Management of coal combustion wastes

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CCC/231 ISBN 978-92-9029-551-8

January 2014

Abstract

It has been estimated that 780 Mt of coal combustion products (CCPs) were produced worldwide in 2010. Only about 53.5% were utilised, the rest went to storage or disposal sites. Disposal of coal combustion waste (CCW) on-site at a power plant may involve decades-long accumulation of waste, with hundreds of thousands, if not millions, of tonnes of dry ash or wet ash slurry being stored. In December 2008, a coal combustion waste pond in Kingston, Tennessee, USA burst. Over 4 million cubic metres of ash sludge poured out, burying houses and rivers in tonnes of toxic waste. Clean-up is expected to continue into 2014 and will cost $1.2 billion. The incident drew worldwide attention to the risk of CCW disposal. This caused a number of countries to review CCW management methods and regulations. The report begins by outlining the physical and chemical characteristics of the different type of ashes generated in a coal-fired power plant. The amounts of CCPs produced and regulations on CCW management in selected countries have been compiled. The CCW disposal methods are then discussed. Finally, the potential environmental impacts and human health risks of CCW disposal, together with the methods used to prevent them, are reviewed.

Acronyms and abbreviations

ACAA	American Coal Ash Association
ADAA	Ash Development Association of Australia
ASTM	American Society for Testing and Materials
BA	bottom ash
BS	boiler slag
CCB	coal combustion by-product
CCP	coal combustion product
CCR	coal combustion residue
CCW	coal combustion waste
DOE	Department of Energy
EA	Environment Agency (UK)
EC	European Commission
Ecoba	European Coal Combustion Products Association
ECOS	Environmental Council of the States (USA)
EFA	economiser fly ash
EIP	Environmental Integrity Project (USA)
EoW	end of waste
EPA	Environmental Protection Agency (USA)
EPA	Environmental Protection Authority (Australia)
EPA NSW	Environmental Protection Authority, New South Wales (Australia)
EPRI	Electric Power Research Institute (USA)
ESP	electrostatic precipitator
EU	European Union
EWC	European waste catalogue
EWC-STAT	European Waste Classification for Statistics
FA	fly ash
FBC	fluidised bed combustion
FGD	flue gas desulphurisation
LOI	loss on ignition
LoW	List of Wastes (European Union)
MCL	maximum contaminant level
MEP	Ministry of Environmental Protection (China)
Mt	million tonnes
NAAQS	National Ambient Air Quality Standards (USA)
NAS	National Academy of Sciences (USA)
NETL	National Energy Technology Laboratory (USA)
NDRC	National Development and Reform Commission (China)
NODA	Notice of Data Availability (USA)
NRC	National Research Council (USA)
OECD	Organisation for Economic Co-operation and Development
OSM	Office of Surface Mining (USA)
PC	pulverised coal
POEO	Protection of the Environmental Operations (Australia)
RCRA	Resource Conservation and Recovery Act (USA)
SBLP	serial batch leaching procedure
SDA	semi-dry absorption
SGLP	synthetic groundwater leaching procedure
SMCRA	Surface Mining Control and Reclamation Act (USA)
SPLP	Synthetic Precipitation Leaching Procedure
TCLP	Toxicity Characteristic Leaching Procedure

TVA	Tennessee Valley Authority
UN	United Nations
WRAP	Waste Resources Action Programme (UK)
WWCCPN	World Wide Coal Combustion Product Network

Contents

1 Introduction

Burning coal in a coal-fired power plant produces a variety of solid residues known as coal combustion products (CCPs), coal combustion residues (CCRs) or coal combustion wastes (CCWs). CCPs are now more commonly called coal combustion by-products (CCBs) because of their increasing beneficial use. How the material is referred to generally depends on the context in which it is being discussed. Regardless of what it is called, these terms refer to the same material. In order to promote consistent nomenclature, members of the World Wide Coal Combustion Products Network (WWCCPN) called for a harmonisation of the terminology of CCPs (Heidrich and others, 2013).

Coal demand is expected to continue to grow in the emerging economies for the next ten years, driven in particular by the power generation industries in China and India (IEA, 2011). Therefore, the production of CCP will increase inevitably. The world total production of coal combustion products in 2010 was approximately 780 Mt as shown in Figure 1 (Feuerborn, 2012; Heidrich and others, 2013). Table 1 lists the production of total CCPs and the rate of utilisation from 2009 to 2011 (some countries may consider mine and quarry fill as utilisation). It is clearly shown that only around 53% of CCPs are utilised, the rest went to storage or disposal sites. If the estimate for 2010 CCP production was correct, there were approximately 360 Mt of CCPs which needed to be disposed of in 2010 and this figure is expected to continue (if not increase) for many years.

Beneficial use of CCPs is covered in the IEA Clean Coal Centre reports by Adams (2004) and Smith (2005a,b, 2006), ash management in coal-fired power plants by Couch (2006), and impact on ash in respect of quality and quantity of the changes in the power generation sector by Barnes (2010). Therefore this report will focus on CCP disposal, and will use the term CCWs instead of CCPs (where appropriate).

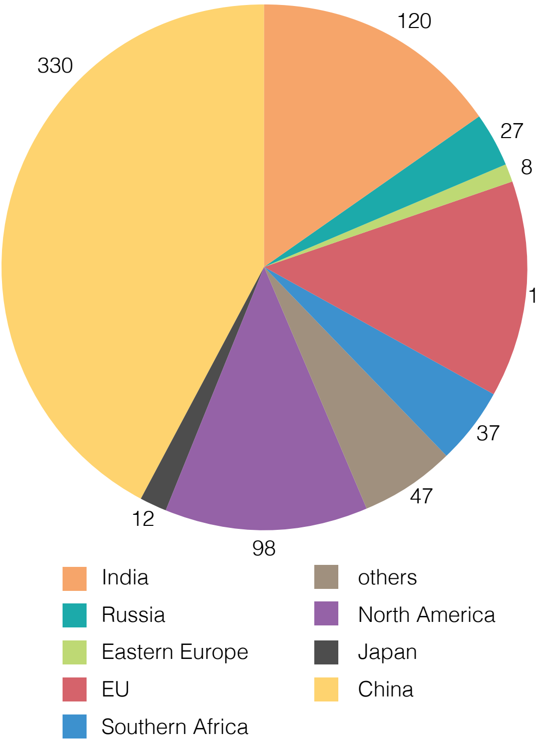

Figure 1 Worldwide production of coal combustion ashes in 2010 (Feuerborn, 2012)

The type of CCP produced in a power plant is determined by the type of coal that is burnt and the design of the boiler. The characteristics of each type of coal ash depend not only on the type of coal and its source and quality, but also on the coal mining and preparation method, combustion process, and the emission control technology used. The environmental and human health impacts from the disposal of CCW are related to its characteristics.

Disposal of CCWs on-site at a power plant may involve decades long accumulation of waste and the deposition of hundreds of thousands, if not millions, of tonnes of dry ash (in a landfill) or wet ash slurry (in a surface impoundment). In December 2008, a coal combustion waste pond in Kingston, Tennessee, USA burst. Over 4 million m^3 of ash sludge poured out, burying houses and rivers in tonnes of toxic waste. Clean-up is expected to continue into 2014 and will cost $1.2 billion (Luther, 2012). The incident drew worldwide attention to the risk of wet CCW disposal. This caused a number of countries to

Table 1 Coal combustion production and utilisation rates for selected countries

	2011		2010		2009		Source
	Production, Mt	Utilisation, %	Production, Mt	Utilisation, %	Production, Mt	Utilisation, %	
Australia	13.1	46	14.1	41	13.7	18	Ash Development Association of Australia (2011)
China	480a	68*	400†	67*	375†	60†	* NDRC (2013), †Yang and others (2010)
Israel	1.09	100	1.07	100	1.13	100	Israeli National Coal Ash Board (2013)
India	131.1	55.8	123.5	62.6	116.7	57.1	Indian Central Electricity Authority (2011)
Japan	9.67	95	10.7	95	10.7	97	Federation of Electric Power Companies of Japan (2012)
Korean R	8.64	67.8	8.41	84.2	8.35	62.8	Wee J (2012)
Poland	20.7	59	19.6	58.3	17.6	60.1	Szczygielski K (2012)
Russia			26.6	18.8	26.1	18.4	Putilov and Putilova (2012)
USA	118.0	43.5	118.1	42.5	114.0	44.3	ACAA (2011)
EU 15¶			48‡	91.4‡	51.8§	90.7§	‡ Caldas-Vieira and Feuerborn (2013), § Ecoba (2013)

¶ The utilisation rate for 2010 includes 39.8% and for 2009 includes 39.2% for the restoration of opencast mines, quarries, and pits

review industry wastes management and regulations.

The US EPA has carried out a large amount of work on CCW disposal, with most of the published work available on the their website (*see* www.epa.gov). Information on CCW management from other countries is not as readily available.

The report begins by outlining the physical and chemical characteristics of the different type of ashes generated in a coal-fired power plant. The amounts of CCPs produced and regulations on CCW management in selected countries have been compiled. The CCW disposal methods are then discussed. Finally, the potential environmental impacts and human health risks of CCW disposal are reviewed, together with the methods used to prevent them.

2 Coal combustion wastes

As the organic compounds of coal combust, ash is generated inside the boiler furnace from the unburnt inorganic components and unburnt carbon. The type of ash produced varies depending on the mineral components of the coal and the combustion technique used. Based on physical and chemical forms derived from coal combustion methods and emission controls, fly ash (FA), bottom ash (BA), boiler slag (BS) and fluidised bed combustion (FBC) ash as well as the products from dry or wet flue gas desulphurisation, such as semi-dry absorption (SDA) product and FGD gypsum, are produced. In America, another type has been classified, known as *economiser fly ash* (EFA). EFA is fly ash that is captured at the economiser unit, located along the flue gas pathway between the furnace and the electrostatic precipitators (ESP) or baghouses. As a result, EFA contains some small, angular, non-volatilised particles entrained in the flue gas stream and some spherules condensed from flue gas at higher temperature than those at the ESPs or baghouses. EFA is also coarser than fly ash collected at ESPs or baghouses (Jones and others, 2012). This report will not classify EFA separately since it is a special form of fly ash. Two more types of ashes are defined in the draft definitions put forward by WWCCPN (Heidrich and others, 2013). Hollow ash particles that form in the furnace gas stream sometimes contains smaller ash spheres. They float on water and are usually collected from the lagoon, where ash/water disposal systems are being used. These ashes are named as *cenospheres*.

