Overview of Coal Combustion Residuals (CCR) and TVA Kingston Case Study

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Executive Summary

Coal is formed from the plant or animal organic matter trapped in sedimentary rock. There are four types of coal, including lignite (formed at shallow burial depths), sub-bituminous coal (formed at depths of ~500 to 2,000 meters), bituminous coal (formed at depths of ~2,000 to 5,000 meters), and anthracite (formed below ~5,000 meters). The amount of energy stored in the coal increases with depth.

World coal production and consumption has been increasing since the International Energy Agency (IEA) began gathering data in 1971, but decreases in both production and consumption were observed from 2014 to 2016. However, coal remains the second-largest source of electricity generation in the United States. As a result, a large volume of coal combustion residuals (CCR) is generated annually in the US from coal-fired power plants. For example, ~107 million tons of CCR were generated in the US in 2016. There are four types of CCR, including fly ash, bottom ash, boiler slag and flue gas desulfurization (FGD) materials.

The Resource Conservation and Recovery Act (RCRA) addresses the nationwide management of municipal and industrial solid wastes. CCR has been within the realm of regulation under RCRA since 1978, when six "special wastes" (including CCR) became exempt from regulation as hazardous waste under RCRA Subtitle C. Since 1978, there has been debate over whether CCR should be regulated under RCRA, and whether that regulation should be as hazardous waste (under Subtitle C) or as nonhazardous waste (under Subtitle D). In 2008 there was a catastrophic failure of a dike at the Tennessee Valley Authority's Kingston, Tennessee coal-fired power plant, leading to the release of 5.4 million cubic yards of CCR into a system of rivers. A massive cleanup operation commenced. As a result of public outcry over this incident, the EPA began the process of proposing to regulate CCR under RCRA for the first time. In 2016, the EPA finalized a rule to regulate CCR as nonhazardous waste under RCRA Subtitle D. The EPA identified that there are risks posed to human health and the environment by CCR, but that risk could be

mitigated with regulations under Subtitle D. However, it deferred a final determination on the hazardous characteristics of CCR until a later date.

CCR that is beneficially re-used is exempt from the rule. However, if CCR is placed into a landfill or surface impoundment, then groundwater monitoring around that unit is required. This requirement demonstrates the EPA's recognition that certain trace elements may leach from CCR. Trace elements identified by the EPA in their final CCR rule as subject to groundwater monitoring include As, B, Ba, Be, Cd, Co, Cr, F$^-$, Hg, Li, Mo, Pb, Sb, Se, Tl, and Ra-226 and Ra-228 combined. These as well as additional trace elements have been demonstrated to be present in CCR, with the concentrations varying based upon the source of the coal and type of CCR. It has also been demonstrated that trace elements will leach from CCR, with the nature and composition of the leachate varying with the source of the coal, the type of CCR and environmental conditions (primarily pH).

Given that the Tennessee Valley Authority (TVA) Kingston release has been cited by the EPA as an impetus for the final CCR rule, it will be discussed here as a case study. The TVA Kingston release resulted in contamination of a river system with trace elements including As and Se. 3.5 million cubic yards of CCR were removed from the river system and disposed of offsite, while ~2.3 million cubic yards of CCR were compacted and disposed of onsite within a 240 acre disposal cell. 510,000 cubic yards of residual CCR are still present. CCR that was released contained As predominantly in the oxidized form (arsenate, As^V) and Se predominantly in the reduced form Se^{IV}. Once deposited in the Emory River, As remained primarily as As^V while 30 to 50% of Se in the CCR transformed from Se^{IV} to reduced species of Se (including organic Se^{II}). Total concentrations of As and Se in the CCR and in sediment indicate impacts to the environment, and moderate toxicity has been observed. In order to evaluate long-term impacts from the onsite landfill and the residual CCR, long-term monitoring began in 2013 and is expected to continue for 30 years.

The TVA Kingston release demonstrates a clear need to regulate the structural components of CCR units, but environmental impacts from the leaching of trace elements is less clear. The environment appears to have recovered even with the presence of 510,000 cubic yards of residual CCR in the system of rivers, and no additional remediation is planned. Also, an ecological risk assessment showed that leaching of trace elements from the onsite 240 acre landfill containing 2.3 million cubic yards of CCR was not a significant exposure pathway for ecological receptors.

1 Coal Production and CCR Generation

1.1 Coal Formation

Coal is formed from the plant or animal organic matter trapped in sedimentary rock. In order for fossil fuels to have formed, reducing environments must have been present when the organic material was deposited, preventing the oxidation of complex organic hydrocarbon material (Robb, 2005). As shown in **Figure 1**, in the marine environment, burial of primarily phytoplankton results in the formation of kerogen (sedimentary organic matter that is insoluble in organic solvents and contains a polymer-like structure), oil and gas, while in the terrestrial environment, burial of primarily land-derived plants results in the formation of peat, lignite, sub-bituminous coal, bituminous coal and anthracite
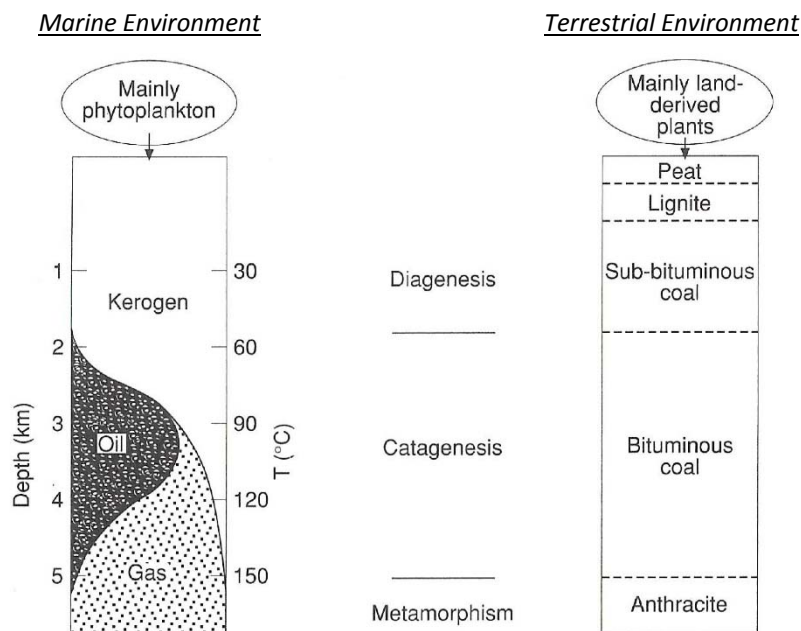
Figure 1 – Formation of fossil fuels in marine and terrestrial environments.
*Figure from Robb, 2005.*

Peat is a precursor to coal and consists of the fibrous remains of organic material, and forms when the rate of organic material accumulation exceeds the rate of decomposition. Peat accumulation occurs in marshes, bogs and swamps, where reduced environments limit the oxidation of the organic

material, allowing for the accumulation and preservation of that material (Brady & Weil, 2008). One large period of organic matter accumulation that resulted in the formation of coal was the Carboniferous Period of the Paleozoic Era, occurring ~299 to 359 million years ago. During the Carboniferous Period, a warm, moist climate resulted in wetlands being far more extensive than today, resulting in the development of large lowland swamps with increased peat accumulation (Stanley, 2005).

As indicated in **Figure 1**, peat and lignite are formed at shallow burial depths above ~500 meters within the terrestrial environment, with sub-bituminous coal formed at depths of ~500 to 2,000 meters (Robb, 2005). Bituminous coal is formed at depths of ~2,000 to 5,000 meters, and anthracite is formed below ~5,000 meters. The amount of energy stored in the coal increases with depth.

