#20

Dellas

ECOTOXICOLOGICAL IMPLICATIONS OF AQUATIC DISPOSAL OF COAL COMBUSTION RESIDUES IN THE UNITED STATES: A REVIEW*

CHRISTOPHER L. ROWE[1]*, WILLIAM A. HOPKINS[2] and JUSTIN D. CONGDON[2]

[1] *University of Maryland Center for Environmental Science, Chesapeake Biological Laboratory, Solomons, Maryland, U.S.A.;* [2] *University of Georgia, Savannah River Ecology Laboratory, Aiken, South Carolina, U.S.A.*

(author for correspondence, e-mail: Rowe@cbl.umces.edu)*

(Received 18 April, 2001; accepted 14 March, 2002)

Abstract. We provide an overview of research related to environmental effects of disposal of coal combustion residues (CCR) in sites in the United States. Our focus is on aspects of CCR that have the potential to negatively influence aquatic organisms and the health of aquatic ecosystems. We identify major issues of concern, as well as areas in need of further investigation.

Intentional or accidental release of CCR into aquatic systems has generally been associated with deleterious environmental effects. A large number of metals and trace elements are present in CCR, some of which are rapidly accumulated to high concentrations by aquatic organisms. Moreover, a variety of biological responses have been observed in organisms following exposure to and accumulation of CCR-related contaminants. In some vertebrates and invertebrates, CCR exposure has led to numerous histopathological, behavioral, and physiological (reproductive, energetic, and endocrinological) effects. Fish kills and extirpation of some fish species have been associated with CCR release, as have indirect effects on survival and growth of aquatic animals mediated by changes in resource abundance or quality. Recovery of CCR-impacted sites can be extremely slow due to continued cycling of contaminants within the system, even in sites that only received CCR effluents for short periods of time.

The literature synthesis reveals important considerations for future investigations of CCR-impacted sites. Many studies have examined biological responses to CCR with respect to Se concentrations and accumulation because of teratogenic and reproductively toxic effects known to be associated with this element. However, the complex mixture of metals and trace elements characteristic of CCR suggests that biological assessments of many CCR-contaminated habitats should examine a variety of inorganic compounds in sediments, water, and tissues before causation can be linked to individual CCR components. Most evaluations of effects of CCR in aquatic environments have focused on lentic systems and the populations of animals occupying them. Much less is known about CCR effects in lotic systems, in which the contaminants may be transported downstream, diluted or concentrated in downstream areas, and accumulated by more transient species. Although some research has examined accumulation and effects of contaminants on terrestrial and avian species that visit CCR-impacted aquatic sites, more extensive research is also needed in this area. Effects in terrestrial or semiaquatic species range from accumulation and maternal transfer of elements to complete recruitment failure, suggesting that CCR effects need to be examined both within and outside of the aquatic habitats into which CCR is released. Requiring special attention are waterfowl and amphibians that use CCR-contaminated sites during specific seasons or life stages and are highly dependent on aquatic habitat quality during those periods.

* Contribution 3558-CBL from the Chesapeake Biological Laboratory.

Environmental Monitoring and Assessment **80:** 207–276, 2002.

Whether accidentally discharged into natural aquatic systems or present in impoundments that attract wildlife, CCR appears to present significant risks to aquatic and semiaquatic organisms. Effects may be as subtle as changes in physiology or as drastic as extirpation of entire populations. When examined as a whole, research on responses of aquatic organisms to CCR suggests that reducing the use of disposal methods that include an aquatic slurry phase may alleviate some environmental risks associated with the waste products.

Keywords: accumulation, aquatic animals, coal ash, electric power, energy, heavy metals, sublethal effects, trace elements

1. Introduction

Coal is widely recognized as a fuel source associated with substantial environmental impacts. Mining, transport, and storage of coal are associated with habitat degradation and environmental pollution (Dvorak *et al.*, 1977). Large-scale, industrial combustion of coal produces both air-borne and solid wastes, the former having been under stringent regulation by federal and state governments for several decades. In contrast, solid coal combustion residues (hereafter CCR) which account for 90% of fossil fuel combustion wastes in the U.S. (USEPA, 1988) remain only under state regulation, which varies in rigor by jurisdiction. In some states, basic environmental protection standards for CCR disposal sites such as use of groundwater monitoring programs, leachate collection systems, and impermeable impoundment liners are not required. For example, in a national survey of 259 coal utilities having greater than 100 megawatt capacity, nearly 40% reported operating under no standards for groundwater quality (EPRI, 1997).

Federal regulations on CCR disposal remain in exemption following the 1980 Bevill Amendment to the Resource Conservation and Recovery ACT (RCRA; USEPA, 1988). The rationale for the amendment to RCRA was that: 1) the wastes were produced in large volumes, 2) there was little information available on characteristics and environmental behavior of the wastes, and 3) the limited data available suggested that risks posed by the wastes were low (EPRI, 1997). However, research conducted in the past two decades has revealed that CCR is a chemically complex mixture that can pose substantial risks to the environment. In particular, mounting evidence suggests that disposal of CCR in natural and man-made aquatic systems results in environmental degradation and poses health risks to wildlife. The goal of this paper is to review the literature related to environmental risks posed by aquatic disposal of CCR and to make recommendations for future research. Our purpose is not to provide a thorough review of CCR disposal technologies, or chemical and physical properties of CCR. Treatments of these and related issues are available in the literature (Adriano *et al.*, 1980; Roy *et al.*, 1981; EPRI, 1987a and b; Bignoli, 1989; Sharma *et al.*, 1989; Eary *et al.*, 1990; Mattigod *et al.*, 1990; Carlson and Adriano, 1993; Prasad *et al.*, 1996). However, to provide general background on CCR, we provide a brief a summary below.