Table 2 Types of coal combustion wastes (Couch, 2006; Luther, 2010a)

Waste type	Description
Fly ash	A fine powdered ferroaluminosilicate material captured through a particulate control device in the chimney or stack of plants fired with coal or lignite at 1100–1400°C. Various types of particles are found, including cenospheres (hollow particles), plerospheres, filled with smaller aggregations known as microspheres, and opaque magnetite spheres related to the pyrite content of the original coal.
Bottom ash	A coarse, gritty material, these agglomerated ash particles are those that are too large to be carried in flue gas. The ash impinges on the furnace walls or falls through open grates to an ash hopper below the furnace (fired at 1000–1200°C), either dry or as a slurry (via the addition of water). It has a porous surface structure and is coarse, with grain sizes spanning from fine sand to fine gravel.
Boiler slag	A vitreous grained material drained from the wet bottom boilers operated at 1500–1700°C. When molten slag comes in contact with quenching water, it fractures, crystallises, and forms pellets. This boiler slag material is made up of hard, black, angular particles that have a smooth, glassy appearance. The particles are uniform in size, hard, and durable, with a resistance to surface wear.
Fluidised bed combustion (FBC) ash	Ash which is produced in fluidised bed combustion boilers. The technique combines coal combustion and flue gas desulphurisation in the boiler at temperatures of 800–900°C. FBC ash is rich in lime and sulphur.
Flue gas desulphurisation (FGD) gypsum	A natural gypsum-like product which is obtained by wet flue gas desulphurisation, typically with a limestone slurry sorbent and special treatment of the adsorbed products.
Spray dry absorption (SDA) FGD products	A fine grained material resulting from dry flue gas desulphurisation with lime acting as the sorbent.
FGD other products (including other FGD scrubbers)	Products from other FGD scrubbing processes not covered in the above two categrories.

Another type is *Conditioned ash*, which refers to the fly ash which is mixed with water (10–20% by dry mass typically) in order to be transported in normal tipping vehicles. The different types of coal combustion product are described in Table 2.

Fly ash, bottom ash, boiler slag, and FGD residues are referred to as high-volume wastes by the American industry because they are generated in very large quantities (EPA, 1988). Coal-fired power plants also generate waste streams that the industry typically calls low-volume wastes, which are formed during equipment maintenance and water purification processes. However, some of these wastes (such as cooling tower blowdown) can be generated in substantial quantities. Although these wastes are usually small in volume compared to ash and environmental control residues, they can contain higher concentrations of certain constituents that may cause environmental concern. They therefore require care in handling and disposal. Types of low-volume wastes generated by coal-fired power plants, which are not considered in this report, include:

- boiler blowdown;
- coal pile runoff;
- cooling tower blowdown;
- demineraliser regenerants and rinses;
- metal and boiler cleaning wastes;
- pyrites;
- sump effluents.

The physical and chemical characteristics of each type of CCW have a bearing on its potential for utilisation and potential risk to human health or the environment when disposed of. Physical, chemical, mineralogical, morphological and radioactive properties of CCWs vary as they are influenced by coal source and quality, combustion process, the presence of cofired materials, degree of weathering, particle size and age of the ash, and so on. The American Society for Testing and Materials (ASTM) recognises two classes (C and F) of coal combustion products based on the relative concentration of silica, aluminum, and iron oxide that correspond to a high-lime and low-lime content fly ash. Class F ash is pozzolanic in nature, and contains more than 70 wt% $SiO_2 + Al_2O_3 + Fe_2O_3$ and low lime. This fly ash is produced from the combustion of harder, older anthracite and bituminous coal. Class C ash from burning younger lignite or subbituminous coal, in addition to having pozzolanic properties, also has some self-cementing properties. Class C fly ash generally contains 50–70 wt% $SiO_2 + Al_2O_3 + Fe_2O_3$ and high lime (Ahmaruzzaman, 2010). Alkali and sulphate (SO_4^{2-}) contents are generally higher in Class C type fly ash (Seshadri and others, 2013). In Australia, the majority of CCPs produced is categorised as Class F – being mainly silica and alumina (80–85%) and <10% calcium oxide (CaO). CCPs produced in India are mainly the Class C type.

In this section, CCWs are separated into two groups to discuss their characteristics: ashes and emissions control residues.

2.1 Characteristics of ashes

Ashes here refer to those generated from conventional combustion boilers without FGD treatment and include fly ash, bottom ash and boiler slags.

2.1.1 Physical characteristics

Physical characteristics of coal combustion ash of interest for disposal are particle size and its distribution, compaction behaviour, permeability, and shear strength (*see* Table 3).

The greater the assortment of particle sizes in the material, the more it can be compacted to achieve greater density and shear strength, and lower permeability. Fly ash particles are fine-grained, typically

Table 3 Physical characteristics of fly ash, bottom ash and boiler slag (EPA, 1988; EPRI, 2009)

Properties	Fly ash	Bottom ash/Boiler slag
Particle size, μm	1–100	100–0,000
Compaction behaviour		
compressibility, %	1.8	1.4
bulk density, kg/m^3	1041	1762
moisture content, %	10–35	12–26
porosity, %	0.4–0.5	0.25–0.4
Permeability, cm/s	10^{-6}–10^{-4}	10^{-3}–10^{-1}
Shear strength		
cohesion, Pa	0–1.17x10^6	0
angle of internal friction, °	25–45	35–45

silt-sized, ranging from 1 to 100 μm in diameter with a median particle diameter of 20 to 25 μm (EPRI, 2009). The particle size distribution of most bituminous coal fly ash is generally less than 75 μm. Although subbituminous coal fly ash is also silt-sized, it is generally slightly coarser than bituminous coal fly ash (Ahmaruzzaman, 2010). Bottom ash and boiler slag are coarser than fly ash and the size of the particles can range from fine sand to fine gravel, or about 100 to 10,000 μm in diameter (EPA, 1988).

Compaction behaviour refers to the amount of settling that takes place after disposal and the rate at which such settling occurs. Compressibility, density, and moisture content are factors affecting compaction behaviour. When compacted and dry, most fly ash and bottom ash behave very similarly to cohesive soil (EPA, 1988).

Permeability reflects the rate at which water will seep through the waste material in a given period of time. It provides a good first estimate of the rate and quantity of leachate migration. A number of factors can influence the degree of permeability, such as the size and shape of particles, the degree of compaction, and the viscosity of water. Properly compacted fly ash often has a low permeability, similar to that of clay, while the permeability of bottom ash is usually slightly higher. Boiler slag permeability is higher, similar to that of fine gravel (EPA, 1988).

Shear strength is an important determinant of the shape and structural stability of wastes disposed of in landfills. A strong material can form steep slopes and support heavy loads from above. Two indicators of shear strength are cohesion, a measure of the attraction between particles due to electrostatic forces, and the angle of internal friction, an indicator of the friction between particles. Dry, non-alkaline ash has no cohesion. Dry ash that is alkaline demonstrates some cohesion and, when compacted, increases in strength over time. The angle of internal friction associated with ash varies with the degree of compaction, although it is similar to that for clean, graded sand (EPA, 1988).

2.1.2 Chemical characteristics

The waste itself represents non-combustible constituents in coal. Therefore, its characteristics are strongly influenced by the source coal itself, such as lignite, bituminous coal, or subbituminous coal, and the techniques used for handling and storage. Combustion processes and the operating conditions of the boiler also have an effect. Factors affecting ash chemical characteristics are very plant and coal specific.

While the components of each type of ash vary, depending on the above factors, all CCWs are likely to include certain amounts of toxic constituents. Depending on the sulphur content of the feed coal, the pH of fresh FA and BA generally ranges between 4.5 and 12 (Dellantonio and others, 2010). In general, over 90% of ash is made up of silicon, aluminium, iron, and calcium in their oxide form. Minor constituents such as magnesium, potassium, sodium, titanium, and sulphur account for about 8% of the mineral component, while trace constituents such as arsenic, cadmium, lead, mercury, and selenium, together make up less than 1% of the total composition (*see* Table 4). The partitioning of

Table 4 Element concentrations (mg/kg) in fly ash and bottom ash (EPRI, 2009)

	Fly Ash*	Bottom Ash*
Aluminum	70,000–140,000	59,000–130,000
Calcium	7,400–150,000	5,700–150,000
Iron	34,000–130,000	40,000–160,000
Silicon	160,000–270,000	160,000–280,000
Magnesium	3,900–23,000	3,400–17,000
Potassium	6,200–21,000	4,600–18,000
Sodium	1,700–17,000	1,600–11,000
Sulphur	1,900–34,000	BDL–15,000
Titanium	4,300–9,000	4,100–7,200
Antimony	BDL–16	All BDL
Arsenic	22–260	2.6–21
Barium	380–5100	380–3600
Beryllium	2.2 - 26	0.21–14
Boron	120–1000	BDL–335
Cadmium	BDL–3.7	All BDL
Chromium	27–300	51–1100
Copper	62–220	39–120
Lead	21–230	8.1–53
Manganese	91–700	85–890
Mercury	0.01–0.51	BDL–0.07
Molybdenum	9.0–60	3.8–27
Nickel	47–230	39–440
Selenium	1.8–18	BDL–4.2
Strontium	270–3100	270–2000
Thallium	BDL–45	All BDL
Uranium	BDL–19	BDL–16
Vanadium	BDL–360	BDL–250
Zinc	63–680	16–370

BDL Below Detection Limit

* Source for most fly ash and bottom ash data is EPRI CP-INFO Database. Beryllium, thallium, mercury (bottom ash only) and boron (bottom ash only) are from the EPRI PISCES Database

these elements is primarily determined by volatilisation and condensation processes involved in the combustion of coal. Trace toxic elements are leachable and the rate of constituent release to the environment is affected by leaching conditions (Kosson and others, 2009). Radionuclides of uranium (U) and thorium (Th) series were reported in FA (Tadmore, 1986). The majority of FA is not significantly enriched in radioactive elements or in associated radioactivity compared to common soils or rocks (Zielinski and Finkelman, 1997). Unburnt carbon, depending on the combustion conditions, can also appear in CCWs. The wastes are also likely to include a certain level of toxic organic compounds, such as dioxins and polycyclic aromatic hydrocarbons (PAH) (EPA, 1988; EPRI, 2009; Luther, 2010a).