1.2   Coal Production and Consumption

The information and figures in this section are pulled from the International Energy Agency (IEA) Coal Information 2017 report (IEA, 2017), which provides detailed worldwide statistics for coal
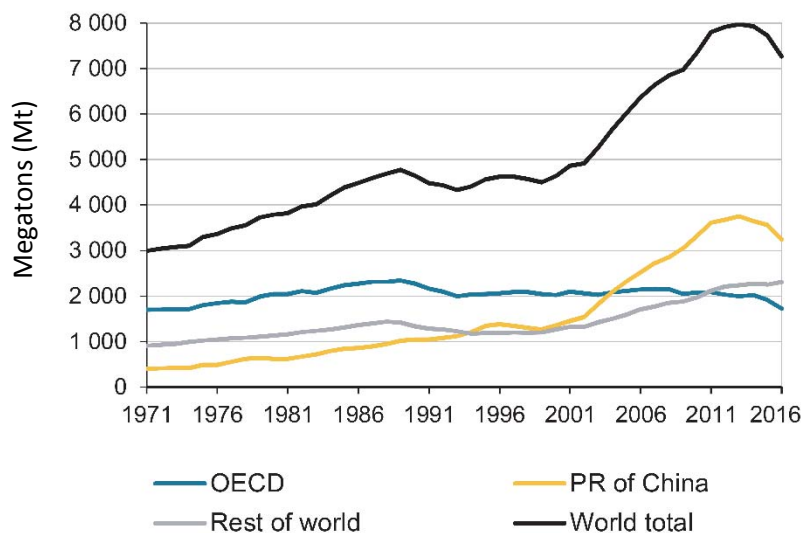
Figure 2 *– World total coal production from 1971 – 2016. OECD refers to 35 counties that are members of the Organization for Economic Co-operation and Development, including the United States and many European countries. Figure from IEA, 2017.*

production and consumption. For the purposes of IEA's report, coal is defined to include hard coal (anthracite), bituminous coal (coking coal and other bituminous coal), and brown coal (sub-bituminous coal and lignite). As indicated in **Figure 2**, world coal production has increased steadily since 1971. However, between 2014 and 2016 a decrease in production was observed, from 7,934.1 Megatons (Mt) in 2014 down to 7,268.6 Mt in 2016 (IEA, 2017). China has been the world's leading coal producer since 1985, while production in the US has been declining since 2008.

As shown in **Figure 3**, world coal consumption has also been increasing since 1971, from 2,000 megatons of coal equivalent (Mtce) in 1971 to 5,360 Mtce in 2016 (IEA, 2017). However, consumption has decreased steadily since 2014, from 5,600 Mtce in 2014 to 5,360 in 2016. China has largely been driving consumption as the world's largest consumer of coal, but changes in their economic growth model and concerns about air pollution have decreased consumption. Consumption in the US reached its peak in 2005, at 796 Mtce in 2005 dropping to 494 Mtce in 2016 (IEA, 2017).
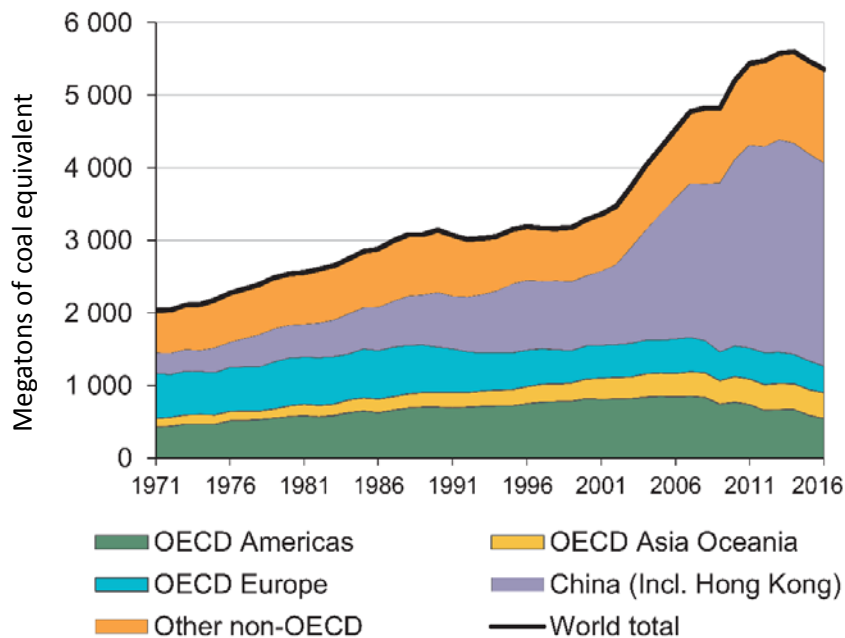
Figure 3 – World total coal consumption from 1971 – 2016. Megatons of coal equivalent is the sum of anthracite, other bituminous coal, coking coal, sub-bituminous coal and lignite converted to a common energy unit. Figure from IEA, 2017.

1.3   CCR Generation in the United States

Although coal consumption is dropping in the United States, coal remains the second-largest source of electricity generation in the country. According to the U.S. Energy Information Administration (EIA), in 2016 1,239,149 thousand megawatt hours of electricity generated in the US was generated using coal, versus 4,131,693 thousand megawatt hours of energy production (U.S. EIA, 2017). Therefore ~30% of electricity generated in the US in 2016 was generated from coal.

As a result of this consumption, a large volume of coal combustion residuals (CCR) is generated annually. There are four types of CCR: fly ash, bottom ash, boiler slag and FGD materials (USEPA, 2017a). Fly ash is produced when finely ground coal is burned in a boiler to produce electricity, while bottom ash is produced during the burning of coal, with particles being too coarse and heavy to be carried up the smoke stacks. Boiler slag is molten bottom ash with a smooth and glassy texture that is collected at the bottom of slag-type and cyclone-type furnaces. FGD materials are products of the process used to reduce sulfur dioxide emissions from the exhaust system of a coal-fired boiler. According to a survey conducted by the American Coal Ash Association, 107 million tons of CCR were produced in the US in 2016 (American Coal Ash Association, 2016).

2 CCR Regulation

2.1 Regulatory History

RCRA addresses the nationwide management of municipal and industrial solid waste. CCR has been within the realm of regulation under RCRA since 1978 (USEPA, 1978). These standards included a proposal to exempt six "special wastes" from the RCRA Subtitle C regulations pending further study. One of the six special wastes was utility waste, including fly ash and bottom ash. The possible hazard of trace elements was identified in the proposed rule.

In 1980, Congress amended RCRA with the Solid Waste Disposal Act Amendments of 1980 (U.S. Congress, 1980). Under the "Bevill amendment", the EPA excluded certain solid wastes related to the

energy industry from regulation as hazardous waste, including CCR. This exemption was temporary, pending the EPA's consideration of eight "Bevill factors." The eight Bevill factors consisted of: "1) the source and volumes of such material generated each year; 2) present disposal and utilization practices; 3) potential danger, if any, to human health and the environment from the disposal and reuse of such materials; 4) documented cases in which danger to human health or the environment from surface runoff or leachate has been proved; 5) alternatives to current disposal methods; 6) the costs of such alternatives; 7) the impact of those alternatives on the use of coal and other natural resources; and 8) the current and potential utilization of such materials" (U.S. Congress, 1980). The EPA was required to follow up and report on these factors to determine whether these wastes should be regulated as hazardous wastes.

The EPA missed the 1982 deadline to report to Congress on CCR. The final report, titled "Report to Congress on Wastes from Combustion of Coal by Electric Utility Power Plants," was instead submitted in 1988 (USEPA, 1988). The report concluded that trace elements in CCR including fly ash, bottom ash, boiler slag and flue gas emission control waste could pose risks to human health and the environment, but those wastes generally do not exhibit RCRA hazardous waste characteristics. The reports also determined that regulating CCR as hazardous waste would vastly increase the costs of disposal.

Following up on its 1988 Report to Congress, the EPA published its first regulatory determination on CCR in 1993 (USEPA, 1993). In this report, it was concluded that regulation of CCR as hazardous waste under RCRA Subtitle C was not appropriate as CCR posed limited risks, and there were already adequate state and federal regulatory programs in place for these materials. The EPA's second regulatory determination in 2000 addressed the management of additional coal combustion wastes not addressed in the 1993 determination, and came to similar conclusions (USEPA, 2000).   This determination also allowed beneficial reuse of CCR.