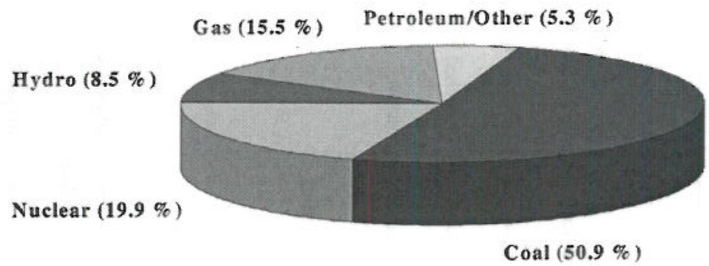

Figure 1. Net electricity generation in the U.S. by fuel source, 1999 (USDOE, 2000).

The organization of the main body of this review follows a typical risk assessment format, beginning with a discussion of sources of exposure to organisms and leading to discussions of accumulation, lethal and sublethal effects on individuals, and ecological (population and community-level) effects. While the tables are meant to provide exhaustive references to pertinent studies as well as provide data in support of the text, not all studies listed in tables are specifically discussed in the text. Rather, the text provides overviews of specific topic areas with reference to information in the tables when necessary. Because several systems have been particularly well-studied with respect to accumulation and/or effects, we include brief case studies based upon these systems within appropriate topic areas. Tables specifically related to the case studies are presented in the Appendix. Throughout the text and tables we refer to study organisms by the common or group names used by the original authors. Scientific names of all organisms discussed are provided in Appendix Table I.

2. Production and Disposal of CCR in the U.S.

With a growing human population, electricity demands continue to increase. Although an increased reliance on other energy sources in the U.S. in recent decades has resulted in a slight decrease in dependence on coal (USDOE, 1999), the largest portion of electric utility capability in the U.S. remains fueled by coal (Figure 1; USDOE, 2000). Reliance on coal for power generation has resulted in a concomitant rise in high- and low-volume waste production, with fly ash being the largest component (see below and Table I). Technologies used to reduce airborne emission of harmful particulates such as fly ash have resulted in large volumes of these wastes being removed from exhaust stacks and the subsequent need for disposal of the particulate materials. Production of fly ash, which makes up approximately 60% of the CCR waste stream, has increased in the U.S. from about 24 million tonne in 1970 to nearly 57 million tonne in 1998 (EPRI, 1997; EPA, 1997; ACAA, 1998; Figure 2).

TABLE I

Characteristics of high and low volume CCR (Van Hook, 1979; Carlson and Adriano, 1992; EPRI, 1987a and b; 1997)

Waste Type	Description	Chemical Constituents
	A. High Volume Wastes	
Fly Ash	Fine particulate residue collected in emission-control devices. Comprises ~ 60% of high volume wastes.	Various elements, including As, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sr, V, Zn. Most enriched in volatile elements (e.g. As, B, Cl, F, S, Se).
Bottom Ash and Slag	Fine and coarse grain residue remaining in the boiler following combustion.	Various trace elements, including As, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sr, V, Zn.
Flue Gas Desulfurization (FGD) Wastes (Scrubber Sludge)	Fine grain residues removed from stack via addition of limestone slurry to the flue stream.	Fly- and bottom ash constituents, often enriched in Ca-S salts and carbonates.
Fluidized Bed Boiler (FBB) Wastes	Residues mixed with ash resulting from mixing limestone and coal in the furnace on an air- fluidized bed.	Ash constituents plus Ca-S salts and carbonates.
Coal Gasification Ash (CGA)	Waste produced from conversion of coal to gaseous and liquid fuels, and is similar to fly ash but contains a higher proportion of coarse particulate material.	Ash constituents, iron sulfides, acids.
	B. Low Volume Wastes	
Air Heater, Precipitator Wash Waters	Effluent generated by high pressure washing of fly ash from air heaters and precipitators.	Ash constituents.
Boiler Chemical Cleaning Wastes	Wastewater produced from descaling boiler tubes.	Ash constituents, solvents and corrosion inhibitors.
Boiler Blowdown	Low purity water resulting from continued recirculation during steam production.	Dissolved minerals, phosphate, hydrazine.
Cooling Tower Blowdown	Low purity water periodically removed from cooling systems.	Dissolved minerals, anti-fouling and anti-fungal compounds.

TABLE I
Continued.

Waste Type	Description	Chemical Constituents
Coal Pile Runoff	Runoff wastestream produced from precipitation on coal pile stores.	Trace elements, PAHs, acids (bituminous coal) or alkaline compounds (subbituminous coal).
Coal Mill Rejects	Solids rejected from milling process.	Rocks, metal fragments, minerals, hard coal, iron and sulfur compounds.
Demineralizer Regenerant and Resins	Acidic and basic solutions from regenerating ion exchange beds.	Acids, bases, mineral salts.
Surface drainage	Collected runoff from floors, yards, and low pressure service water.	Various organic and inorganic materials.

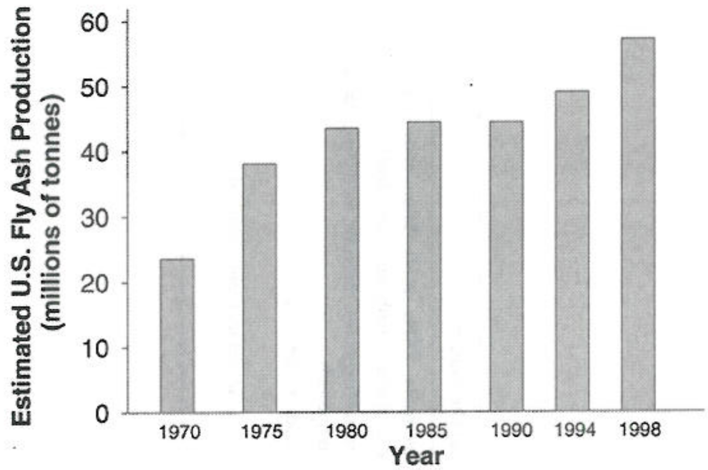

Figure 2. Estimated annual production of fly ash in the U.S., 1970 to 1998 (EPRI, 1997; USEPA, 1997; ACAA, 1998).