The principal components of bituminous coal fly ash are silica, alumina, iron oxides, and calcium oxides, with varying amounts of carbon, as measured by the loss on ignition (LOI). Bituminous coal ashes generally contain much less calcium than subbituminous coals, and yield slightly acidic to slightly alkaline solutions (pH 5–10) on contact with water. Subbituminous coal and lignite fly ash typically contain relatively high concentrations of calcium, often exceeding 15% (expressed as CaO), and produce alkaline solutions (pH 11–12) on contact with water (*see* Table 5). Very little anthracite is burnt in utility boilers, so there are only small amounts of anthracite fly ash (Ahmaruzzaman, 2010; EPA, 1988; EPRI, 2009). Ahmaruzzaman (2010) compared the normal range of the chemical constituents of Indian bituminous coal fly ash with those of Indian lignite fly ash and subbituminous coal fly ash. Indian lignite and subbituminous coal fly ash typically has a higher calcium oxide content and LOI, and may have a higher concentration of sulphate compounds than fly ash from bituminous coals.

The distribution of elements among the different types of ash is largely determined by the firing temperature of the boiler, which in turn affects the proportions of volatile elements that end up in the fly ash and bottom ash. Some elements, such as sulphur, mercury and chlorine, are almost completely volatilised and leave the boiler in the flue gas rather than remaining in the bottom ash or boiler slag. Some of these more volatile elements may condense on the surface of the fly ash particles as the flue gas cools. For example, much higher quantities of arsenic, copper and selenium are found in fly ash

Table 5 Typical composition (wt%) of FGD residues and fly ashes in the USA (Berland and others, 2003)

	FGD gypsum	Sulphitic wet FGD material	Spray dry scrubber product	Bituminous coal fly ash	Subbituminous coal fly ash	Lignite fly ash
SiO_2	0.1–6.3	0.1–7.4	7.9–10.6	43–64	30–45	12–49
Al_2O_3	0.1–5.1	0.1–6.0	4.1–4.7	11–32	17–21	6.3–16
Fe_2O_3	0.1–5.7	0.1–6.0	1.9–4.4	3.5–25	3.9–6.5	3.7–14
CaO	27–32	38–52	1.0–1.9	0.7–20	17–30	16–34
MgO	1.0–4.9	1.3–6.1	33.6–37.8	0.6–1.6	3.4–6.4	3.8–11
Na_2O	0–0.6	0.1–0.9	0.2–0.3	0.2–4.0	0.2–8.4	0.6–6.1
K_2O	0–0.6	0–0.6	0.3–0.6	0.5–3.0	0.3–1.0	0.4–2.1
CO_2			10.6–20.1			
SO_3	44–46	54–63	17.4–30	0.1–11	1.1–5.0	0.4–30
SO_4			10.5–24.6	1.2		
Cl^-			0.9–3.0	<0.1		
C (organic)			0.1–0.2	1.0		
Solids	*5–10*	*5–10*				

than are found in bottom ash or boiler slag (EPA, 1988). Bhangare and others (2011) studied ashes from five Indian power plants and found that the concentration of 13 elements (Pb, Cu, Cd, Zn, Fe, Mn, Cr, Ni, Mg, Li, Co, Hg, and As) are higher in fly ash than that in bottom ash and coal. The results showed that the elements were more enriched in fly ash than in bottom ash. The elements were classified into three groups based on their volatility:

- elements which are condensed equally between bottom and fly ashes but not in the fugitive fly ash;
- elements which are more concentrated in the fly ash collected in electrostatic precipitators and the fugitive fly ash than in the bottom ash;
- elements which are mainly present in gas phase and in minor concentrations in fly and bottom ashes.

Elements can volatilise during coal combustion and the volatility is dependent on the mode of occurrence, concentration, physical change, chemical reactions, and combustion technology. The elements associated with sulphide minerals and organic matter are more volatile compared to those in other chemical forms, such as oxides.

Changing the fuel by introducing a different coal blend or cofiring biomass with the coal, for example, can alter the chemical composition of the resultant ashes. Biomass ash does not contain toxic metals like coal ash. The ash-forming constituents in biomass fuels are quite diverse depending on factors such as the type of biomass, type of soil and harvesting methods. In general, the major ash-forming inorganic elements in biomass fuels are Ca, K, Na, Si and P. However, some biomass fuels have a high silicon content, for example rice husk, while some have a high alkali metal content, such as wood (Ahmaruzzaman, 2010). On the other hand, KEMA conducted a series of tests to determine the elemental composition and parameters, such as the relative enrichment factors of the elements in the ashes and the removal efficiencies of the gaseous elements in the ESP and FGD materials. It appeared that those parameters obtained for biomass co-combustion in general are equal to those at 100% coal firing (te Winkel, 2010). Barnes (2010) also reviewed the impacts on fly ash properties of co-combustion of other fuels with coal. The conclusion is that the composition of the cofiring fly ash tends to be dominated by the composition of the coal, as most co-fuels have a relatively low ash content. However, in some cases, a co-fuel may be rich in a specific component, for example vanadium and nickel in petcoke, and care must be taken to ensure that these do not adversely affect the environment.

Upon combustion, radio nuclides naturally found in the feed coal are preferentially accumulated in the finer FA fractions. Generally, radio nuclide activities in CCWs are in the upper range of natural background concentrations for granites and shales; however, there is large regional variation. Radioactivity of CCWs are discussed in more detail in Chapter 5.

2.2 Characteristics of pollutant control residues

The current trend towards zero emissions presents a challenge to power station operators to minimise the effects of coal combustion in terms of limiting atmospheric pollution and reducing the amount of residues generated. Various air pollution control systems have been developed to reduce sulphur oxides, nitrogen oxides, carbon oxides and other greenhouse gas emissions. These systems have an impact on some of characteristics of CCWs.

The characteristics of emission control residues differ both physically and chemically from conventional FA, mainly because of the reagents used to control pollutants and modifications made to the combustion technology. Emission control processes that can affect fly ash composition include the use of ammonia-based systems to control NOx, powdered activated carbon injection to control mercury and sodium-based sorbents to control SO_3 (EPRI, 2009). In terms of carbon capture, there are three techniques applied to coal-fired power plants:

- pre-combustion capture which requires the use of integrated gasification combined cycle (IGCC);
- post-combustion capture which is similar to conventional pollutant control systems;
- oxy-combustion which involves burning coal in an oxygen rich environment to produce a concentrated steam of CO_2.

These techniques are described in detail by Davidson (2007, 2009, 2010, 2011, 2012). Ashes from IGCC are not covered in this report. Post-combustion processes will not have any impact on the resulting CCWs as there are no changes to the coal combustion and desulphurisation processes. However, the quality of CCWs is affected in the oxy-fuel process due to its high combustion temperature (Caldas-Vieira and Feuerborn, 2012, 2013). Barnes (2010) believes oxyfuel combustion presents a radically change to the combustion environment. Although his research did not quantify the impact of the oxyfuel environment on ash, it is still a very important area for ash development. Fryda and others (2012) studied ash deposition of Russian coal and lignite under oxyfuel and air combustion conditions. Results show higher deposition propensities under oxyfuel conditions. Particle size seems larger for the Russian coal oxy fired ash but not for lignite. The density and porosity may also be affected but the authors did not investigate further. Fryda and others (2012) also reported that chemical and mineralogical compositions do not seem to differ significantly between air and oxyfuel conditions. However, Wu and others (2013) studied ash deposition behaviours in a 3 MWe pilot plant under both oxyfuel and air combustion conditions and reported a higher content of SO_3 and Fe_2O_3 and lower content of SiO_2 and Al_2O_3 in oxyfuel fly ash.

Combustion modifications to reduce NOx, such as low NOx burners, can result in high carbon in ash because of a lower maximum flame temperature in the boiler. This is particularly common for older units that have been retrofitted. Newer boilers are less prone to give high carbon in ash, but the longer residence times and cooler furnace affect the ash mineralogy which in turn affects its performance negatively in, for example, construction products. Post-combustion processes to reduce NOx include selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR), both of which produce ammonia-rich ash. These carbon- and ammonia-rich ashes can be processed for use in plastic filler applications (Huang and others, 2003). However the ammonia ‘slip’ needs to be managed very carefully in order to avoid nuisance to end users.

SO_2 can be removed in fluidised bed combustion (FBC) by the addition of crushed limestone to the bed material. The bed is held in suspension by injecting air through a perforated floor. The limestone reacts with the released SO_2 to form calcium sulphate ($CaSO_4$). FBC ash (both fly ash and bottom ash) is produced inside the boiler. FBC FA is carried in the flue gas and leaves the furnace in the upper part of the boiler. It is mostly collected in electrostatic precipitators or fabric filters. FBC BA is collected at the bottom of the boiler and represents the coarser part of the ash. FBC fly ash and bottom ash may be mixed and extracted together as FBC ash. FBC ash is a mixture of conventional coal combustion ash (fly ash and bottom ash), the SO_2 reaction product (primarily $CaSO_4$) and unspent sorbent. Hence, FBC ash is highly alkaline which affects the reactivity of the ash and influences its suitability for certain applications. It contains higher levels of calcium, sulphate and carbon than conventional ash and lower amounts of silica, alumina and iron oxide. The FBC ash does not meet ASTM requirements to qualify as either class C or class F fly ashes (Shon and others, 2011). This is not only due to the chemical requirement but also due to the exclusions of materials from FBC with lime (Feuerborn, 2013). The coal is combusted at temperatures of approximately 800–950°C, a range which is lower than in the traditional pulverised coal units (1300–1700ºC). The relatively low combustion temperature not only supports sulphur capture and reduces NOx formation considerably but also permits the use of various types of fuel. This has increased the popularity of the technology as there is a growing demand to combust or co-combust more secondary fuels, such as petcoke, sewage sludge and high-energy industrial wastes. FBC produces large volumes of ash because of the amount of sorbent used; approximately 1000 kg of coal burnt can generate 200 kg of FBC ash (Wang and others, 2006). Limestone used as a sorbent for the SO_2 released during coal combustion is effective in capturing SO_2, but the resulting ashes require handling, disposal and utilisation procedures radically different to those for PC ash (Anthony and others, 2003).