In 2008, there was a catastrophic failure of a dike at TVA's Kingston coal-fired power plant, leading to the release of 5.4 million cubic yards of fly ash and bottom ash sludge over a 300 acre area including a branch of the Emory River. The release was followed by a massive cleanup operation overseen by the EPA and the state of Tennessee. As a result of public outcry over this incident, and in order to follow up on its 2000 regulatory determination, the EPA began the process of proposing to regulate CCR under RCRA for the first time (USEPA, 2017b).

There were two options within the proposed rulemaking (USEPA, 2010). The first was to reverse the August 1993 and May 2000 Bevill regulatory determinations and list CCR as a special waste subject to regulation under RCRA Subtitle C (as hazardous waste) when being disposed of in landfills or surface impoundments. The second option was to leave the Bevill determination in place and regulate CCR under RCRA Subtitle D (as nonhazardous waste) by issuing national minimum criteria. Under both options, the EPA was proposing to establish safety requirements for dams to address the structural integrity of surface impoundments to prevent catastrophic releases.

The final rule was published in the Federal Register in 2015 (USEPA, 2015a), with the rule becoming effective in 2016 (USEPA, 2017b). The EPA decided to regulate CCR as nonhazardous waste under RCRA Subtitle D, rather than as hazardous waste under Subtitle C. The EPA identified that there are risks posed to human health and the environment by CCR, but that risk could be mitigated with new regulations under Subtitle D. These regulations include requirements for CCR landfills and surface impoundments, with exemptions for CCR that is beneficially used.

2.2 Beneficial Reuse

When CCR is beneficially reused, it is not subject to the final rule. In order to fall under the category of beneficial reuse, the use of CCR must fulfill the following criteria, it must provide a functional benefit; it must be a substitute for a virgin material; it must meet product specifications and/or design standards; and if the CCR is unencapsulated and 12,400 tons or more are used in a non-roadway

application, it must be demonstrated that environmental releases to groundwater, surface water, soil and air will be similar or less than that of the original material, and that the releases are below regulatory levels to protect human and ecological receptors (USEPA, 2017c).

According to a survey, 56% of the 107 million tons of CCR produced in 2016 was beneficially reused (American Coal Ash Association, 2016). There are challenges that prevent a larger percentage of CCR from being beneficially reused. Material that does not meet the criteria listed above cannot be beneficially reused. In addition, utility companies have identified numerous challenges regarding beneficial reuse of CCR. A survey of utility companies presented at the 2013 World of Coal Ash Conference identified the following challenges: flux in the market for CCR, including inconsistencies in demand and distance to end users; regulatory impacts, including uncertainty about regulation and the negative public image of CCR; economic concerns, including the cost of rail and truck transportation; unrelieablility of the supply; and difficulties associated with internal management and coordinating the re-use (Rokoff et al., 2013).

Beneficial reuse falls into two categories: encapsulated and unencapsulated (USEPA, 2017c). Encapsulated use consists of binding the CCR such that its mobility to the environment is minimized – for example, binding it in wallboard, concrete, roofing material or bricks. Unencapsulated use consists of using the CCR in an unbound form, such as in structural fill or embankments.

Fly ash is primarily used as a replacement for Portland cement in concrete, potentially increasing the strength of the concrete and reducing production costs (Yao et al., 2015). 38% of the 22.6 million tons of fly ash produced in 2016 was reused as an ingredient in concrete, concrete products or grout (American Coal Ash Association, 2016). The EPA has determined that replacement of non-CCR products with fly ash in concrete mixtures results in similar or decreased environmental impacts, and that any releases are at or below regulatory requirements designed to be protective of human health and the environment (USEPA, 2014b). Other uses for fly ash are as structural fill or in embankments, and mining

applications. Fly ash may also be used as a geopolymer binder in pervious concrete, with bottom ash as the coarse aggregate material (Zaetang et al., 2015).

It has been demonstrated rare earth elements including dysprosium (Dy), erbium (Er), europium (Eu), neodymium (Nd), scandium (Sc), terbium (Tb) and yttrium (Y) are present in fly ash generated from burning coal in the US (Taggart et al., 2016). Rare earth element concentrations vary depending upon the source of the coal, but concentrations are sufficiently high that production of rare earth elements from fly ash may be feasible. However, the feasibility of recovery is limited by available extraction technologies (Meawad et al., 2010).

Bottom ash is typically used as a cheaper alternative for base material in road construction or as a grit material for blasting, and it may also be used as a partial substitute for Portland cement in concrete mixtures (Kurama & Kaya, 2008). Other uses are as structural fill or in embankments, as an ingredient in gypsum panel products, and for waste stabilization or solidification (American Coal Ash Association, 2016).

FGD materials are primarily used as ingredients in gypsum panel products and for waste stabilization and solidification, but they may also be used as structural fill or in embankments, or as ingredients in concrete (American Coal Ash Association, 2016). The EPA has determined that the use of FGD materials in gypsum panel products results in similar or decreased environmental impacts, and that any releases are at or below regulatory requirements designed to be protective of human health and the environment (USEPA, 2014b).

2.3 Summary of Final Rule

If CCR is not beneficially reused, then it is subject to the final rule. This section summarizes the final rule – see U.S. EPA 2015a, pages 21302 to 21309. Also referenced is the EPA's webinar presentation giving an overview of the final rule (USEPA, 2015b). The final rule applies to new and existing CCR landfills and surface impoundments, including any expansion of these units, where the CCR

is generated by the combustion of coal by electrical utilities and independent power producers. The units may be located onsite or offsite. The rule also applies to inactive CCR impoundments at active sites, unless the unit is closed within 36 months of the date the final rule was published. Landfills no longer receiving CCR and CCR units at facilities where electricity is no longer produced are exempt from the rule, as is CCR generated from non-utility boilers, CCR generated from coal mines, or CCR that is beneficially used.

The rule puts into place several location restrictions for certain new and existing CCR landfills and surface impoundments (referred to as "CCR units" in the rest of this section). Restrictions are in place to ensure CCR units are placed above the uppermost aquifer in a given area, unless it can be demonstrated that there is not a connection between the CCR unit and the uppermost aquifer. CCR units must be placed outside of wetlands, unless it can be demonstrated that there is no degradation to sensitive wetland ecosystems. In addition, CCR units must be located away from fault zones and seismic impact zones, unless it can be demonstrated that structural integrity can be maintained during an earthquake within these areas. Finally, CCR units cannot be located in unstable areas, unless it can be demonstrated that the presence of these areas will not impact the structural integrity of the unit.

Further requirements include the installation of composite or alternative composite liners and the installation of leachate collection and removal systems for new CCR units; requirements focused on the structural integrity of new units; a requirement to control fugitive dust; installation of run-off and run-on controls; flood control requirements; inspection requirements; groundwater monitoring; closure and post-closure requirements; and requirements for recordkeeping, reporting, and maintenance of a publicly accessible internet site where compliance with the new rule is documented.

The groundwater monitoring requirement demonstrates the EPA's recognition that certain trace elements may leach from CCR. Groundwater monitoring is required for all CCR surface impoundments, landfills and lateral expansions. Semi-annual monitoring is required for the following parameters, which

the EPA has identified as leading indicators of releases associated with CCR: B, Ca, Cl⁻, F⁻, pH, SO_4^{2-} and total dissolved solids. If there is a statistically significant increase over background concentrations for any of these parameters, then additional groundwater sampling is required within 90 days. Samples must then be analyzed for the following parameters: As, B, Ba, Be, Cd, Co, Cr, F⁻, Hg, Li, Mo, Pb, Sb, Se, Tl, and Ra-226 and Ra-228 combined. Should additional investigation confirm that the CCR unit is the source of contamination, corrective measures are required. If the leaking unit is an unlined surface impoundment, it must either be retrofitted with a liner or closed. Otherwise, a corrective measures assessment must be conducted, followed by selection and implementation of a remedy. Remedies would vary depending upon the site, but could include isolation (capping and installation of subsurface barriers), immobilization (in-situ solidification or stabilization), chemical treatment, excavation or monitored natural attenuation.