Because enormous quantities of wastes are produced from coal combustion, there has been a need for economically efficient disposal systems. An economically-attractive disposal method has been aquatic disposal, which is less labor intensive than land-or mine-filling (Carlson and Adriano, 1993). Typically, aquatic disposal of CCR involves pumping slurried wastes from the production site to constructed basins that, in many cases, ultimately discharge into natural water bodies. Aquatic basins serve as a physical treatment, relying on gravitational settling of particulate material from the slurried waste stream. Approximately 45% of coal-fired power plants rely on aquatic basins for disposal of CCR (EPRI, 1997). In terms of volume disposed, approximately two-thirds of CCR was disposed of using aquatic basins prior to 1980 (EPRI, 1997). Today, aquatic basins still account for disposal of approximately one-third of CCR produced (EPRI, 1997; Figure 3).

3. Composition of CCR

The composition of CCR can be quite variable (Tables I and II), reflecting differences in parent coal composition (Dvorak, 1977, 1978), inclusion of other fuels in the combustion processes, combustion and cleaning technology, and disposal techniques (Carlson and Adriano, 1993). Because coal is itself a concentrated source of many trace elements, oxidation and loss of carbon from the solid substrate during combustion produces a residual ash material that is further concentrated in non-volatile elements. Addition of materials collected from boiler flues and air scrubbing units to the bulk CCR stream can return volatile components to the CCR stream which would otherwise have been lost during combustion. Moreover, waste

TABLE II

Inorganic and organic constituents of fly- and bottom ash components of CCR. Inorganic contents were determined in ash samples derived from 42 coal-fired power plants in the U.S., and are presented as ranges of means (ppm dry mass). Organic analyses are for semi-volatile EPA-priority organic pollutants in methylene chloride concentrates of base/neutral fractions of 6 fly ash and 7 bottom ash samples selected from the 42 power plants providing samples for analyses. Concentrations of organic compounds are presented as ranges of means (ppb in extract). All data from EPRI (1987a). Note that samples were analyzed for a total of 42 organic contaminants; only those two which were detected are included below

Element	Fly	Bottom	Element	Fly	Bottom	Element/Compound	Fly	Bottom
Al	46000–152000	30500–14500	Mo	7–236	3–443	V	< 95–652	< 50–275
As	8–1385	< 5–37	Na	1300–62500	814–41300	U	11–30	< 5–26
Ba	251–10850	150–9360	Ni	23–353	< 10–1067	Zn	27–2880	4–515
Ca	7400–223000	2200–241000	P	1100–10340	< 500–4630	bis(2-ethylhexyl)phthalate	17–286	6–204
Cd	6–17	< 5	Pb	21–2120	5–843	Di-n-octylphthalate	ND - 3.2	ND - 6.2
Cl	180–1190	< 150–2630	S	1300–64400	460–74000			
Cr	37–651	< 40–4710	Sb	11–131	< 10			
Cu	45–1452	27–146	Se	6–47	< 2–10			
Fe	25000–177000	20200–201000	Si	89500–275000	51000–312000			
K	3000–25300	2600–24000	Sn	8–56	< 9–90			
Mg	1600–41800	2500–46000	Sr	204–6820	182–6460			
Mn	44–1332	56–1940	Ti	1310–10100	1540–11300			

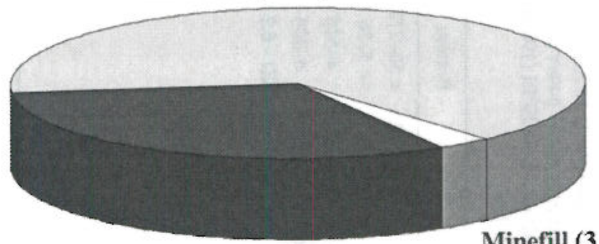

Figure 3. Percentage of CCR disposed of in landfills, aquatic basins, and minefills in the U.S. (EPRI, 1997).

management practices vary among facilities, and may entail combining numerous waste products associated with coal combustion and typical plant operations into a single, chemically complex CCR effluent. Depending upon the site in question, the CCR stream can thus contain a variety of waste types, including fly ash (typically the largest component), bottom ash, flue gas desulfurization (FGD) wastes, fluidized bed boiler (FBB) wastes, coal gasification ash (CGA), and multiple types of low volume comanaged wastes (EPRI, 1997). The result of modern, industrial coal combustion practices is thus a solid CCR waste enriched in numerous elements and compounds, some of which may pose risks of toxicity to organisms that interact with the wastes in natural or man-made habitats (Tables I and II). Of the three commonly employed disposal techniques (landfills, aquatic basins, and minefills), comanagement of multiple waste types is most prevalent at facilities using aquatic basins for disposal. In a survey of 259 disposal facilities, 91% of sites using aquatic basins simultaneously disposed of high and low volume waste types, whereas 70 and 75% of landfills and minefills, respectively, received the mixed effluents (EPRI, 1997).

The largest proportion of CCR is in the form of solids such as ash (USEPA, 1988) that contain a variety of potentially toxic elements and compounds (Tables I and II). Thus, from the standpoint of potential environmental impacts associated with CCR, the solid ash fraction appears to be a component of CCR that requires particular attention. The emphasis of this paper will be on environmental impacts of solid CCR in aquatic environments, with a primary focus on effects on aquatic organisms. Moreover, we will focus on inorganic contaminants associated with CCR disposal in aquatic systems which appear to be much more prevalent than organic contaminants (Table II), and thus have received greater attention from researchers.

4. Environmental Impacts of CCR in Aquatic Systems

4.1. Exposure to Contaminants

4.1.1. *Sources of Contaminants to Biota*

Disposal of CCR into aquatic systems can physically and chemically alter habitat conditions via sedimentation and changes to sediment particle size distribution, turbidity, pH, conductivity, and inputs of contaminants (Theis, 1975; Carlson and Adriano, 1993; Dvorak 1977, 1978). Numerous aquatic systems have been studied with respect to these habitat modifications, the focus primarily being on inorganic contaminants associated with CCR. Concentrations of several trace elements (primarily As, Cd, Cr, Cu, Pb, and Se) have been particularly well characterized in several CCR-impacted systems because of the abundance of these elements in CCR and/or concerns associated with the known toxicological actions of these elements. Whereas in some systems the focus of chemical screening was primarily on dissolved fractions of one or a few trace elements in water, surveys in other systems suggest that numerous trace elements are elevated in CCR-impacted systems not only in water, but also in suspended solids and sediments (Table III).