FGD is the process which is added to a pulverised coal fired boiler and other boilers to extract SO_2 from the flue gas. The reagents that are used as sorbents to scrub flue gases include limestone ($CaCO_3$), lime (CaO) and caustic soda (NaOH). There are many different FGD processes which include wet scrubbers, semi-dry or spray dry absorption systems, and dry sorbent injection processes, all of which generate residues. A wet scrubber process produces slurry waste, which is a saleable by-product (provided there is a local market). The semi-dry and dry FGD processes produce solid waste, which is easily disposed of compared to ash from wet scrubbers (Seshadri and others, 2013; Smith, 2006).

All FGD residues are comprised of spent reagent, which is made up of the chemical compounds that result from the reaction of the absorbent with sulphur oxides in the flue gas, plus any unreacted portion of the absorbent. The sludge may also contain water and fly ash. Several factors determine how much spent reagent, water and ash are present in the FGD sludge. These factors include the type of scrubber system used, combustion conditions, as well as other pollution abatement and control technologies, the characteristics of the coal, and the SO_2 emission limit that the power plant is required to meet by law (EPA, 1988).

Hence, the main pollutant control residues for disposal are fluidised bed combustion (FBC) ash and flue gas desulphurisation (FGD) residues or sludge.

2.2.1 Physical characteristics

In general, the same physical properties important in determining the disposal of ash from pulverised coal fired boilers (*see* Section 2.1.1) are also important determinants of the disposal characteristics of pollutant control residues. These physical characteristics, such as particle size, compaction behaviour, permeability and shear strength, vary considerably depending on the scrubber system employed, fuel type and source, combustion conditions, and so on.

FBC ashes

As FBC ashes are conventional ashes with additional SO_2 reaction products and unspent sorbent and unburnt carbon, their physical characteristics are similar to pulverised coal (PC) ashes with some slight modifications. Anthony and others (2003) summarised four differences of FBC ashes compared to conventional PC ashes:
- they react vigorously with water (CaO is present) and two of their components are relatively water soluble, namely $CaSO_4$ and CaO, after conversion to $Ca(OH)_2$;
- they are fine grained, non-plastic and self-cementing;
- they contain few, if any, spherical particles;
- dimensional and freeze-thaw stability in the finished products are often poor.

FBC ashes appear dark grey in colour which may be attributed to the removal of NOx and SOx (Seshadri and others, 2013). The particle size of FBC ashes is larger than that of ashes generated from conventional PC boilers because coal is not pulverised prior to combustion in FBC systems. According to Electricité de France (EDF) investigations, FBC fly ash extracted from the ESP is a silt-sized material with a particle size range of 0.5 to 100 µm, with a median diameter of 21 ± 3 µm, depending on cyclone design. FBC bottom ash is a rough, sand-like material. Its particle size ranges from 0.1 to 20 mm with a median diameter of 0.25 ± 0.1mm (Lecuyer and others, 2001). Chugh and others (2000) reported similar FBC fly ash particle sizes (20–30 µm) and these had a prismatic shape. The uniformity coefficient is similar to Class C and F fly ash. However, the FBC bottom ash was much coarser in size (70–80 µm), with a lower uniformity coefficient than FBC fly ash. The LOI of FBC ash is higher than PC ash, and it also has a much smaller amount of glassy material because of the lower temperature in the combustor.

The size, density and strength of FBC ash have also been described by Galpern and others (1997):

- 6–13% of particles are larger than 10 mm, while 8–20.5% of the particles are small (<0.63 mm);
- the bulk density of the FBC bottom ash is 1130–1310 kg/m^3;
- the compression durability is 1.77–3.53 MPa;
- the crushability is 24.8–31.2%.

FGD residues

Depending on the type of FGD system employed, the particle size distribution of FGD residues can vary substantially from plant to plant. Typical particle size is between 0.88 and 1 mm. Sludge from wet scrubbers tends to have a narrow range of particle sizes. The particles produced by dual-alkali systems are finer than those generated in direct lime or limestone scrubbers, which are already very fine: 80 wt% are finer than 0.025 mm effective diameter. The dry scrubbers generally produce sludge containing larger particles. For example, Naik and others (2003) cite typical particle size distributions for calcium sulphite (unoxidised) and calcium sulphate (oxidised) residues, graded from coarser sand, silt, to fine clay, as:

	Unoxidised, %	Oxidised, %
Sand size	1.3	16.5
Silt	90.2	81.3
Clay	8.5	2.2

The density of FGD sludge depends directly on the method of handling. Wet sludge mixed with ash will have a higher density than untreated sludge, while chemical fixation increases the density even more. The average bulk density of FGD gypsum depends on the particle shape, and is approximately 1200 kg/m^3 for block shaped crystals and roughly 600 kg/m^3 for needle-like crystals. The density of the particles in dry sludge varies widely. The bulk density of spray dry absorption residues generally varies from 780 to 1250 kg/m^3 compared to 1000 kg/m^3 for fly ash. When there is only a small proportion of ash, the bulk density is about 600 kg/m^3. The density of the furnace sorbent injection particles was 2650 kg/m^3, greater than that of particles from duct sorbent injection at 2410 kg/m^3.

The permeabilities of untreated FGD sludge from the different wet scrubber systems are generally similar. Mixing the sludge with fly ash does not necessarily change the degree of permeability, although if fly ash acts as a fixative when added to the sludge, the mixed waste product will have reduced permeability. Chemical fixation can also decrease permeability. Sludge from dry scrubber systems has low permeability relative to sludge from wet systems.

The solids content of scrubber sludge is a function of many things, including whether the sludge is treated prior to disposal, the size of the particles in the sludge, the sulphur content of the coal, the amount of ash present in the sludge, and the desulphurisation process used. The percentage of solids in untreated sludge is usually in the range 20–40%, although it can be as high as 60%. Depending on the method of treatment used before disposal (if any), the percentage of solids can be much higher. In fact, some chemical fixation processes are designed to transform the sludge into a cement-like product.

The shear strength of FGD sludge is referred to as ‘unconfined compressive strength,’ which reflects the load-bearing capacity of the sludge. The unconfined compressive strength is sensitive to the moisture content and age of the sludge. Untreated wet sludge has no compressive strength and is similar to toothpaste in this respect. Mixing with ash or lime increases compressive strength, as does chemical fixation. In addition, as the treated sludge ages, its compressive strength increases. Compared to raw coal with 20% moisture content or wet sand with 10% moisture, gypsum with 9% moisture has a much greater unconfined yield strength of around 10 kPa at a 10 kPa normal stress. After seven days of storage, it becomes even more cohesive, giving an unconfined yield strength of around 20 kPa which means it is essentially non-flowing. For spray dry absorption residues, the typical moisture content of the conditioned residues is about 10%. Conditioning with water aids compaction and the residues may then display thixotropic properties.

The information in this section is taken mainly from EPA (1988) and Smith (2006), with additional information incorporated, where appropriated.

2.2.2 Chemical characteristics

FBC ashes

With added limestone, the FBC process achieves SO_2 removal by the following steps:

$CaCO_3 \rightarrow CaO + CO_2$

$CaO + SO_2 + ½O_2 \rightarrow CaSO_4$

As Adams (2004) summarised, the limestone particles, when heated, calcine from a low porosity solid to a porous matrix of CaO, with about 50% porosity. The CaO then reacts with sulphur dioxide. However, the $CaSO_4$ product blocks the pores of the calcined sorbent. Overall, utilisation is typically 20–40%, leaving a solid which contains significant amounts of CaO, in addition to $CaSO_4$, and fuel derived ash components. Thus, the main components of the solid residues from FBC, using limestone as a sorbent, are those arising from the sulphation reaction ($CaSO_4$) and the by-products of this reaction (CaO, $CaCO_3$).

The chemical composition of the ash depends directly on coal characteristics, namely(Lecuyer and others, 2001):

- the chemical composition of the inorganic fraction of coal;
- the sulphur content of coal and the SO_2 removal rate .

The variability of the chemical composition is high, even for the same firing unit, according to the various fuels used. This variability can hinder ash management, because a constant quality of ash cannot be guaranteed. Depending on the coal used, the main features (or constraints) of the chemical composition are usually the high calcium and high sulphate contents resulting in a high alkalinity (pH 12–12.7). However, lower contents of Si, Al, Ba, Cr, Cu and Mn compared to PC ashes are found. This may be due to the lower temperature used in FBC resulting in less volatilisation of the components of the coal mineral matter. One notable exception is ash from the firing of low sulphur anthracite and bituminous coals; these FBC residues are composed primarily of fuel ash constituents, since sorbent does not dominate ash chemistry (Lecuyer and others, 2001; Sellakumar and others, 1999).

FGD residues

The characteristics of FGD residues are detailed in Smith (2006). The following summary is based on the reviews by Smith (2006) and Twardowska and Szczepanska (2004).