Interestingly, the EPA again deferred a final decision regarding the Bevill amendment factors (USEPA, 2015b). As discussed above, it was a requirement of the Solid Waste Disposal Act Amendments of 1980 for the EPA to evaluate whether CCR should be regulated as hazardous waste under Subtitle C of RCRA by considering the eight Bevill factors. Those included an evaluation of whether CCR poses a risk to human health or the environment resulting from its chemical composition. The EPA has decided that additional information is needed before making a final decision regarding the Bevill factors. However, this deferment of a final decision does not prevent the beneficial reuse of CCR.

3 Composition and Leaching

Much of the new rule focuses on requirements related to siting CCR units and structural requirements for CCR units. However, the remainder of this paper will focus on the chemical composition of CCR as it relates to potential soil and groundwater contamination.

3.1 Trace Elements Composition

Meawad et al. (2010) compiled data from three studies to present the median concentration of trace elements in fly ash, bottom ash, boiler slag and FGD materials. Their data presented in **Table 1** show that trace elements are present in all four types of CCR.

As discussed in Section 4, the EPA in their final CCR rule include about 16 elements as subject to groundwater monitoring. B is the most significant of these because concentrations of B above background trigger monitoring for the full list of parameters. Twelve of these 16 trace elements are included in **Table 1** (see bolded parameters).

Table 1
Median trace element content of fly ash, bottom ash, boiler slag and flue gas desulfurization (FGD) materials, in ppm. Bold constituents are a subset of the list included as groundwater monitoring parameters in the final CCR rule. Table from Meawad et al., 2010.

Element	Fly Ash	Bottom Ash	Boiler Slag	FGD
Ag	3.2	3	37	3.3
As	43.4	4.7	4.5	
B	311	90	49.5	60
Ba	806	633	413	162
Be	5	2.2	7	29.3
Cd	3.4	3.1	40.5	3.9
Co	35.9	24	–	–
Cr	136	120	–	–
Cu	112	61.1	32	46.1
F	29	50	–	–
Hg	0.1	0.009	9.5	4.8
Mn	250	297	–	–
Ni	77.6	79.6	83	68.1
Pb	56.8	13.2	8	25.3
Sb	4.6	4	0.8	6
Se	7.7	0.8	4.5	4.5
Sr	775	800	–	–
Tl	9	NA	38.5	9
V	252	141	75	65
Zn	148	52.6	35.8	90.9

Fly ash is a mixture of fine particles including clays, quartz, iron oxides and aluminosilicate glass, and is abundant in Si, Al and Fe oxides (Deonarine et al., 2015). Fly ash also contains smaller concentrations of oxides of Ca, K, Mg, P, S and Ti. Trace elements are present in fly ash, including As, Cd, Cr, Hg, Ni, Pb, Sb, Se, U and Zn (Deonarine, Kolker, & Doughten, 2015). **Figure 4** illustrates the trace elements concentrations of three fly ash samples, compared with average concentrations for United States coals.
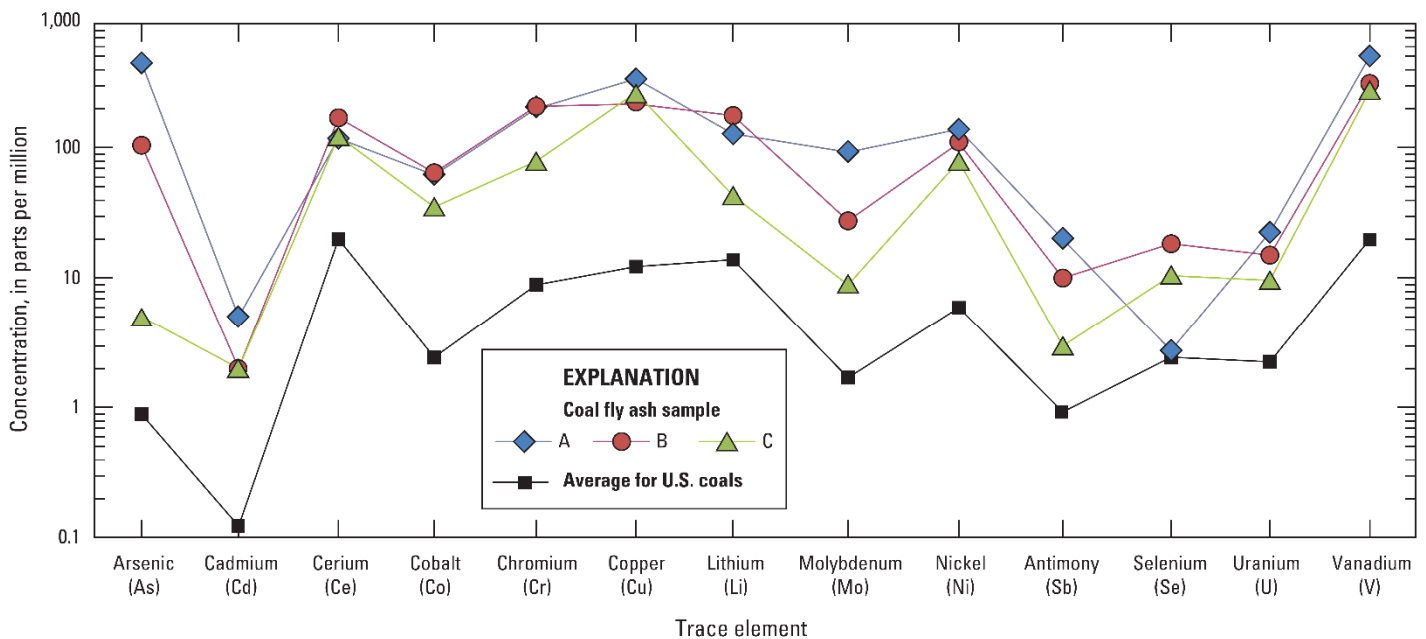
Figure 4 – Concentrations of trace elements in fly ash samples from the United States. Samples A and B are from the Appalachian Basin while sample C is from the Powder River Basin of Wyoming. An average for United States coals is also presented. Figure from Deonarine, Kolker, & Doughten, 2015.

A study of the concentrations of trace elements in 23 fly ash samples from 23 coal-fired power plants in Europe provides additional information on the chemical composition of fly ash (Moreno et al., 2005). Samples were collected from power plants in Spain, the Netherlands, Greece and Italy. The chemical composition was determined by Inductively Coupled Plasma Mass Spectrometry and Inductively Coupled Plasma Atomic Emission Spectrometry after acid-digestion of the samples. Concentrations are presented in **Table 2**.

Table 2
Trace element composition of 23 fly ash samples collected from throughout Europe. Bold constituents are included in the groundwater monitoring parameter list in the final CCR rule. Table from Blissett and Rowsan, 2012 after Moreno et al., 2005.

Element	Trace element composition (ppm)		
	25th Perc.	Median	75th Perc.
As	40	55	97
B	135	259	323
Ba	639	1302	1999
Be	6	8	12
Cd	1	2	2
Co	30	35	48
Cr	137	148	172
Cu	73	86	118
Ge	3	7	15
Hg	0.2	0.2	0.3
Li	150	185	252
Mo	7	11	13
Ni	87	96	144
Pb	59	80	109
Rb	50	108	147
Sb	4	4	8
Se	6	7	13
Sn	7	8	10
Sr	384	757	1647
Th	25	30	37
U	9	12	18
V	202	228	278
Zn	123	154	175

The results indicate that the concentrations of trace elements in CCR vary with the type of CCR and with the source of the coal from which the CCR was generated. The following section discusses the leaching of trace elements from CCR.