The results of chemical surveys presented in Table III reflect the elevated concentrations of contaminants associated with CCR in dissolved and particle-associated forms. However, to examine the potential risks that elevated CCR-derived contaminants in aquatic systems may pose for wildlife, the propensity for contaminants to be accumulated from the environment must be examined, as must the biological responses associated with contaminant accumulation. These topics are treated in the following sections of this document.

4.1.2. *Trace Element Accumulation by Biota*

There is a large amount of data demonstrating that plants and animals inhabiting CCR-contaminated sites or chronically exposed to CCR in laboratory or field-based experiments accumulate trace elements, sometimes to very high concentrations (Table IV). Accumulation of trace elements from water and sediments by vascular and non-vascular plants suggests the potential for trophic transfer of bioaccumulative elements to grazers. For example, in the D-Area facility, SC, numerous types of producers accumulated trace elements from sediments and/or water, themselves apparently serving as vectors of the contaminants to several grazing invertebrates (Table IV; Cherry and Guthrie, 1976, 1977; Guthrie and Cherry, 1979). Occurrence of some trace elements at very high concentrations in micro- and macroinvertebrates also suggests that predatory vertebrates may accumulate some trace elements to levels that may ultimately result in lethal or sublethal effects (Hopkins, 2001). In Stingy Run, OH, high tissue burdens of some contaminants in odonates may have been a source of contaminants to several species of fish which accumulated trace elements in numerous tissues (Table IV; Lohner and Reash, 1999; Reash *et al.*, 1999). Such relationships between tissue trace element

TABLE III

Mean or ranges of trace element concentrations in water (ppb), suspended solids (ppm dry mass), and sediments (ppm dry mass except where noted) in aquatic sites contaminated by CCR. NR = not reported, BDL = below detection limits. Decimal places reflect those presented by the original authors

Site	Description	As	Cd	Cr	Cu	Pb	Se	Reference
			Water (ppb)					
Belews Lake, NC	Prior to ash effluent discharge	BDL	NR	NR	NR	NR	BDL	Olmsted *et al.*, 1986
Belews Lake, NC	Ash effluent entering lake	190–253	NR	NR	NR	NR	157–218	Cumbie, 1978
Belews Lake, NC	Lake water, 2 yr following initial ash effluent discharge	4–10	NR	NR	NR	NR	7–14	Cumbie, 1978
Belews Lake, NC	Lake water, 2 yr following initial ash effluent discharge	6.6	NR	NR	NR	NR	12.6	Olmsted *et al.*, 1986
Belews Lake, NC	Lake water, 5 yr following initial ash effluent discharge	4.3	NR	NR	NR	NR	9.5	Olmsted *et al.*, 1986
Belews Lake, NC	Lake water, 8 yr following initial ash effluent discharge	3.1	NR	NR	NR	NR	8.8	Olmsted *et al.*, 1986
Belews Lake, NC	Lake water, 22 yr following initial ash effluent discharge, 11 yr after discharge had ceased	NR	NR	NR	NR	NR	< 1.0	Lemly, 1997
Martin Creek Reservoir, TX	Fly ash ponds discharging into reservoir	NR	NR	NR	NR	NR	2,200–2,700	Garrett and Inman, 1984
Columbia Generating Station, WI	Drainage from ash pit entering Rocky Run Creek	NR	2.4–2.9	35–65	4–43	NR	NR	Magnuson *et al.*, 1980

TABLE III
Continued.

Site	Description	As	Cd	Cr	Cu	Pb	Se	Reference
Fruitland, NM	Ash pond surface water	33	1	3	2	NR	60	Dreesen *et al.*, 1977
Fruitland, NM	Ash pond effluent water	27	1	2	3	NR	57	Dreesen *et al.*, 1977
Lansing, NY	Farm pond receiving airborne drift of coal ash	NR	NR	NR	NR	NR	0.35	Gutenmann *et al.*, 1976
Harrodsburg, KY	Ash settling pond	NR	0.46	NR	4.38	NR	NR	Benson and Birge, 1985
Roger's Quarry fly ash reservoir, Oak Ridge, TN	During period of active use	NR	NR	NR	NR	NR	25	Southworth *et al.*, 1994
Roger's Quarry fly ash reservoir, Oak Ridge, TN	After cessation of discharge	NR	NR	NR	NR	NR	< 2	Southworth *et al.*, 1994
Stingy Run, OH	Stream draining ash reservoir, measurements prior to ash effluent inputs	BDL	NR	BDL	NR	NR	BDL	Reash *et al.*, 1988
Stingy Run, OH	Stream draining ash reservoir; measurements following ash effluent inputs[a]	21–24	NR	62–129	NR	NR	19–33	Reash *et al.*, 1988
Stingy Run, OH	Stream draining ash reservoir [b]	< 4–14.3	0.7–0.8	1.6–29.7	2.9–6.2	< 2–2.1	3.2–11.8	Lohner and Reash, 1999; Lohner *et al.*, 2001
Little Scary Creek, WV	Stream drainage ash reservoir	64	NR	NR	13	NR	32	Reash *et al.*, 1999
Glen Lyn, VA	Ash basin input [c]	NR	30–43	NR	270–2,880	NR	NR	Cairns and Cherry, 1983
Glen Lyn, VA	Ash basin outfall [c]	NR	2–150	NR	5–20	NR	NR	Cairns and Cherry, 1983

concentrations in fish and accumulation by invertebrate prey species were apparent in several systems in which biotic samples were surveyed (Table IV). Note that some authors have reported body burdens in concentrations per unit wet mass, whereas others have reported concentrations relative to dry mass. We indicate in the tables the different ways in which concentrations were presented by the original authors.