The chemical composition of FGD products varies from plant to plant depending on the operating conditions, the coal fired and desulphurisation regents employed. Generally, as Ca and S make up about 90% of the FGD residues, the mineralogy is dominated by $CaSO_3$ or $CaSO_4$, as well as unspent sorbents such as $Ca(OH)_2$ and CaO, rendering such residues highly alkaline. The limestone scrubbing process is generally represented by the following overall reactions:

$SO_2 + CaCO_3 \rightarrow CaSO_3 + CO_2$

$CaSO_3 + 1/2O_2 + 2H_2O \rightarrow CaSO_4{\cdot}2H_2O$

The chemical properties of FGD residues are summarised in Table 5 using data collected from FGD systems operating in the USA in the 1990s.

The most common wet FGD processes remove SO_2 in a water suspension of ground limestone in wet

FGD scrubbers and semi-dry adsorption towers. The end product is calcium sulphite ($CaSO_3$) and sulphite crystals in the form of a water suspension separate from FA. Generally calcium sulphite is converted to calcium sulphate ($CaSO_4$) by forced oxidation in wet limestone FGD processes. The wet FGD processes do not influence the properties of the other CCWs (FA, BA, BS) and the environmental behaviour of FA when the wet flue gas desulphurisation is processed does not differ from that of the 'pure' FA without FGD process. But the wet FGD processes create a considerable amount of FGD solid residues, which increase the burden for disposal.

The chemical composition of spray dry scrubber residues depends on the sorbent (commonly lime) used for desulphurisation and the proportion of fly ash collected with the FGD residues, generally consisting of fly ash (70–80%) entrained with reacted and unreacted sorbent. The principal reaction product of spray dry scrubbers is calcium sulphite as well as minor amounts of $CaCO_3$ and $CaCl_2$. Gypsum may also form under more oxidising conditions. Unused sorbent remains as CaO in the residues and is the most variable component. Due to the presence of unreacted CaO, the end product is reactive and highly alkaline. The high proportion of FA moderates reactivity of the FGD reaction products. The chemical composition of spray dry scrubber residues show significant increase in S, Ca and carbonates compared to the 'pure' FA. Residues from duct sorbent injection are similar to those from the spray dry absorption process, consisting primarily of $Ca(OH)_2$ and $CaSO_4.1/2H_2O$.

The end product of the semi-dry FGD processes consists of dry FA and the reaction products of the injected sorbents and SO_2 in the flue gases. The reaction products from semi-dry FGD processes consist of calcium compounds: $CaSO_3$, $CaSO_4$, $Ca(OH)_2$, $CaCO_3$ and smaller amount of $CaCl_2$. The composition of the end product from the semi-dry desulphurisation processes, in particular the proportion of calcium compounds, depends upon the proportion of FA, which differs significantly for different systems and varies from 2% to 75%. Accordingly, the end products can thus be either high- or low-FA material. The content of $CaSO_3$, which is a main component of the end product, generally ranges from 10% to 60%. $CaSO_4$ occurs in smaller amounts and $Ca(OH)_2$ prevails over $CaCO_3$. The proportion of FA and FGD reaction products determines the chemical composition of the end product of a semi-dry FGD process.

The general pattern of trace element occurrence in the FA matrix remained similar over the years studied by Twardowska and Szczepanska (2004), despite many different and variable sources of coal supply to the power plants studied. Dry FGD residues experienced decreased metal concentrations because of the addition of low-metal FGD product. Decrease in metal concentrations of 50–70% have been found in semi-dry FGD residues. This can be explained by the 'diluting' effect of trace element-free reaction products. Therefore, FA is the main source of trace elements in the FGD residues. To conclude, the product from dry and semi-dry FGD processes may have a considerable effect on the properties of FGD residues and release of constituents from this material due to its influence on pH, the contents and forms of sulphur and calcium compounds, as well as on concentrations of trace elements. With respect to trace element concentration, FGD products caused a positive 'diluting' effect on the FA.

Total concentrations of 16 PAH in the dry FGD residues ranged from 182 to 312 µg/kg. They also appeared to be close to those of 'pure' FA, with respect to quantitative composition and proportions of PAH compounds. While no distinct effect of dry FGD processes on PAH occurrence was noticed, the semi-dry processes result in an order of magnitude reduction in the concentrations of 16 PAHs both in high-FA and low-FA semi-dry FGD residues, mainly due to the decrease of naphthalene content, which was the dominant compound.

2.3 Summary

FA has a fine texture whilst that of FBC ashes and FGD residues is coarse. The dark grey colour of FBC ashes may be the result of an absence of NOx scrubbing and SOx removal processes. The

Table 6 Physical characteristics of CCWs (Seshadri and others, 2013)

	FA	FBC	FGD
Texture	fine	fine to coarse granules	coarse granules
Colour	white	dark grey	white
Particle size, µm	0.01–100	0.2–200	1–740
Bulk density, kg/m^3	960–1600	1520–680	780–1250
Moisture content, wt%	7.8–9	21.5–27.5	8–10
Volatile matter, wt%	4.7	33.1	19.6
Ash content, wt%	79.6	9.1	6.7

particle size ranges from 0.01 to 740 µm in the order of FA < FBC < FGD. Bulk density and moisture content are high in FBC ash. Ash content is low in FBC and FGD compared to their conventional counterpart. Table 6 compares the physical characteristics of FA, FBC ashes and FGD residues.

The chemical properties of CCWs are largely determined by the metal oxides. In FA, the predominant elements are Si, Al, Fe, and Ca. For FBC and FGD residues, Ca and S based compounds may predominate due to the calcium based additives employed to capture SO_2. With respect to trace metal contents, FGD products exert a positive 'blending' effect on the FA. The semi-dry desulphurisation processes cause a considerable reduction in organic compounds which are proven carcinogens, though their occurrence is generally low and non-problematic. The chemical composition of FGD residues will also affect the environmental behaviour of the disposed or utilised end products.

3 Legislation

Regulations on CCWs disposal vary from country to country. The foundation for regulating CCWs relies on the classification of CCWs. Traditionally, industrial wastes can be classified as *solid wastes* or *hazardous wastes*. In most countries, CCPs are considered as solid wastes. This may be for the purpose of encouraging the use of coal and avoiding an unnecessary regulatory burden on the utility industry. It may also be for the benefit of re-using CCPs.

To date, the only international treaty which addresses CCPs, administered by the United Nations (UN) and the Council of the Organisation for Economic Co-operation and Development (OECD), is the *Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and Their Disposal*, usually known as the *Basel Convention* (*see* www.basel.int). The Convention was opened for signature in 1989, and entered into force on 5 May 1992. As of March 2013, 180 countries, as well as the European Union, are parties to the Convention, while Haiti and the USA have signed the Convention but not ratified it. The *Basel Convention* sets out a two-step process for determining whether a waste is hazardous:

- the waste must belong to one of the categories listed in Annex I to the Convention*, Categories of wastes to be controlled*;
- it must possess at least one of the characteristics listed in Annex III to the Convention, *List of hazardous characteristics*.

CCPs are not a hazardous waste unless they or their leachates contain Annex I substances at levels that cause them to exhibit Annex III characteristics. However, in some instances, leachates from CCW disposal sites have an impact on the environment.

Further to the Basel Convention, the OECD *Decision of the Council C (2001) 107/Final* (OECD, 2004) concerns the control of trans-boundary movements of wastes destined for recovery operations. It specifies categories of waste which can be subjected to recovery procedures. This list includes fly ash, bottom ash and slag tapped from coal-fired power plants and specifies the conditions for recovery. The OECD recommendation notes that: ‘Classification of waste according to its hazardous properties should be independent of local or regional conditions. The Basel Convention aims to control the trans-boundary movement of hazardous waste. Site-specific analysis is inappropriate in that context because the result would be different classification determinations at different sites for the same waste, which would be confusing at best. Therefore, the system of classification is based upon the principle of intrinsic hazard of the waste or its chemical constituents’ (Ilyshechkin and others, 2012).

This chapter briefly reviews regulatory issues for CCPs in European Union and a number of countries around the world.

3.1 European Union (EU)

On the Europa website (www.europa.eu) there is an EU law section which is searchable by year and regulation number. Summaries of EU legislation can be found from the Europa site at: europa.eu/legislation_summaries/index_en.htm. Waste management regulations, including the *Basel Convention*; *General Framework* directive on waste, waste management statistics, and landfill of waste, are filed under the *Environment* section. On the European Commission (EC) part of the Europa website, there is a collection of EU waste legislation (ec.europa.eu/environment/waste/legislation/index.htm). The site provides links to individual pieces of EU legislation under the five sub-categories:

- Framework European Union legislation on waste;

- European Union legislation on waste management operations;
- European Union legislation on specific waste streams;
- Reporting and questionnaire legislation;
- Useful links to other in/directly related legislation.

A summary of the EU legislation most relevant to CCW management is below.

Apart from following the Basel Convention and OECD Decision C, using the same terminology, principle, and general intent of the Basel Convention, the EU compiled the *European Waste Catalogue* (EWC), a classification system for waste based on a combination of what they are and how they are produced. The member states are required to incorporate the EWC into their relevant legislation (*see* UK section). The EWC was then replaced by the *European Waste Classification for Statistics* (EWC-STAT), now in its fourth version (as of 2010).

The EWC-STAT is mainly a substance oriented aggregation of the waste types defined in the European List of Wastes (LoW) (Commission Decision 2000/532/EC). The result is a 1:n relationship between EWC-STAT and LoW, which allows for the unambiguous conversion of the waste types, classified according to the LoW, into EWC-STAT categories. The transposition table between the EWC-STAT and LoW is established in Annex III of EC regulation 2150/2002 (Eurostat, 2010). The EC regulation 2150/2002 was amended in 2010 as commission regulation (EU) no. 849/2010 (EC, 2010). In this document, combustion waste is listed in Annex I items 44 (non- hazardous) and 45 (hazardous) coordinating to item 12.4 in the list of EWC-STAT version 4. In Annex III, coal fly ash, bottom ash, slag and boiler dust are classified as *non-hazardous* waste in section *12.42 – Slags and ashes from thermal treatment and combustion*.

