORNL studies	120 d <i>Pimephales promelas</i> survival and reproduction (ORNL)	Bulk ash	1	1	0
	7 d <i>Pimephales promelas</i> embryo-larval (ORNL)	Emory River sediment	1	1	0
Totals			215	173	42

LTM = long-term monitoring; TVA = Tennessee Valley Authority.

<sup>a</sup>Toxicity based on a statistically significant difference in responses of the investigative sample relative to an upstream reference control sample.

44% of the studies. In bioaccumulation studies, *C. fluminea* exhibited no significant bio-uptake of metals or metalloids. A prebioaccumulation study with *L. variegatus* demonstrated that this species was unable to burrow into the substrate. Because ash is comprised of a variety of sizes of spherical particles, as it sits ash self-compacts into an almost concrete-like mass nearly impenetrable to burrowing organisms. *H. azteca* reportedly burrow into the upper layer of sediment under laboratory conditions (Ingersoll et al. 2000), but they, too, were unable to exhibit burrowing activity in these exposures to whole ash samples. All plume studies and all but 1 stilling pond effluent sample study during this phase of testing resulted in no toxicity to *C. dubia* and *P. promelas* (Table 1; Supplemental Data).

#### Postdredging residual ash period studies

In the residual ash investigations, the 10-day screening toxicity studies with *H. azteca* exposures to whole sediments collected from the Clinch River resulted in toxicity for 1 of 8 samples, whereas *C. dilutus* studies with Clinch River samples resulted in toxicity for 2 of 8 samples. For Emory River samples, the 10-day screening studies resulted in toxicity for 5 of 8 *H. azteca* studies and 7 of 8 *C. dilutus* studies. In long-term definitive toxicity tests, 2 of 4 *H. azteca* studies exhibited toxicity for Clinch River sediment samples, whereas all 4 Emory River studies resulted in measurable toxicity. In the partial life cycle studies with *C. dilutus*, 1 of 4 Clinch River samples resulted in toxicity whereas all 4 Emory River samples exhibited some degree of toxicity (Table 1; Supplemental Data; Stojak et al. 2015).

#### Long-term monitoring studies

In the long-term monitoring 10-day studies conducted in November 2013 with *H. azteca*, no toxicity was detected for either species (Table 1; Supplemental Data).

#### Oak Ridge National Laboratory studies

In the nonconventional assessments conducted by ORNL, no toxicity attributable to coal ash exposure was observed with adults, embryos, or larvae of *P. promelas* during long-term laboratory exposures, or with *L. microlophus* during in vitro laboratory spawning tests with adults exposed in situ for over 2 years after the ash release (Greeley et al. 2014b, 2014c).

## DISCUSSION

The results from this diverse range of studies overwhelmingly indicate that there was no toxicity of the Kingston fly ash in exposures to aquatic organisms inhabiting the overlying water column. Results from an extended ash elutriate study with fathead minnows (Stanley et al. 2013) and an embryolarval development study with fathead minnows exposed directly to fly ash (Greeley et al. 2014a) further support these findings. These laboratory studies (Table 1; Supplemental Data) and a study with amphipod, chironomid, and freshwater mussels exposed to fly ash (Wang et al. 2013) support the finding that the potential toxicity to benthic species (both infaunal and epifaunal) is minimal to moderate, depending on the species and the percentage of ash present in the sediment.

The baseline ecological risk assessment (BERA) performed to evaluate potential risks to biota from the residual ash in Watts Bar Reservoir included correlations of risk with concentrations

of contaminants in sediments. That analysis indicated that only river sediments containing greater than 40% ash are likely to cause toxicity to benthic fauna (ARCADIS 2012). The toxicity was primarily correlated with exposure to As, and to a lesser extent with covarying ash-related metals or metalloids (ARCADIS 2012, Stojak et al. 2015). Analysis of benthic community structure showed, at most, a weak correlation with percent ash further illustrating the lack of benthic invertebrate population and community-level effects (Buys et al. 2015). The BERA's conclusion that resident benthic invertebrate populations in the Emory River were, at most, at moderate-to-low risk due to residual ash contaminants reflected a counterbalancing of the sediment toxicity results by the benthic community survey findings (Walls et al. 2015). The BERA results, including the long-term toxicity tests, played a significant role in selecting Monitored Natural Recovery as the removal action for the residual ash, and the sediment toxicity test results were critical in developing project remedial goals (Carriker et al. 2015). The long-term monitoring toxicity test results (November 2013) of no measurable toxicity for Emory River and Clinch River sediment samples (35% and 21% ash, respectively) support those findings (Table 1; Supplemental Data).

Preliminary analysis of data for porewaters collected at various locations and times throughout the Kingston Ash Recovery Project by investigators using various collection and porewater extraction methods seems to show a pattern of porewater As concentrations similar to the toxicity results (Supplemental Data). Relatively low and stable As concentrations occurred at less than approximately 40% to 50% ash content in sediments, followed by a roughly linear relationship between porewater As and ash content above that point. Although the data must undergo more rigorous analysis to examine this apparent relationship, this preliminary analysis also supports the correlation of benthic species toxicity with As reported in the BERA. It suggests that up to approximately 40% ash, native sediments have some capacity to bind As, thereby reducing its bioavailability.

## CONCLUSIONS

The wealth of data derived from the conservative laboratory toxicity tests conducted since the 2008 fly ash release to the Emory and Clinch rivers clearly indicate that the risks to resident species are moderate and limited to locations with ash content greater than 40%. An additional investigative study demonstrated that benthic species may be prone to adverse effects from physical attributes of ash (survival) in addition to chemical effects (growth). Although the BERA findings indicated a correlation of ash content and As to toxicity, benthic population, and community structure were only weakly related to ash content. Consequently, subsequent risk management decisions were counterbalanced by benthic community survey findings. Sampling and testing included in the comprehensive long-term monitoring plan (benthic and fish population studies in addition to laboratory toxicity studies) support the findings from preliminary testing that there will be no long-term effects from this fly ash release for aquatic and benthic biota, with natural recovery sufficient for areas with residual ash. The findings from the laboratory toxicity studies do not contradict, but rather, are supportive of peer-reviewed literature accounts of field studies at other sites, and emphasize the importance of coal source and site-specificity to toxicity that has been elucidated very clearly in those reports.

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## SUPPLEMENTAL DATA

### Data S1.

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