The importance of trophic vectors for trace element accumulation by vertebrates in CCR-contaminated systems was demonstrated by a recent series of experiments on the lake chubsucker, a benthic fish. Exposure to CCR-contaminated sediments alone (with uncontaminated water and food provided) resulted in rapid accumulation of trace elements (Table IV; Hopkins *et al.*, 2000b). When the same species of fish was exposed to CCR under semi-natural mesocosms conditions (water, sediments, and prey collected from the CCR disposal site), trace element accumulation was much greater than in fish previously exposed to sediments alone (Table IV; Hopkins, 2001), and effects on growth and survival were greatly exacerbated. Trace element accumulation by invertebrates was likely the most important factor influencing accumulation by fish, and led to body burdens in fish more than an order of magnitude higher than burdens found in fish exposed to contaminated sediments alone (i.e., provided with uncontaminated water and food; Table IV).

Amphibians, reptiles, birds, and mammals also accumulate contaminants from CCR-contaminated sites as a result of their feeding niche/trophic status, and/or long life spans which expose them to contaminants over exceptionally long periods of time (Table IV). For example, the banded water snake is a relatively long-lived predator with high trophic status (preying upon other vertebrates such as fish and amphibians). Banded water snakes collected from a CCR-contaminated system have the highest hepatic concentrations of Se and As yet reported in a reptile (Table IV; Hopkins *et al.*, 1999a). In addition, a series of laboratory studies with the banded water snake demonstrated the importance of ingestion of contaminated prey items in accumulation of contaminants. Adult and juvenile snakes were fed contaminated prey items (fish) collected from a CCR-contaminated swamp (D-area site, SC) for up to two years. Resulting accumulation was pronounced, with particularly high concentrations of Se accumulating in liver, gonads, and kidney (Table IV; Hopkins *et al.*, 2001; Hopkins *et al.*, 2002a). Concentrations of Se greatly exceeded concentrations known to induce reproductive failure in birds and fish (Lemly, 1993, 1996). Moreover, snakes fed alternating diets of contaminated and uncontaminated prey (Hopkins *et al.*, 2002a) also accumulated Se burdens above the reproductive toxicity thresholds proposed by Lemly (1993, 1996). Results from these studies suggest that even periodic feeding on prey items derived from CCR-contaminated sites can result in high tissue burdens in predatory vertebrates. Therefore, terrestrial vertebrates inhabiting nearby habitats could accumulate trace elements from prey items dispersing from the contaminated sites, even if the remaining portion of a predator's diet consists of prey items with no history of contaminant exposure.

A particularly well-studied system with respect to trace element accumulation in aquatic vertebrates as a result of CCR contamination is Hyco Reservoir, NC. Investigators have examined several tissues in numerous species of fish to quantify Se accumulation. Hyco Reservoir is thus examined more thoroughly in the case study to follow.

4.1.3. *A Case Study of Selenium Accumulation by Fish: Hyco Reservoir, NC*
Hyco Reservoir is a 1764 ha cooling reservoir serving a 2495 MW coal-fired power plant in Roxboro, North Carolina. As well as heated water discharge, the reservoir also received effluents from coal fly ash basins (CPL, 1981). Fish declines and a fish kill in autumn of 1980 (CPL, 1981) prompted several investigations to examine coal-related contaminants and potential effects on the aquatic community within the reservoir. Here we provide an overview of Se accumulation by fish in Hyco Reservoir, because of the large number of species examined in that system. Biological responses to Se accumulation in Hyco Reservoir are presented elsewhere in this document where sublethal and ecological effects of CCR are considered (Sections 4.2 and 4.3).

Water chemistry surveys in Hyco Reservoir found that dissolved Se concentrations were quite high (Table III), whereas waterborne concentrations of other CCR-derived trace elements did not appear to be elevated (CPL, 1981). Measurements of organic contaminants (PAHs, PCBs, pesticides, herbicides) showed no elevations above detection limits (CPL, 1981). Sampling of fish tissues revealed similar patterns as did the water chemistry surveys: fish inhabiting Hyco Reservoir experienced significant tissue burdens of Se, while other trace elements (Hg, As, Cu, Cr, Zn) were not elevated above normal (Appendix Table II; CPL, 1981). Tissue levels of organic contaminants (PAHs, PCBs, pesticides, herbicides) were below detection limits, except for DDD and DDE which were detectible but within normal background concentrations (CPL, 1981). Because of the predominance of Se in water and tissues, subsequent investigations of the Hyco system focused primarily on Se accumulation and its effects on aquatic organisms (Appendix Table II).

Selenium accumulation was observed in several trophic groups in Hyco Reservoir. Accumulation of Se by plankton may have been a source of Se accumulation to planktivorous and ultimately higher-level predatory fish (Appendix Table II). Selenium accumulation varied among fish species. Muscle Se concentrations were generally highest for bluegill and several other sunfish, and lowest for catfish (Appendix Table II). Liver Se concentrations in bluegill collected from Hyco Reservoir were about 50 times greater than liver concentrations in reference fish (Sager and Cofield, 1984), and were considerably higher than liver Se concentrations of other species (Appendix Table II). Gonadal Se concentrations also appeared higher for bluegill sunfish than other species and there were sex-specific differences in Se concentrations in gonads; ovarian Se concentrations were about twice the concentrations observed in testes (Appendix Table II; Sager and Cofield, 1984; Baumann and Gillespie, 1986). Moreover, bioaccumulation led to Se concentrations in ovaries of bluegills about 1000 times above ambient water concentrations (Baumann and Gillespie, 1986).

It is clear from studies to date that, when CCR is discharged into aquatic systems, some potentially toxic trace elements in water, sediments, and suspended solids (Table III) are accumulated by biota and further transferred through the food web (Table IV; Appendix Table II). Biological responses resulting from exposure and accumulation would thus be predicted. For example, the propensity for Se to accumulate in fish from Hyco Reservoir, especially within ovarian tissues, suggests that some species in this system may have been at risk of reproductive impairment. Demonstrated lethal and sublethal responses of biota to CCR-derived contaminants will be the subject of the following sections.