The LoW is the waste classification in the EU for administrative purposes, for example, permitting and supervision in the field of waste generation and management. According to Decision 2000/532/EG, the LoW should be revised regularly on the basis of new knowledge and, in particular, of research results. Whereas the EWC-STAT is used for the reporting of data to Eurostat, it does not prescribe a specific classification to be used for data collection. Countries are free to use any waste classification as long as they can produce the defined formats in the required quality. In practice, most of the countries collect their data according to the LoW and subsequently convert it into the required EWC-STAT categories on the basis of the transposition table in Annex III of EU regulation no 849/2010 (Eurostat, 2010).

The disposal of ash in landfill is governed by EU directives, such as the Council Directive 1999/31/EC. The Directive was amended in 2003 and 2008. Related acts and reports can be found on the Europa website at: europa.eu/legislation_summaries/environment/tackling_climate_change/l21208_en.htm. In December 2008, the mechanism of end of waste (EoW) criteria was introduced by Article 6 of the Waste Framework Directive (available at: susproc.jrc.ec.europa.eu/activities/waste). The objective of EoW criteria is to remove the administrative burdens of waste legislation for safe and high-quality waste materials, thereby facilitating recycling. EoW is the point at which full recovery of a waste is deemed to have taken place, allowing the material to be considered as a product or waste (Carroll, 2013). The environmental regulators in member states interpret the Waste Framework Directive and devise local EoW criteria. These EoW criteria have considerable regulatory and commercial impacts, including impacts on CCW disposal.

On 1 June 2007, the REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals) regulation entered into force. The aim of the regulation is to improve the protection of human health and the environment from the risks of chemicals while enhancing the competitiveness of the EU chemicals industry (Feuerborn, 2010). Each producer or importer of CCPs has to register the marketed or imported substances at the European Chemistry Agency situated in Helsinki, Finland. This regulation is mainly concerned with CCP utilisation and so will not be discussed any further.

The European Coal Combustion Products Association (Ecoba) was founded in 1990 by European energy producers to ensure full beneficial and high grade utilisation of all CCPs. Ecoba is active in the development of European standards.

All the EU member countries classify CCPs as non-hazardous and they interpret the EU regulations into their own domestic legislation. This report will discuss the UK as an example (*see* Section 3.7). Other EU countries' CCPs management practices are listed in Table 7.

In the countries which promote co-combustion of biomass, such as the Netherlands, considerable

Table 7 CCP regulations used in selected countries in Europe

	Hazardous	Regulation used and reference
European Union	No	EWC-STAT (EC, 2010)
Austria	No	Landfill regulation, in Federal waste management plan 2011 vol 1. Available at: www.bundesabfallwirtschaftsplan.at
Croatia	No	Waste regulation, in Regulation on categories, types and classification of waste with a waste catalogue and list of hazardous waste. Available at: www.mzoip.hr/default.aspx?id=7259 (Mar 2009)
Czech Republic	No	National law, Kyte and others (1999)
Denmark	No	Waste regulation, in Classifying green list waste under the 'Waste shipments regulation' (no 1013/2006) practical guidelines. Available at: www.mst.dk/English/Waste/Shipment_of_waste/Regulation_on_waste.htm (2011)
Finland	No	Waste regulation, in Government decree on waste. Available at: www.finlex.fi/en/laki/kaannokset/2012 (Apr 2012)
France	No	Landfill regulation, Kyte and others (1999)
Germany	No	Waste regulation, in Further development of European regulations on shipments of waste. Available at: www.bmu.de (2007)
Greece	No	Waste regulation. Solid waste disposal versus mining and mining wastes, available at: www.biutec.at/safemanmin
Ireland	No	Waste regulation, EC (2011) Shipments of hazardous waste exclusively within Ireland regulations. Available at: www.irishstatutebook.ie/2011/en/si/0324.html
Italy	No	Waste and landfill regulation, Crillesen and others (2006)
Netherlands	No	Waste regulation, Crillesen and others (2006)
Portugal	No	Waste regulation, Kyte and others (1999)
Romania	No	Landfill regulation, in Environmental Performance Reviews Series No. 37 Romania, available at: www.unece.org/fileadmin/DAM/env/epr/epr_studies/Romania_II.pdf (2012)
Slovak Republic	No	Waste regulation, in Decree of the Ministry of the Environment of the Slovak Republic Regulation No. 284/2001, available at: archive.basel.int/legalmatters/natleg/byparties/dnn-frmbody.php?partyId=134 (2001)
Spain	No	Waste and landfill regulations, Crillesen and others (2006) and Kyte and others (1999)
United Kingdom	No	Waste and landfill regulations, UK EA (2006)

work has been done to verify the toxicity of the ashes. Results show that no dangerous substances are found in the co-combustion ashes. The Netherlands classifies cofiring ashes as non hazardous according to the EWC (te Winkel, 2010).

3.2 Australia

According to the Senate 2008 report *Management of Australian waste streams*, Australia lacks fundamental information on most aspects of waste generation and management (Ilyushechkin and others, 2012). In Australia, CCPs are defined as an industrial waste (Heidrich, 2011) and regulations mirror those being applied in the EU and in the USA. There are no specific federal regulations that apply to the disposal of CCW. At the State level, only those states that contain the major coal mines and coal-fired power plants, for example, New South Wales (NSW), Queensland, Victoria and Western Australia (WA), have paid attention to the disposal of CCW. It seems that all states rely on general environmental regulations to control coal ash disposal rather than regulations that specifically apply to ash (Riley, 2005). This approach appears to be similar to that used in the EU, whereas, in the USA there are regulations that specifically apply to the disposal of CCWs (*see* Sections 3.1 and 3.2.7 respectively).

Based on Riley's (2005) summary of a report written by Aynsley and others in 2003 and on the recent report by Ilyushechkin and others (2012), the Australian legislation on CCWs is summarised as follows:

- Hazardous Waste Act 1989 is identified as the most relevant legislation of the Commonwealth of Australia. The Act is an implementation of the Basel Convention. The authors indicated that there are four UN hazardous characteristics that could be applied to coal ash;
- In NSW, coal ash falls under waste legislation. Effective as of 14 June 2011, the Australian Environmental Protection Authority (EPA) has stipulated a general exemption under Protection of the Environmental Operations (POEO) regulation 2005 part 6, clause 51 and 51A. This is the *Coal ash exemption 2011* (EPA NSW, 2011). The document first defines the terms used in the exemption, followed by the general conditions for coal ash land disposal. Responsibilities of generator, processor, and consumer are stated. It also provides the chemical and other material property requirements, sampling and testing requirements, and test methods;
- In the Australian Capital Territory Act, use and disposal of coal ash follow the general environmental regulations. There are no specific rules for CCWs;
- In Victoria, fly ash is a prescribed waste under the *Prescribed waste regulation 1998* (which is repealed now). Non-prescribed industrial wastes are controlled by Victoria EPA when disposed to landfill. These wastes can be accepted at solid inert landfills or municipal solid waste sites licensed by Victoria EPA to accept this type of waste;
- In South Australia (SA), details on legislation are limited. According to Riley (2005), coal ash is regulated as controlled waste, primarily due to the over-ruling definition of the Commonwealth. However, Ilyushechkin and others (2012) report that guidelines for solid waste, including criteria for assessment, classification and disposal, are currently under development. There are no specific definitions for coal ash and other CCPs in SA;
- In Western Australia, no legislation on the use and disposal of CCPs is available. Coal ash as a waste is covered by various environmental regulations. The *Waste Avoidance and Resource Recovery Act 2007* is one of the regulations to define waste;
- In the Northern Territory, in the *Crown Land Act* 1931-91, there are sections that reference permits to use land and waters for the purpose of ash disposal ponds;
- With effect from the 30 November 2007, Queensland EPA published and enforced an exemption for beneficial use. However, this approval is only applicable to the Ash Development Association of Australia (ADAA) and its members. Another exemption was introduced in March 2010. The notice of *General approval of a resource for beneficial use – coal combustion products (CCPs) used in bound final products* was issued by the Department of Environment and Heritage Protection of Queensland;

- In Tasmania, the classification scheme is used mainly for landfill permit reporting requirements and charging landfill fees. There are no specific definitions for CCPs in the classification scheme. The *Landfill Sustainability Guide 2004* provides guidance.

3.3 China

In China, CCW is classified as solid industrial waste (Yang and others, 2010). The two Chinese government agencies that are most closely connected with coal ash management are the National Development and Reform Commission (NDRC) and the Ministry of Environmental Protection (MEP). The NDRC is responsible for managing coal ash utilisation, while the MEP is concerned with preventing coal ash from polluting the environment. In the NDRC, departments relating to coal ash management are:

- Development and Planning Division;
- Regional economic departments, such as Department of Western Regional Development;
- Department of Resource Conservation and Environmental Protection.

For example, the Development and Planning Division's key responsibilities are to draw up a mid-to-long-term plan that provides the fundamental principles for national economic and social development. Within this framework, it could develop plans that provide clear directions for the handling of coal ash disposal and the prevention of environmental pollution within a specific time frame. The MEP is divided into 14 departments. Those responsible for handling coal ash are:

- Department of Science, Technology and Standards;
- Department of Total Pollutants Control;
- Department of Environmental Impact Assessment;
- Department of Environmental Monitoring;
- Department of Pollution Prevention and Control.

Of these, the Department of Environmental Impact Assessment is responsible for conducting environmental impact assessments on coal-fired power plant construction projects, including ash management. The Department of Pollution Prevention and Control is the most directly concerned with the prevention of coal ash pollution.

The policy most closely related to the prevention of coal ash pollution is the *Law on the Prevention and Control of Environmental Pollution by Solid Waste* (revised 2004 and in force from 1 April 2005). The MEP is responsible for its enforcement. Other existing relevant environmental protection laws and a variety of local and regional statutes on environmental protection and resource utilisation are also applied to CCWs management. However, all of these are the basic laws. Their contents are fairly general and are therefore difficult to apply in practice.

Some mandatory standards are concerned with preventing the dispersal, leakage or run-off from CCW. Of those used, the *Standards for Pollution Control on the Storage and Disposal Sites for General Industrial Solid Waste* is the most relevant. There are also more than 20 national and industry standards that are currently in use or are being drawn up. Only a minority of these are mandatory; the others are voluntary standards that industry is merely encouraged to adopt. Therefore, irrespective of the content of these standards, their effectiveness is too limited to serve as a strong control to the practices of coal-fired power plants. This is one of the main reasons why it is very difficult for China to make any major headway in preventing coal ash pollution (Yang and others, 2010).