3.2 Leaching from CCR

There are many methods for assessing the leaching capability of CCR. Commonly used are the Toxicity Characteristic Leaching Procedure (TCLP; USEPA Method 1311), and Synthetic Precipitation Leaching Procedure (SPLP; USEPA Method 1312). However, these methods have limitations. TCLP is designed to simulate leaching of organic and inorganic compounds from a landfill and is appropriate for determining whether a RCRA solid waste is hazardous, while SPLP is designed to simulate leaching from

acidic precipitation (USEPA, 2017d). These tests are therefore simulating specific scenarios under narrow ranges of pH, and are not appropriate to evaluate the leachability of trace elements from CCR, which may exist in various forms and be exposed to various pHs in the environment.

As a result of these limitations, the EPA has developed a set of four Leaching Environmental Assessment Framework (LEAF) methods (USEPA, 2017e). The four test methods consist of the following methods:

- Method 1313 – Liquid-Solid Partitioning as a Function of Extract pH Using a Parallel Batch Extraction Procedure
- Method 1314 – Liquid-Solid Partitioning as a Function of Liquid-Solid Ratio for Constituents in Solid Materials Using an Up-Flow Percolation Column Procedure
- Method 1315 – Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials Using a Semi-Dynamic Tank Leaching Procedure
- Method 1316 – Liquid-Solid Partitioning as a Function of Liquid-Solid Ratio Using a Parallel Batch Extraction Procedure

While the use of these four tests is not a regulatory requirement, and these test methods do not replace the use of TCLP to determine whether a waste is hazardous under RCRA, the EPA does recommend using the tests in certain situations, including for the evaluation of CCR (USEPA, 2017d). The following discussion of each LEAF method is from the EPA's LEAF How-To Guide: Understanding the LEAF Approach and How and When to Use It (U.S. EPA, 2017d).

The first is Method 1313 – Liquid-Solid Partitioning as a Function of Extract pH Using a Parallel Batch Extraction Procedure. It provides a procedure for analyzing samples over a range of pHs, from pH 2 to pH 13. The test is done at a set liquid-to-solid (L/S) ratio of 10 mL/g-dry material. This is helpful for CCR leachate evaluations where the CCR may be present in the environment at varying pHs. Method 1314 – Liquid-Solid Partitioning as a Function of Liquid-Solid Ratio for Constituents in Solid Materials Using an Up-Flow Percolation Column Procedure, consists of pumping deionized water through a

column of the subject material. It provides leachate concentrations, pH and conductivity as a function

of incremental changes in and the cumulative value of the L/S ratio. This data can be used to estimate

leaching as a function of time.

Method 1315 determines Mass Transfer Rates of Constituents in Monolithic or Compacted

Granular Materials Using a Semi-Dynamic Tank Leaching Procedure.   It provides a procedure for

determining the rate of mass transport from monolithic or granular materials (such as CCR) as a function

of time. Data collected includes the rate of release of the constituents, compacted dry density, and pH

of the leachate as a function of time. Method 1316 determines Liquid-Solid Partitioning as a Function of

Liquid-Solid Ratio Using a Parallel Batch Extraction Procedure. It consists of five batch extractions over a

range of L/S values, from 0.5 to 10 mL/g-dry material.   Data collected includes constituent

concentrations and mass release, pH, and conductivity as a function of the L/S ratio. This test is useful

when the material being tested has low permeability, such as very fine grained CCR.

Data from the LEAF tests can be combined to evaluate CCR under a range of pHs, L/S ratios, and

various field conditions. TCLP and SPLP provide much more limited data given they are designed to

simulate leaching under specific scenarios of pH and L/S ratio. Given the limitations of the TCLP and

SPLP methods, the following discussion of leaching results from CCR should be viewed with a critical eye.

Fly ash may leach trace elements such as As, B, Cr, Mo, Ni, Se, Sr and V, although leaching can be

reduced through in-situ chemical treatment (Bhattacharyya et al., 2009). However, the testing was

done at a single-point pH of 4.2 using the SPLP method, therefore the results are unlikely to be

representative of typical conditions where CCR is exposed to the environment.

Da Silva et al. (2018) used two LEAF methods to evaluate the leachability and trace element

concentrations of CCR including fly ash, bottom ash and FGD materials.   CCR from seven coal-fired

power plants in Florida was analyzed using SPLP, LEAF Method 1313, and LEAF Method 1316. Using the

SPLP method, trace element concentrations in leachate were highest for fly ash and bottom ash, and

that the buffering capacity of all samples was high. Comparison of SPLP results with results from Method 1313 and Method 1316 indicated that concentrations of trace element including As, Cd, Cr, Pb and Se vary by method, and therefore pH and L/S ratio are important factors in determining the leachability of trace elements from CCR.

Based on an extensive literature review, the following elements exhibit low leachability from fly ash under typical environmental conditions when pH is 7-10: Be, Cd, Co, Cu, Fe, Mg, Mn, Ni, Pb, Si, Sn, Th, Tl, U and Zn (Izquierdo & Querol, 2012). However, within the same pH range, As, B, Cr, Mo, Sb, Se, W and V were at their maximum leachability from fly ash; As, B, Cr, Mo and Se should be of particular concern given their toxicity and high water solubility and therefore high mobility in surface water and groundwater (Izquierdo & Querol, 2012). Therefore these elements should be focused upon for the purposes of CCR management. Indeed, as discussed in Section 4, B is the primary trace element focused upon by the EPA in their final CCR rule since it triggers additional monitoring for As, Cr, Mo and Se, among other parameters.

A study of seeps, surface water and groundwater near CCR units at 15 sites in Tennessee, Kentucky, Georgia, Virginia and North Carolina found that concentrations of As, B, Ca, Fe, Mn, Mo, Se, Sr, SO_4^{2-} and V exceeded background concentrations: (Harkness et al., 2016). The study also found that B and Sr isotopes can be used to help identify leaking CCR units, linking CCR units to releases of trace elements into the environment.

As discussed in Section 4, the TVA Kingston release has been cited by the EPA as an impetus for the final CCR rule. It will therefore be discussed below as a case study, with As and Se being the primary site contaminants.

4 TVA Kingston Case Study

4.1 Project Overview

In 2008, a catastrophic failure of a dike at the TVA coal-fired power plant in Kingston, Tennessee led to the release of 5.4 million cubic yards of fly ash and coal ash sludge over a 300 acre area, including a branch of the Emory River (Natural Resource Trustees, 2015). **Figure 5** shows aerial images of the storage unit both before and after the failure. As a result, there are a number of requirements in the final CCR rule related to where CCR units may be located and structural requirements for CCR units. However, this discussion will focus on the environmental component of the release.

Following the release, in 2009 an Administrative Order on Consent (AOC) was issued by the EPA requiring cleanup under the Comprehensive Environmental Response, Compensation, and Liability Act. The AOC laid out short-term, mid-term and longer-term strategic objectives (USEPA, 2009). Short-term objectives included preventing the release from harming public health and the environment, removing sufficient CCR to restore flow to the Emory River and reduce downstream migration, and ensuring proper short-term management of CCR. Mid-term objectives included removing all remaining CCR after the short-term objectives were met, restoring waters negatively impacted by the release, and ensuring proper disposal of CCR. Longer-term objectives included a comprehensive site assessment and cleanup to address residual contamination.