4.2. Effects of CCR on Individuals

4.2.1. *Lethal Effects*

Lethality of CCR to aquatic organisms has been observed in laboratory and field studies (Table V). For example, comparative studies by Birge (1982) showed that CCR effluent was acutely toxic to embryonic fish and amphibians in the laboratory (Table V). Birge (1982) also conducted laboratory bioassays to examine relative toxicities of 22 individual CCR-related elements to goldfish, rainbow trout, and narrow-mouth toads. Based upon comparisons of 7 and 28 d LC_{50} values, narrow mouth toads were found to be the most sensitive species to 17 of the elements (in order of decreasing toxicity: Hg, Zn, Cr, Cu, Cd, As, Pb, Co, Ge, Al, Sn, Se, Tl, Sr, Sb, Mn, W), whereas rainbow trout were most sensitive to 5 elements (Ag, La, Ni, V, Mo). Acute laboratory studies on other vertebrates and invertebrates have also demonstrated lethality responses by several species when exposed to water, sediments, or suspended solids from CCR-contaminated sites (Table V).

Field and outdoor mesocosm studies also suggest that for some species, acute or chronic exposure to CCR can ultimately be lethal (Table V). For example, in a 5 d field-caging study, shrimp, darters, and salamanders were extremely sensitive to conditions in a CCR-contaminated site, whereas other invertebrates and fish experienced much lower mortality rates (Table V; Guthrie and Cherry, 1976). A recent exposure of benthic fish in outdoor mesocosms for 45 days indicated that prolonged exposures to CCR, as would occur in contaminated habitats, may result in extremely high mortality (75%; Hopkins, 2001).

As a whole, results of field- and laboratory-based lethality studies (Table V) suggest that, if lethality is to be used as an endpoint for examining ecological risks of CCR, numerous species must be simultaneously examined due to extreme species-specific differences in sensitivity. Particular attention should be devoted to the duration and conditions of exposure; a recent study indicates that reductions in resource abundance during chronic exposure to CCR increases the sensitivity of fish to CCR (Hopkins *et al.*, 2002a). Moreover, the absence of a lethal response by

TABLE V

Results of studies of lethality of CCR to aquatic animals. Tissue trace element concentrations were usually unmeasured or unreported in these studies. When tissue burdens were measured, the reference is denoted by '[a]' and concentrations are included in Appendix Table IV

Species	Exposure method	Exposure duration	Observed effect(s)	Reference
		Invertebrates		
Amphipod	Laboratory exposure to water from ashpit drainage ditch	4 d	Low survival of early instars compared to adults	Magnuson *et al.*, 1981
Shrimp	Caged *in situ* at drainage basin outflow	5 d	100% mortality	Guthrie and Cherry, 1976
Shrimp	Caged *in situ* in drainage basin outflow ditch	5 d	100% mortality	Guthrie and Cherry, 1976
Shrimp	Caged *in situ* at confluence of outflow ditch and a creek	5 d	45% mortality	Guthrie and Cherry, 1976
Odonates	Caged *in situ* at drainage basin outflow	5 d	50% mortality	Guthrie and Cherry, 1976
Odonates	Caged *in situ* in drainage basin outflow ditch	5 d	No mortality	Guthrie and Cherry, 1976
Odonates	Caged *in situ* at confluence of outflow ditch and a creek	5 d	No mortality	Guthrie and Cherry, 1976
Crayfish	Caged *in situ* at drainage basin outflow	5 d	No mortality	Guthrie and Cherry, 1976
Crayfish	Caged *in situ* in drainage basin outflow ditch	5 d	No mortality	Guthrie and Cherry, 1976
Crayfish	Caged *in situ* at confluence of outflow ditch and a creek	5 d	No mortality	Guthrie and Cherry, 1976
		Fish		
Channel catfish	Caged *in situ* at drainage basin outflow	5 d	No mortality	Guthrie and Cherry, 1976
Channel catfish	Caged *in situ* in drainage basin outflow ditch	5 d	No mortality	Guthrie and Cherry, 1976
Channel catfish	Caged *in situ* at confluence of outflow ditch and a creek	5 d	No mortality	Guthrie and Cherry, 1976
Mosquitofish	Caged *in situ* at drainage basin outflow	5 d	40% mortality	Guthrie and Cherry, 1976
Mosquitofish	Caged *in situ* in drainage basin outflow ditch	5 d	No mortality	Guthrie and Cherry, 1976
Mosquitofish	Caged *in situ* at confluence of outflow ditch and a creek	5 d	No mortality	Guthrie and Cherry, 1976
Largemouth bass	Caged *in situ* at drainage basin outflow	5 d	20% mortality	Guthrie and Cherry, 1976
Largemouth bass	Caged *in situ* in drainage basin outflow ditch	5 d	No mortality	Guthrie and Cherry, 1976

TABLE V
Continued.

Species	Exposure method	Exposure duration	Observed effect(s)	Reference
Largemouth bass	Caged *in situ* at confluence of outflow ditch and a creek	5 d	No mortality	Guthrie and Cherry, 1976
Darters	Caged *in situ* at drainage basin outflow	5 d	100% mortality	Guthrie and Cherry, 1976
Darters	Caged *in situ* in drainage basin outflow ditch	5 d	100% mortality	Guthrie and Cherry, 1976
Darters	Caged *in situ* at confluence of outflow ditch and a creek	5 d	33% mortality	Guthrie and Cherry, 1976
Largemouth bass, fingerlings	Stocking of isolated coves of reservoir receiving coal ash effluent with 200,000 fingerlings.	7 d	100% mortality	Olmsted *et al*., 1986
Channel catfish, juveniles	Caged *in situ* for exposure to acidic seepage from a coal ash pond	2 wk	Secretion of protective mucus; 100% mortality	Coutant *et al*., 1978
Rainbow trout	Exposure to different concentrations of suspended ash in static systems	96 hr	Mortality at some concentrations; no discernible pattern	Cairns and Cherry, 1983
Bluegill sunfish	Exposure to different concentrations of suspended ash in static systems	96 hr	Mortality of 30 to 80% of individuals at 1800–6000 ppm Total Suspended Solids	Cairns and Cherry, 1983
Banded sculpin	Released into coal ash-impacted stream 2–3 yrs after cessation of discharge into stream	Multi-year	No effects detected	Carrico and Ryan, 1996
Red ear sunfish, embryos	Laboratory exposure to fly ash effluent dilutions (water only)	3 d	100% mortality in undiluted effluent; 58% mortality in effluent dilted to 10%	Birge, 1978
Goldfish, embryos	Laboratory exposure to fly ash effluent dilutions (water only)	3 d	43% mortality in undiluted effluent; 24% mortality in effluent dilted to 10%	Bridge, 1978
Lake chubsuckers, juveniles	Laboratory exposure to sediments from a CCR impacted site (uncontaminated water and food provided)	124 d	25% mortality	Hopkins *et al*., 2000b[a]

TABLE V
Continued.