While the economic ministries and their resource utilisation policies have grown stronger, the environmental ministries and their environmental protection legislation have been relatively weakened. There has been a lack of co-operation between the bodies governing comprehensive utilisation and pollution control. The most common way the MEP enforces its law is by levying a pollution charge. The *National Standards and Management Practices for Sewage Fee Collection* stipulates that if either non-specialised or specialised storage facilities or handling facilities do not meet environmental protection standards on the discharge of industrial solid waste (that is, they are unable to prevent pollutant dispersal, leakage or run-off), a one-off solid waste emissions fee will be levied. One tonne of discharged coal ash carries a 30 yuan fee. In practice, unless a coal-fired plant fails to build any kind of coal ash storage facility, or is discovered directly discharging coal ash waste into water sources or is engaged in other very serious polluting behaviour, the chances of it being charged pollution fines are extremely low. Even if it is charged RMB 30 yuan per tonne, this is not an effective deterrent. But because of a lack of effective regulations, apart from levying fees, the MEP has no other recourse to force a coal-fired plant to improve its coal ash storage facilities. Yang and others (2010) have uncovered that some local environmental bureaux have ignored the environmental laws, creating a lack of enforcement in environmental monitoring. Sometimes construction of a coal-fired power plant starts without even its environmental impact assessment being approved.

In the 1990s, there were major developments in administrative laws and regulations aimed at encouraging the utilisation of coal ash. The most relevant one is *Administrative measures on comprehensive utilization of coal ash* (1994). The rapid development of China's coal-fired power industry comes with an ever increasing output of coal ash, reaching 480 Mt by 2011 and expected to reach 570 Mt by 2015, which poses environmental issues. Also, according to China's current policy, once coal ash has been recycled into other byproducts or finished products (for example, concrete, bricks, dam materials), it no longer falls under the jurisdiction of environmental protection laws on solid waste. This has created a gap in environmental protection legislation and government monitoring on coal ash utilisation. Unlike the European Union, which regulates recycled coal ash products as commercial chemical products, China lacks such a corresponding regulation. There is an urgent need for such new rules and codes of conduct to be introduced. In responding to this urgency, the previous 1994 management code has been revised by the NDRC, Ministry of Science and Technology, Ministry of Finance and seven other ministerial organisations. The new regulation, which aims to stimulate the utilisation coal ash, was published in January 2013 and took effect on 1 March 2013. The regulation encourages the application of coal ash in producing wall materials, cement and concrete. Alumina extraction from high-aluminum coal ash is also suggested. Relevant government departments will subsidise the research and study of integrated use of coal ash, according to the new rules. There are five chapters in the new regulation document with 28 items (NDRC, 2013). The main changes are:

1. Clarification of the concept of coal combustion ash;
2. Enhancement of whole process management, including:
   a. Apart from ordinary coal-fired power plant, power plants firing coal gangue and coal slimes are also included;
   b. for new power plants and plant extensions, coal ash management must be included in the feasibility research and application;
   c. avoid building permanent ash ponds. If they must be built, the size should not be bigger than three years storage;
   d. encourage alumina extraction from high-aluminum coal ash;
3. Identification of the duties for each organisation in terms of coal ash management;
4. Implementation of this new regulation starting from the province level;
5. Co-operation with other related laws and regulations, such as the law on solid wastes management.

3.4 Japan

Information in English on Japan's CCPs situation is very limited. The Japanese government place much emphasis on the need for recycling and minimising waste. On its website (*see* www.jcoal.or.jp), the Japan Coal Energy Center (JCOAL) states that it has been developing technologies for effective usage of coal ash as one of its main activities. JCOAL collaborates with the Federation of Electric Power Companies and the Japan Fly Ash Association in the collection of statistics on coal ash production and utilisation. With more than 95% utilisation of CCPs (Federation of Electric Power

Companies of Japan, 2012), it is likely that specific legislation on CCPs is regarded as unnecessary. Although there are no specific national laws on CCWs disposal, some sections of the Basic Environment Law of Japan are related to disposal of wastes, including CCWs (Ilyushechkin and others, 2012).

3.5 Korean Republic

In the Korean Republic (South Korea), solid waste is classified as household waste or industrial waste. Hazardous waste is defined as designated waste, which is hazardous industrial waste or infectious (medical) waste. Solid wastes, including hazardous waste, are controlled under the Solid Waste Management Act. Ash, including CCPs, is non-hazardous unless its hazardous content exceeds the criteria of the Korean Leaching Test (Yoon and others, 2013). Riley (2005) cites Isherwood's observation that Korea has abundant land for the disposal of ash. However, no specific legislation for CCPs disposal has been identified. In Korea, most fly ash is recycled in kilns and bottom ash is disposed in landfill sites. Since 2003, the import volume of coal ash from Japan has increased dramatically; some of the fly ash generated in South Korea was no longer recycled which has led to increased use of landfill sites (Chung, 2011).

3.6 South Africa

In recent years, the South African government has made efforts to regulate waste management. A series of waste regulations were published or revised (available from www.sawic.org.za). Of these, those relevant to CCWs disposal are:

- Department of water affairs and forestry (2005) *Minimum requirements for the handling, classification and disposal of hazardous waste, 3rd edition*;
- Republic of South Africa (2009) *National environmental management: Waste Act, 2008: No 59 of 2008*, Government Gazette No 32000;
- Republic of South Africa (2010) *National waste information regulations in terms of the National Environmental Management: Waste Act, 2008 (59/2008)*, Government Gazette No 33384;
- Department of Environmental Affairs (2011) *National waste management strategy*, Government Gazette 35306;
- Republic of South Africa (2012) *National environmental management: Waste Act (59/2008): Waste information regulations*, Government Gazette 35583;
- Republic of South Africa (2012) *National environmental management: Waste Act (59/2008): Waste classification and management regulations*, Government Gazette 35572;
- Department of Environmental Affairs (2012) *National waste information baseline report*.

These documents clarified the definitions and classifications of wastes. In South Africa, wastes are placed in two classes, general or hazardous, according to their inherent toxicological properties. Hazardous wastes are further subdivided according to the risk that they may pose at disposal using a hazard rating. Fly ash and bottom ash are classified as general waste, but could potentially be classified as hazardous since they fall into a high volume/low hazardous waste stream according to the South African Department of Water Affairs and Forestry (2005) classification. Therefore, the *National waste information baseline report* put coal fly ash and bottom ash into a separate group as unclassified (Department of Environmental Affairs, 2012). However, there is a call for bottom ash to be classified as hazardous waste and for caution to be applied at its disposed in order to avoid potential high risk to the environment (Euripidou, 2012).

3.7 UK

According to the UK Environment Agency (EA), the UK has integrated the EU regulations into its own domestic legislation. The *List of waste (LOW) regulation 2005* brings the European Waste Catalogue into law in England and Wales and provides codes for all hazardous and non-hazardous wastes. The UK EA issues a guidance on how to interpret the LOW in the form of a waste thesaurus, an alphabetical list of over 1300 descriptions of wastes matched with the appropriate code (UK EA, 2006). Coal fly ash, bottom ash, and slag are listed in Chapter 10 '*Inorganic wastes from thermal processes*' and are not identified as hazardous. The UK EA website has a section on waste containing all the relevant regulations, www.environment-agency.gov.uk/business/topics/waste/default.aspx.

The *Pulverised Fuel Ash and Furnace Bottom Ash Quality Protocol* was launched in October 2010 by the UK Waste & Resources Action Programme (WRAP) and the Environment Agency. This document details EU's end of waste (EoW) criteria for the production of fly ash and furnace bottom ash arising from combustion of coal for use in bound and grout applications in specified construction and manufacturing processes. If these criteria are met, the resulting outputs will normally be regarded as having been fully recovered and to have ceased to be waste and therefore without the need for waste management controls. The protocol applies to England, Wales and Northern Ireland (WRAP, 2010).

Pulverised fuel ash, including coal ash, is a controlled waste in the UK and has no special requirement for disposal at appropriately licensed facilities. Ashes are disposed of in both ash dams and landfill sites, and are regulated under the 1994 waste management licensing regulations. Under the *Statutory Instrument 1996 No 1528 The Landfill Tax (Qualifying Material) Order 1996*, fly ash and bottom ash are classified as inactive and attract a lower rate tax than other wastes (Euripidou, 2012; Kyte and others; 1999; Riley, 2005).

3.8 USA

In America, new regulations for CCW management are under development by EPA and anticipated for 204. For now, the Federal regulations that apply most particularly to CCWs management are:

- Resource Conservation and Recovery Act (RCRA);
- Surface Mining Control and Reclamation Act, 1997 (SMCRA).

Another federal regulation that may, in some circumstances, be relevant to the disposal of coal ash is the *Superfund Act* (*see* www.epa.gov/superfund), established by the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) of 1980. The SMCRA has been reviewed throughly in another IEA CCC report (Sloss, 2013). This section concentrates on the RCRA.

The RCRA provides general guidelines under which all waste is managed. It includes a congressional mandate to the US Environmental Protection Agency (EPA) to develop a comprehensive set of regulations to implement the law. Enacted in 1976, RCRA was intended, in part, to protect human health and the environment from the potential hazard from waste disposal and to ensure that wastes are managed in an environmentally sound manner. Subtitle C of RCRA created a hazardous waste management programme that, among other provisions, required EPA to develop criteria for identifying the characteristics of 'hazardous' waste and to develop waste management criteria applicable to such waste. Subtitle D of RCRA established criteria applicable to 'non-hazardous solid' waste disposal. It also established state and local governments as the primary regulating and implementing entities for the management of solid waste, such as household garbage and non-hazardous industrial solid waste (Luther, 2010b).