Figure 6 shows site features and the areas of time-critical and non-time-critical removals. The time-critical removal actions (Phase 1) were conducted from 2009 to 2010 and consisted of the removal of 3.5 million cubic yards of CCR from the Emory River and east Swan Pond Embayment. The cleanup involved mechanical excavation, hydraulic dredging, and rapid disposal of CCR offsite at the Arrowhead Landfill in Alabama (USEPA, 2017f). Work was performed around the clock, mostly seven days per week, and required extensive processing to dewater the CCR and transport it via railcars to the landfill (Dotson et al., 2013). Non-time critical removal actions (Phase 2) were mostly completed by 2014 and consisted

of the removal of 2.3 million cubic yards of CCR from the north and middle Swan Pond Embayments, which were compacted and disposed of onsite within a 240-acre disposal cell (USEPA, 2017f). The cell was secured within a four-foot wide slurry wall installed to a depth of 50-70 feet below ground surface and was capped with a 40-mil polyethylene liner, a drainage layer, two feet of clay and topsoil, and a vegetative cover (USEPA, 2014c). Phase 3 of the project included an assessment of the 510,000 cubic yards of residual CCR within the Emory River, including human health and ecological risk assessments. In 2012, Monitored Natural Recovery was selected as the remedy to address the residual contamination, and the EPA described the ecosystem as having essentially returned to pre-release conditions by 2014 (USEPA, 2014c). Long-term monitoring began in 2013 and is expected to continue for 30 years.
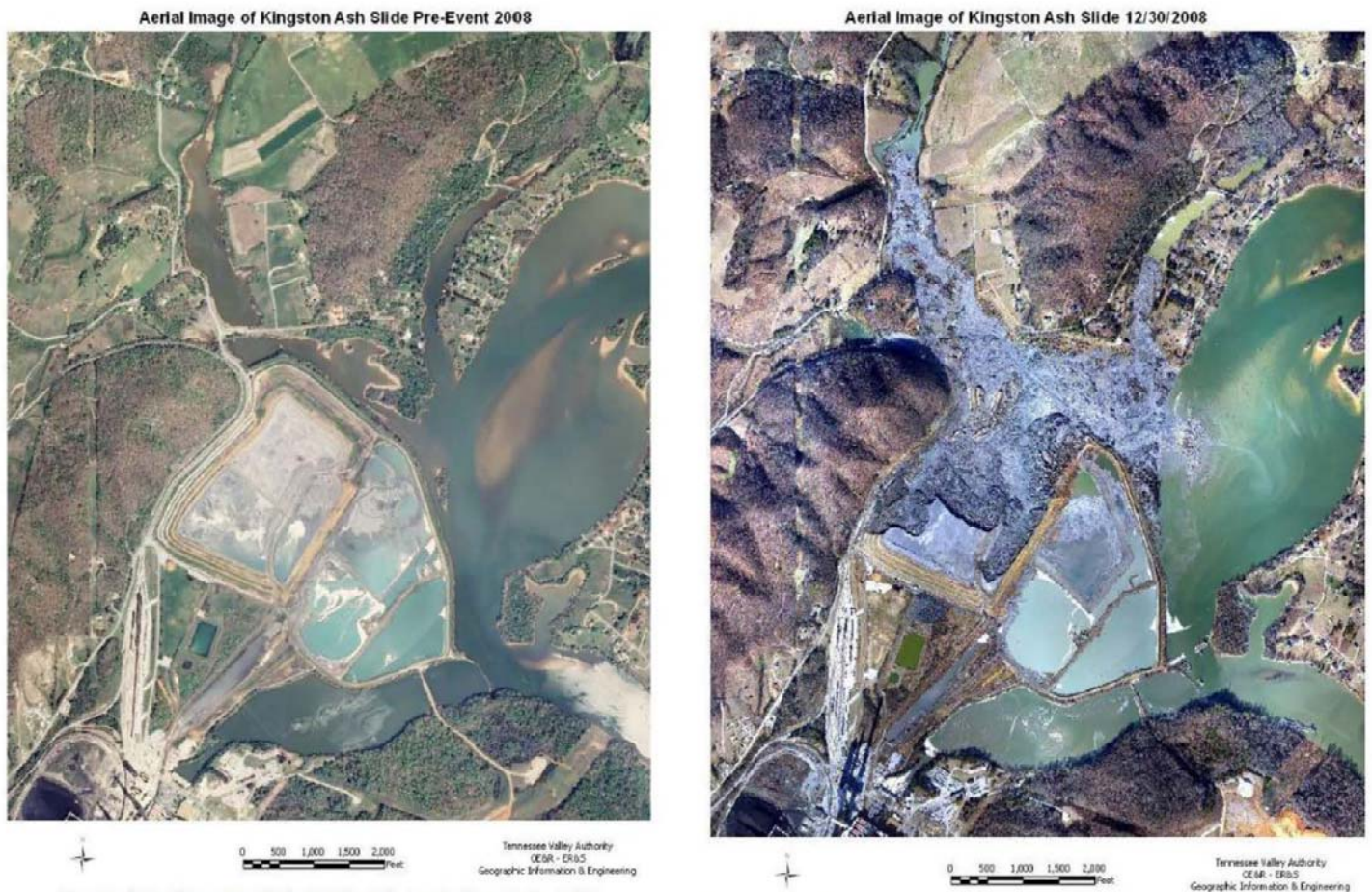
Figure 5 – *Aerial view of TVA Kingston CCR storage unit before and after release on December 22, 2008. Figure from Natural Resource Trustees, 2015.*

Figure 6 – *Overview of TVA Kingston site from USEPA (2017f).*

As and Se were drivers for ecological risk, and were found above reference concentrations in sediment and surface water, representing the nature and extent of CCR-related contamination (TVA, 2012). As a result, As and Se became the focus of the TVA Kingston site.

4.2 As and Se Species

Arsenic can exist in several oxidation states: -III, 0, III, and V. Under acidic and moderately reducing conditions, arsenate (As^V) will coprecipitate with or adsorb onto iron oxides, resulting in arsenic becoming immobile (Wuana & Okieimen, 2011). However, under reducing conditions and at higher pH, arsenite (As^{III}) will be the dominant form and will be more mobile (Wuana & Okieimen, 2011). The primary species of As found in CCR is the oxidized form As^V (Ruhl et al., 2010). The species of As in fly ash samples from the TVA Kingston plant was predominantly As^V (Rivera et al., 2017). The species of As typically leached from CCR is also As^V (Wang et al., 2009).

Selenium can exist in the oxidation states -2, 0, IV and VI. Under aerobic conditions and at high pH, the dominant form will be the more soluble and therefore mobile selenate (Se^{VI}); under reducing conditions and at low pH, the primary form will be selenite (Se^{IV}), which will adsorb onto iron oxides and therefore be less mobile (Brady & Weil, 2008). The primary species of Se in CCR is the reduced form Se^{IV} (Ruhl et al., 2010). Samples of fly ash from the TVA Kingston plant collected in 2009 and 2010, and bottom ash collected in 2009, were analyzed by X-ray absorption near edge structure and contained Se primarily in the elemental Se^0 form, rather than as Se^{IV} (Liu et al., 2013). The amount of leachable Se increased as pH was increased from 3 to 13. When tested at its natural pH of between 7.6 and 9.5, the Se leached from the samples was primarily Se oxyanions as Se^{IV}. Another study of fly ash from TVA Kingston found that Se^{IV} was the primary form in the ash, with lesser concentrations of elemental Se^0 and Se^{II} (in SeS_2) (Rivera et al., 2017). A third study found Se^{IV} to be the primary form in TVA Kingston

ash (ERDC, 2011). Once leached, Se^{IV} is water-soluble and therefore has greater bioavailability than other species (Rivera et al., 2017).

CCR from TVA Kingston therefore likely leached As predominantly in the oxidized form As^V, and leached Se predominantly in the reduced form Se^{IV}. Once ash from TVA Kingston was deposited into the Emory River, As largely remained in the less mobile oxidized form As^V (ERDC, 2011).   30 to 50% of Se in the ash transformed from Se^{IV} to reduced species of Se (including Se^{II}) with no evidence of transformation to Se^{VI} (ERDC, 2011).