Species	Exposure method	Exposure duration	Observed effect(s)	Reference
Lake chubsuckers, juveniles	Laboratory exposure to sediments from a CCR impacted site (uncontaminated water and food provided); Three ration levels provided	78 d	10% mortality in fish provided with medium and high rations; 60% mortality in fish provided with low rations	Hopkins *et al.*, 2002b[a]
Lake chubsuckers, juveniles	Laboratory exposure to sediments from a CCR impacted site (uncontaminated water and food provided)	100 d	17% mortality	Hopkins, 2001[a]
Lake chubsuckers, juveniles	Outdoor mesocosm exposure to sediments, water, and food from a CCR impacted site	45 d	75% mortality	Hopkins, 2001[a]
		Amphibians		
Leopard frogs, embryos	Laboratory exposure to fly ash effluent (water only)	2.5 d	100% mortality in undiluted effluent	Birge, 1978
Fowler's toad, embryos	Laboratory exposure to fly ash effluent (water only)	1.5 d	54% mortality in undiluted effluent	Birge, 1978
Salamanders	Caged *in situ* at drainage basin outflow	5 d	100% mortality	Guthrie and Cherry, 1976
Salamanders	Caged *in situ* in drainage basin outflow ditch	5 d	100% mortality	Guthrie and Cherry, 1976
Salamanders	Caged *in situ* at confluence of outflow ditch and a creek	5 d	80% mortality	Guthrie and Cherry, 1976
Southern toads, larvae	Caged *in situ* in CCR-ash settling basin	Entire larval period (> 60 d)	100% mortality	Rowe *et al.*, 2001a
Bullfrogs, embryos	Laboratory exposure to sediment and water collected from CCR-ash settling basin	Embryonic period (4 d)	32% mortality (10% mortality in controls)	Rowe, unpublished
Bullfrogs, embryos	Laboratory exposure to sediment and water collected from drainage swamp receiving effluent from CCR-ash settling basin	Embryonic period (4 d)	18% mortality (10% mortality in controls)	Rowe, unpublished

TABLE V
Continued.

Species	Exposure method	Exposure duration	Observed effect(s)	Reference
Bullfrogs, embryos/larvae	Laboratory exposure to sediment and water collected from CCR-ash settling basin	Embryonic period and portion of larval period (34 d total)	87% mortality (46% mortality in controls)	Rowe, unpublished
Bullfrogs, embryos/larvae	Laboratory exposure to sediment and water collected drainage swamp receiving effluent from CCR-ash settling basin	Embryonic period and portion of larval period (34 d total)	75% mortality (46% mortality in controls)	Rowe, unpublished
		Reptiles		
Banded water snakes, adults	Fed fish collected from CCR-contaminated site	2 yr	No mortality	Hopkins *et al.*, 2002a
Banded water snakes, juveniles	Fed fish collected from CCR-contaminated site	13.5 mo	No mortality	Hopkins *et al.*, 2001

organisms in acute or chronic tests should not be interpreted as lack of significant biological effects of CCR. Individuals of many species interacting with CCR in natural and artificial systems have been shown to respond sublethally, often in ways in which individual fitness may ultimately be compromised.

4.2.2. *Sublethal Effects of CCR*

Sublethal effects of CCR have been observed in numerous invertebrates and vertebrates in sites in the U.S. (Table VI, Appendix Tables III and IV). Studies have shown that several invertebrates experience changes in dispersal and metabolic processes (Table VI). Fish have been shown to exhibit numerous sublethal responses upon exposure to CCR and accumulation of trace elements. In Little Scary Creek, WV, a system receiving outflow from a CCR retention basin, bluegill sunfish experienced decreased liver weight and white blood cell counts, and elevated serum levels of sodium, potassium, and chloride, although condition factors and general morphology appeared normal (Table VI; Reash *et al.*, 1999). Perhaps the most frequently observed sublethal effects in fish exposed to CCR, however, are abnormalities in developing larvae and histopathological changes in adults. Bluegill sunfish in Hyco Reservoir that were shown to accumulate Se in ovarian tissues (Appendix Table II) produced edamatous larvae which eventually died (Table VI; Gillespie and Baumann, 1986). Also in Belews Lake, NC and other systems, fish have been observed to produce edamatous larvae, as well as to experience numerous histopathological changes (Table VI). In some cases, abnormalities in larvae were associated with reproductive failure and population declines (Section 4.3). In one CCR-contaminated system in particular (Martin Creek, TX), thorough histopathological surveys have revealed widespread changes in native fish associated with accumulation of Se. An overview of findings from histopathological studies in the Martin Creek system is presented in the following case study. In a CCR disposal site on the Savannah River Site, SC, numerous taxa have been shown to respond sublethally to multiple trace elements accumulated from CCR-contaminated sediments, water, and food. The Savannah River site is the subject of a second case study regarding sublethal responses to CCR.

4.2.3. *A Case Study of Selenium Accumulation from CCR and Sublethal Responses by Fish: Martin Creek, TX*

Martin Creek Reservoir is a 2000 ha cooling water reservoir used by a coal-fueled power plant operated by the Texas Utilities Generating Co. The reservoir, constructed in 1974, is located on Martin Creek, Texas, a tributary of the Sabine River. In September, 1978 the utility company began discharging effluents from two fly ash settling ponds into the reservoir (Sorensen *et al.*, 1982a). Shortly thereafter, fish kills in the reservoir were observed (Garrett and Inman, 1984). In May, 1979, approximately 8 months after effluent release had begun, discharge of the effluents into Martin Creek Reservoir ceased. The Martin Creek site provided a unique opportunity to examine the magnitude of biological changes that can occur following

TABLE VI

Sublethal effects of CCR associated with trace element body burdens in animals collected from CCR-contaminated sites or experimentally exposed to CCR. For experimentally exposed organisms, exposure methods are noted. Trace element concentrations are means or ranges expressed as ppm dry mass 'DM' or wet mass 'WM'. Additional information on sublethal effects is compiled in Appendix Tables III to V for systems in which case histories are presented. If known, the specific tissue(s) in which trace elelments were measured are provided. NR = not reported. BDL = below detection limit. Decimal places reflect those presented by the original authors. Scientific names for species examined are provided in Appendix Table I

Species, tissue analyzed for contaminants; protocol	As	Cd	Cr	Cu	Pb	Se	Observed effect(s)	Site (reference)
							Invertebrates	
Amphipods; held for 2d in laboratory streams containing CCR	NR	NR	NR	NR	NR	NR	Reduced downstream movements	Rocky Run Creek, WI (Webster *et al.*, 1981)
Isopods; held for 2d in laboratory streams containing CCR	NR	NR	NR	NR	NR	NR	Reduced downstream movements	Rocky Run Creek, WI (Webster *et al.*, 1981)
Crayfish, muscle (DM); caged for 62 d in ashpit drainage ditch	NR	NR	0.6–1.8	NR	NR	0.4	Reduced metabolic rate	Rocky Run Creek, WI (Magnuson *et al.*, 1981; Forbes *et al.*, 1981)
Crayfish, hepatopancreas (DM); caged for 62 d in ashpit drainage ditch	NR	NR	5.6–6.2	NR	NR	3.6–32.5	Reduced metabolic rate	Rocky Run Creek, WI (Magnuson *et al.*, 1981; Forbes *et al.*, 1981)
Crayfish, muscle (DM); caged for 62 d in creek receiving effluent from ashpit drainage ditch	NR	NR	0.5–0.8	NR	NR	0.2–0.4	Reduced metabolic rate	Rocky Run Creek, WI (Magnuson *et al.*, 1981; Forbes *et al.*, 1981)

TABLE VI

Continued.

Species, tissue analyzed for contaminants; protocol	As	Cd	Cr	Cu	Pb	Se	Observed effect(s)	Site (reference)
Crayfish, hepatopancreas (DM); caged for 62 d in creek receiving effluent from ashpit drainage ditch	NR	NR	2.8–12.6	NR	NR	2.9–12.1	Reduced metabolic rate	Rocky Run Creek, WI (Magnuson *et al*., 1981; Forbes *et al*., 1981)
Fish								
Green sunfish, skeletal muscle (WM); field collected	NR	NR	NR	NR	NR	12.9	Decreased hematocrit, increased condition factor and hepatopancreas-to-bodyweight ratio due to edema, histological abnormalities (liver, kidney, gill, heart, ovary)	Belews Lake, NC (Sorensen *et al*., 1984)
Green sunfish, liver (WM); field collected	NR	NR	NR	NR	NR	21.4	Decreased hematocrit, increased condition factor and hepatopancreas-to-bodyweight ratio due to edema, histological abnormalities (liver, kidney, gill, heart, ovary)	Belews Lake, NC (Sorensen *et al*., 1984)
Fathead minnow, whole body (WM); field collected	NR	0.2	NR	0.6	NR	NR	Decreased sensitivity to metals in acute exposures	Harrodsburg, KY, ash settling pond (Benson and Birge, 1985)
Fathead minnow, internal organs (WM); field collected	NR	0.7	NR	1.9	NR	NR	Decreased sensitivity to metals in acute exposures, perhaps due to metallothionein production	Harrodsburg, KY, ash settling pond (Benson and Birge, 1985)

TABLE VI

Continued.

Species, tissue analyzed for contaminants; protocol	As	Cd	Cr	Cu	Pb	Se	Observed effect(s)	Site (reference)
Fathead minnow, gills (WM); field collected	NR	0.4	NR	1.7	NR	NR	Decreased sensitivity to metals in acute exposures, perhaps due to metallothionein production	Harrodsburg, KY, ash settling pond (Benson and Birge, 1985)
Bluegill, liver (DM); field collected	5.4	4.2	3.5	33.5	NR	53.8	Leukopenia, elevated serum salts, decreased liver mass	Little Scary Creek, WV (Reash *et al.*, 1999)
Bluegill, ovary (DM); field collected	0.6	0.1	2.3	5.8	NR	23.4	Leukopenia, elevated serum salts, decreased liver mass	Little Scary Creek, WV (Reash *et al.*, 1999)
Bluegill, testes (DM); field collected	3.1	0.6	8.3	7.8	NR	24.5	Leukopenia, elevated serum salts, decreased liver mass	Little Scary Creek, WV (Reash *et al.*, 1999)
Bluegill, carcass (WM); field collected	0.05–0.11	0.007–0.01	NR	0.36–0.99	0.05–0.26	6.90–7.20	Reproductive failure	Hyco Reservoir, NC (Gillespie and Baumann, 1986)
Bluegill larvae, whole body (DM); larvae derived from crosses of adults from CCR-contaminated site	NR	NR	NR	NR	NR	28.20	Edema and reduced larval survival	Hyco reservoir, NC (Gillespie and Baumann, 1986)
Bluegill fingerlings, muscle (WM); caged for 8 d in lake receiving CCR	< 0.01–0.03	NR	NR	NR	NR	0.6–3.4	Erratic swimming, exophthalmia, abdominal distention	Belews Lake, NC (Olmsted *et al.*, 1986)
Bluegill fingerlings, viscera (WM); caged for 8 d in lake receiving CCR	< 0.02–0.20	NR	NR	NR	NR	3.6–7.5	Erratic swimming, exophthalmia, abdominal distention	Belews Lake, NC (Olmsted *et al.*, 1986)