The average distributions of the species of As and Se in seasonally exposed sediment, submerged sediment, sediment porewater, mid-depth surface water and epibenthic (from the bottom of the river) surface water are presented in **Figure 7**. This figure shows that organic As, As^{III} and As^V are present in all media, with As^V being dominant in seasonally exposed sediment and As^{III} being dominant in sediment porewater (TVA, 2012). This agrees with the conclusions of ERDC, 2011, that As remained primarily as As^V once deposited in the Emory River. Organic Se^{II} is present in all sampled media and is the primary species of Se in all cases, with Se^{VI} being present in surface water and Se^{IV} present in sediment. This is in agreement with ERDC, 2011 which found that 30 to 50% of Se in the CCR was reduced from Se^{IV} to Se^{II}.
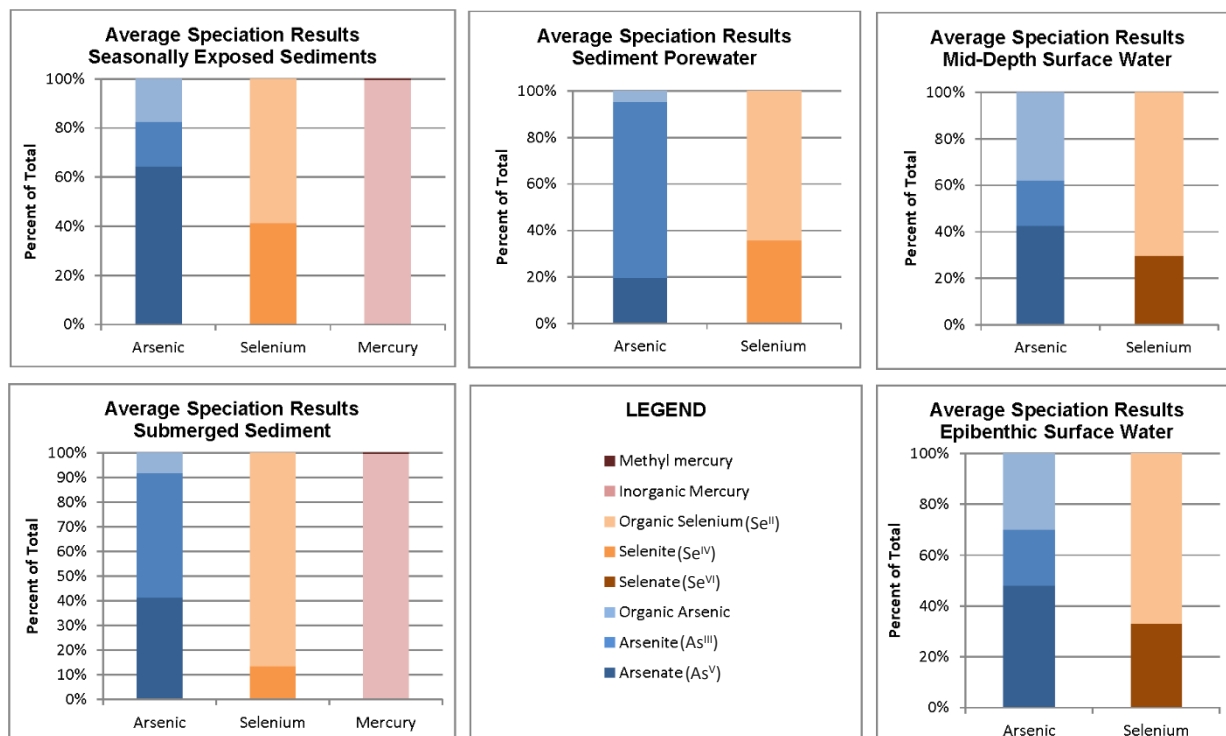
Figure 7 – Average speciation of arsenic and selenium in sediment, porewater and surface water. Figure from TVA, 2012.

4.3 Concentrations, Leaching and Toxicity

A total of 28 samples of the CCR from the disposal cell were collected and analyzed for total element concentrations and radioisotopes. As concentrations in CCR ranged from 28.6 to 166 mg/kg (averaging 74.5 mg/kg), exceeding the regional average surface soil concentration of 19.0 mg/kg, while Se concentrations in CCR ranged from 2.64 to 17.80 mg/kg (averaging 7.89 mg/kg), exceeding the regional average surface soil concentration of 2.44 mg/kg (TVA, 2012).   Therefore total As and Se concentrations within the ash were greater than the concentrations in background soil.

EPA LEAF methods were used on CCR in a leachability study from the site (TVA, 2011). Using Method 1313, initial testing was done at pH values of 5, 7 and 10 (to reflect the pH range of 4.5 to 8.7 observed at the site) at an L/S ratio of 10:1. Based on the results, additional testing was conducted at pH values of 7, 8, 9, 10 and 11. As concentrations increased significantly above pH 9 (ranging from 18.2 ug/L at pH 5 to 702 ug/L at pH 11) while Se concentrations increased significantly above pH 10 (ranging

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from 48.7 ug/L at pH 5 to 134 ug/L at pH 11). Using Method 1316, leaching tests were conducted at L/S

ratios of 0.5, 1, 2, 5 and 10. As concentrations increased from 34.7 ug/L at an L/S ratio of 0.5:1 to 58.2

ug/L at an L/S ratio of 10:1, while Se concentrations decreased from 305 ug/L at an L/S ratio of 0.5:1 to

21.8 ug/L at an L/S ratio of 10:1. These results showed that leachability of As and Se varied with pH and

the L/S ratio, which is in agreement with the discussion in Section 3. They also showed that As and Se

may leach from CCR under the pH observed at the site.

Utilizing the leaching data, groundwater fate and transport modeling was conducted for the

scenario in which CCR would be permanently placed in a landfill in the dredge cell area (the landfill was

later completed in 2014). A Baseline Ecological Risk Assessment evaluated the modelled influx of Se and

As into the system of rivers, over periods of both 30 and 100 years. There were no exceedances of the

Tennessee Water Quality Criteria for Se or As under either modeling condition (TVA, 2012). Therefore,

leaching from the landfill into groundwater and rivers was not a significant exposure pathway.

The distribution of 510,000 cubic yards of residual CCR still present in the Clinch and Emory

Rivers after Phase 1 (time-critical removal) and Phase 2 (non-time-critical removal) is shown in **Figure 8**.

As was detected in seasonally-exposed sediment (containing varying percentages of ash) in the Emory

and Clinch Rivers, averaging 14.3 to 19.2 mg/kg, while Se was detected at average concentrations at

1.77 to 2.45 mg/kg (TVA, 2013). In submerged sediment, average As concentrations ranged from 25

mg/kg in the Emory River to 12.1 mg/kg in the Tennessee River, while average Se concentrations ranged

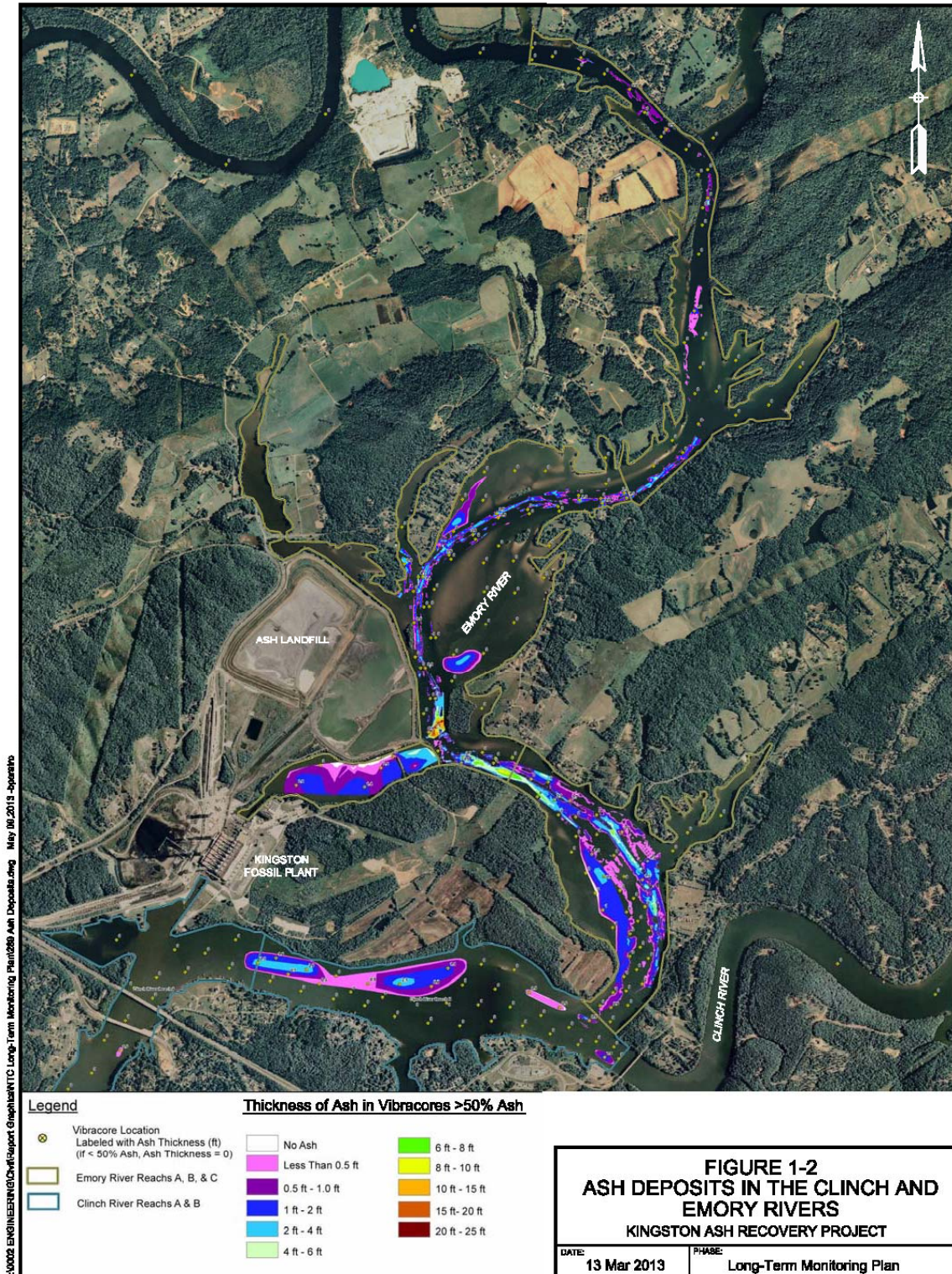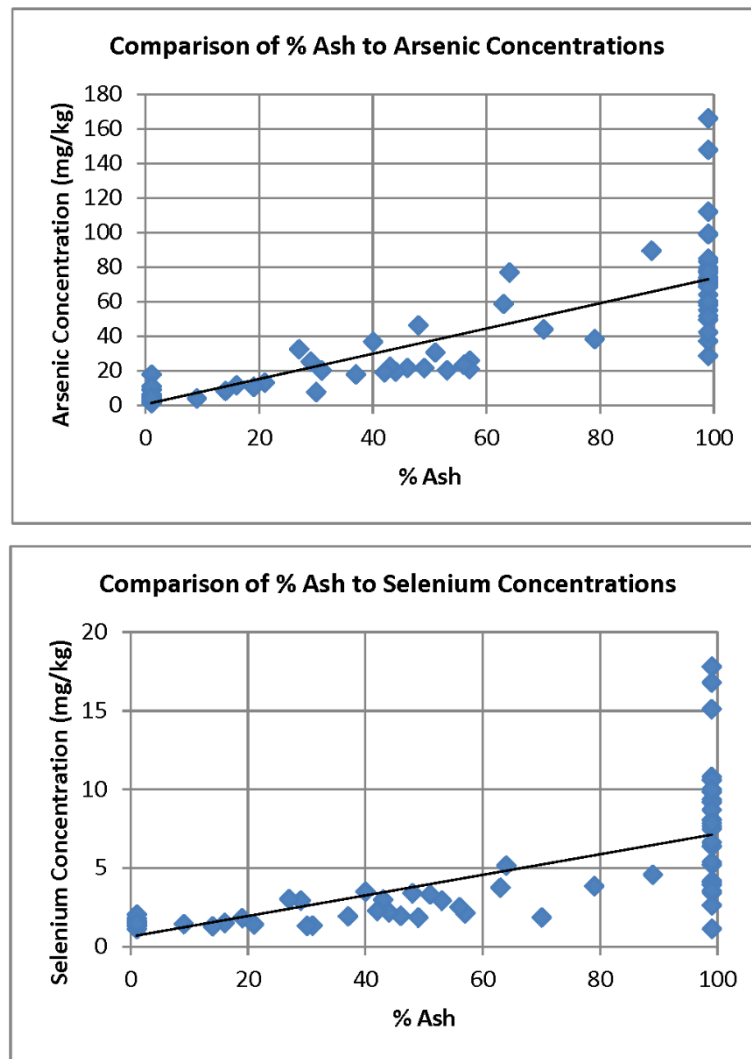from 5.16 mg/kg in the Emory River to non-detect in the Tennessee River.

Figure 8 – *Residual CCR present at the TVA Kingston site after Phase 1 and Phase 2 removals. Figure from TVA, 2013.*

Figure 9 *– Comparison of % ash to arsenic and selenium concentrations, in submerged sediment. Figure from TVA, 2012.*

The concentrations of As and Se in sediment are plotted against the percent ash in **Figure 9**. Concentrations generally increase as ash percentage increases, although there is some variablity due to the expected variation in element concentrations in ash, plus the inprecision of the method used to measure the ash content (polarized light microscopy) (TVA, 2012). The samples shown with 100% ash were from the dredged ash material, with concentrations varying widely as expected due to the inherent variability of trace elements in ash as discussed in Section 3.

A review of toxicity studies from the TVA Kingston river system found that overall risk to species from residual CCR was moderate. No toxicity was observed in aquatic organisms within the water column, however there were risks to benthic fauna in sediment where CCR content is greater than 40%, possibly as a result of exposure to As (Sherrard et al., 2015). Adverse affects on survival due to the physical attributes of the CCR and on growth due to the chemical effects are also likely (Sherrard et al., 2015).

4.4 Summary

Results discussed here indicate that CCR from TVA Kingston contained As predominantly as As^V and Se predominantly as Se^{IV}. Once deposited in the Emory River, As remained primarily as As^V while 30-50% of Se in the ash transformed from Se^{IV} to reduced species of Se (including organic Se^{II}). Total concentrations of As and Se exceeding regional average surface soil concentrations were present in the CCR. Leachability studies demonstrated that As and Se would leach from CCR under the range of pH observed at the site. Utilizing the leaching data, groundwater fate and transport modeling demonstrated that leaching from the landfill into groundwater and rivers was not a significant exposure pathway. However, it was also demonstrated that residual CCR is still present in the environment; As and Se concentrations in sediment increase with increasing CCR percentage; and there is moderate toxicity to benthic fauna in sediment, likely as a result of the As concentrations. In order to evaluate long-term impacts from the onsite landfill and the residual ash, long-term monitoring began in 2013 and is expected to continue for 30 years.

5 Conclusion

The presence of trace elements within CCR that can leach into the environment and the large volume of CCR generated annually in the United States has resulted in the regulation of CCR under RCRA. Given the uncertainty around the environmental impacts of CCR (largely due to the variation in

the elemental composition and leachability of CCR in various environments), it has been regulated as

non-hazardous waste. The TVA Kingston release demonstrates a clear need to regulate the structural

components of CCR units, but environmental impacts from the leaching of trace elements from CCR are

less clear. The environment appears to have recovered even with the presence of 510,000 cubic yards

of residual CCR in the system of rivers, and no additional remediation is planned (just long-term

monitoring). In addition, the 2.3 million cubic yards of CCR compacted and disposed of onsite within a

240 acre landfill have been demonstrated to leach As and Se, yet an ecological risk assessment showed

that leaching from CCR to groundwater was not a significant exposure pathway for ecological receptors.

The EPA has deferred its final determination on whether CCR is hazardous and is requiring

extensive groundwater monitoring (with background assessments) at CCR management units. It may be

the case that based on the results of groundwater monitoring and further risk evaluations, the EPA will

attempt to regulate CCR as hazardous waste in the future.

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