Solid wastes are defined as any discarded material in the RCRA regulations. The regulations specify that a solid waste becomes a hazardous waste when exhibiting one or more of the following characteristics: toxicity, reactivity, ignitability or corrosivity or if EPA specifically lists the wastes as hazardous. If CCW is to be characterised as hazardous, it would likely be because hazardous constituents in the waste exceed regulatory toxicity levels. Generally, CCW does not exceed toxicity levels. However, the nature of CCW is unique, relative to other hazardous waste, because it is generated in huge volumes, with large amounts of inert, benign materials that effectively dilute what may be overall significant amounts of hazardous constituents in an entire CCW landfill or surface impoundment (Luther, 2010a).

The evolution of CCW regulation has a long and complicated history. A timeline is used here to summarise American key developments and efforts on CCW regulations (EPA website: www.epa.gov; Luther, 2010a,b; Rokoff and others, 2011):

1976	RCRA enacted;
1978	EPA first proposed hazardous waste management regulations under subtitle C;
1980	1980 Bevill amendment to the RCRA – CCW was excluded as hazardous waste from subtitle C and required EPA to 'conduct a detailed and comprehensive study and submit a report' to Congress;
1988	EPA published *Report to Congress on wastes from combustion of coal by electric utility power plants*. The report concluded that CCW did not exhibit RCRA hazardous waste characteristics and the current management practices appeared to be adequate;
1993	EPA published a regulatory determination that the four large volume coal combustion wastes did not warrant regulation as hazardous waste under Subtitle C of RCRA;
1999	EPA published *Report to Congress on wastes from the combustion of fossil fuels*;
2000	EPA published final regulatory determination that fossil fuel combustion wastes, including CCW, do not warrant regulation as hazardous waste under Subtitle C of RCRA but national regulations under RCRA subtitle D (solid waste requirements) were warranted for CCW when it is disposed of in landfills or surface impoundments. It was also concluded that no additional regulations were warranted for CCPs used beneficially;
2001	EPA held several meetings with stakeholders regarding the use and disposal of CCPs. In the public notice for its March 2004 meeting, EPA stated that the 'Agency remains concerned about coal combustion byproducts because of the potential for environmental damage; the lack of groundwater protection via monitoring and/or liners; and widely varying state regulatory programs';
2002	EPA issued two draft reports: *Regulation and policy concerning mine placement of coal combustion waste in selected states* and *Mine placement of CCW: state program elements analysis*. The reports reviewed and summarised state regulations and policies concerning the placement of CCW in surface and underground mines;
2006	The National Academy of Sciences (NAS) National Research Council (NRC) issued a report: *Managing coal combustion residues in mines* which recommended that the Department of the Interior's Office of Surface Mining (OSM) takes the lead in CCW disposal standards under the Surface Mining Control and Reclamation Act of 1977;
	EPA and DOE published a joint report: *Coal combustion waste management at landfills and surface impoundments, 1994-2004*. The report concluded that, since 1988, a majority of the states tightened regulation of landfill liners, leachate-collection systems, and groundwater monitoring for new disposal units;
2007	OSM released an Advanced Notice of Proposed Rulemaking (72 FR 12025) regarding *Placement of coal combustion byproducts in active and abandoned coal mines*.
	EPA published reports *Coal combustion waste damage case assessment*, which determined 24 cases of proven damage and 43 cases of potential damage associated with CCW landfills and surface impoundments, and *Human and ecological risk assessment of coal combustion wastes*;
	EPA issued a Notice of Data Availability (NODA) on *Disposal of coal combustion wastes in landfills and surface impoundments* and sought public comments. Documents made available under the NODA included most of the above reports;
2008	The NODA comment period was extended until February 2008;
	22 December 2008 failure at TVA's Kingston plant occurred;
2009	EPA issued *Information request letter on the structural integrity of coal combustion residuals in surface impoundments* to compile, prioritise and improve existing ash impoundments

	which lead to announcement of 49 high hazard sites. Data also showed that over 60% of states do not require liners or groundwater monitoring for surface impoundments;
2010	On 21 June, EPA published *Hazardous and solid waste management system; identification and listing of special wastes; disposal of coal combustion residuals from electric utilities; proposed rule*. It proposed two regulatory options applicable to the management of CCW, basically subtitle C or D. 90 days comment period started. Eight public hearings held across the country and over 450,000 comments received, of which 10,000 were significant; EPA issued NODA on *Coal combustion residual surface impoundments*;
2011	Post election, funding for EPA action labelling coal ash a hazardous waste was cut off in February. This does not prevent EPA from moving forward with rules but may signal a shift in political pressure; March, EPA Administrator Lisa Jackson stated that the agency was undertaking evaluations of beneficial uses of CCPs through Coal Combustion Products Partnership (C_2P_2) programme and would use the findings in creating a final regulation; EPA issued NODA on *Disposal of coal combustion residuals from electric utilities*;
2012	ACAA estimated that EPA would not be able to finalise the coal ash disposal regulation before the end of year;
2013	New EPA regulations and guidance remain pending.

Since EPA published its proposal in 2010, various concerns regarding the proposed options have been expressed. Industry groups argue that the Subtitle C option is not justified and would be too costly to implement which would ultimately have an impact on energy prices. Also it may potentially limit recycling opportunities. The American Coal Ash Association (ACAA) (Adams, 2011) and the Environmental Council of the States (ECOS) (Brown, 2012) share this concern. State representatives are concerned about the cost of implementing a subtitle C regulatory programme. They further argue that EPA has not demonstrated that existing state regulatory programmes are in fact deficient in protecting human health and the environment. Some members of Congress have expressed some of the same concerns. Environmental and citizen groups, such as Earth Justice (*see* www.earthjustice.org/coalash), argue that new data demonstrate that the waste meets the regulatory criteria necessary to list it as hazardous under Subtitle C. Furthermore, the EPA lacks the authority to enforce the disposal criteria as it proposes under Subtitle D. Some members of Congress share this concern as well (Luther, 2010b). Decisions are pending.

As EPA's final regulation has been delayed, Congress has attempted but failed to pass legislation by both the House and Senate that then has been signed into law. A bill to provide for the regulation of coal, *The Coal Residues Reuse and Management Act, H.R. 2273*, was passed by the House of Representatives. Because of the importance of coal ash as a construction material in the transportation industry, H.R. 2273 was offered as an amendment to the House version of the Transportation Bill, H.R. 4348. Although the coal ash amendment was removed from the transportation bill, it was amended to provide certain improvements, and that revised bill has been introduced into the Senate as S 3512. Each of these pieces of legislation would amend Subtitle D of the RCRA to set the bar for the regulation of coal ash by establishing a robust set of minimum federal requirements for the management and disposal of coal ash that will ensure safety and the protection of human health and the environment (Bradley, 2012).

Until EPA finalises the regulation, management of CCPs is the responsibility of individual states. Forty-five states, encompassing 96% of America's coal-fired utility generating capacity, replicate the federal exemption of CCPs as a hazardous waste.

3.9 Comments

There is currently a lively debate in the USA, Europe, and South Africa about regulating coal ashes as hazardous waste, and the respective regulatory authorities are currently trying to finalise their regulations. At the moment, CCPs are generally classified as non-hazardous waste. It is often an additional note such as, ‘if the toxic substance exceeds the limit’, that decides whether the CCPs generated from a plant are hazardous or not.

4 CCW management methods

CCPs that are not utilised are usually sent for disposal which must be managed properly. This chapter discusses the disposal methods commonly used today.

Selecting the ecologically and economically most appropriate coal ash disposal method is a crucial, highly-site specific aspect of coal ash disposal management operations, where topographic, climatic, logistic, and regulatory factors constitute the most important boundary conditions in the decision-making process. Land requirement and consumption are critical aspects of CCW management. Depending on the disposal method, between 3000 and 8000 m^2 of land is required per installed megawatt (Dellantonio and others, 2010).

There are four principal methods for disposing of CCW:

- surface impoundment;
- landfill;
- mine and quarry fill;
- ocean disposal.

According to EPA (1988), nearly 70% of all generating units in the USA manage their CCW on-site. About two-thirds of the on-site facilities are surface impoundments, while most of the others are landfills. Landfills are used for about 95% of all off-site disposal in the USA. Ocean disposal is not commonly used today. It was mentioned in US EPA's report to congress (EPA, 1988) that ocean disposal of CCW was in the research and development phase. In the UK, dumping of fly ash into the sea was practised on a large scale in the North Sea but was stopped in 1992 (Recoal Consortium, 2008). However, related follow-up studies of actual large-scale use are not documented in the literature. Mine fills are covered in another IEA CCC report by Sloss (2013). She comments that the addition of CCW can actually improve the structural integrity of backfills in mine reclamation, whilst also providing neutralising potential to combat acid mine discharge. Smith (2005b, 2006) also discussed mine backfill when reviewing coal fly ash and FGD residues management. Therefore, this report focuses only on surface impoundments and landfills.

4.1 Surface impoundments

The American EPA (EPA, 2010a) states that:

Coal combustion residues (CCR) surface impoundment or impoundment means a facility or part of a facility which is a natural topographic depression, man-made excavation, or diked area formed primarily of earthen materials (although it may be lined with man-made materials), which is designed to hold an accumulation of CCRs containing free liquids, and which is not an injection well. Examples of CCR surface impoundments are holding, storage, settling, and aeration pits, ponds, and lagoons. CCR surface impoundments are used to receive CCRs that have been sluiced (flushed or mixed with water to facilitate movement), or wastes from wet air pollution control devices, often in addition to other solid wastes.

Surface impoundment units, often called wet ponds, hold liquid waste that is generally sluiced directly from a power plant to the impoundment unit, where solids settle out, leaving relatively clear water at the surface that may be re-circulated into the plant or discharged to surface water (*see* Figure 2). Solids may accumulate until the impoundment unit is full, or the pond may be drained and the solids dredged periodically and taken to another disposal unit such as landfill (Luther, 2010a). By using this method, certain types of waste treatment, such as neutralisation of acids, can be accomplished concurrently with disposal. Historically, wet ponding has been one of the most widely used disposal methods for coal ash and FGD wastes because it is simple and easily implemented. In recent years,

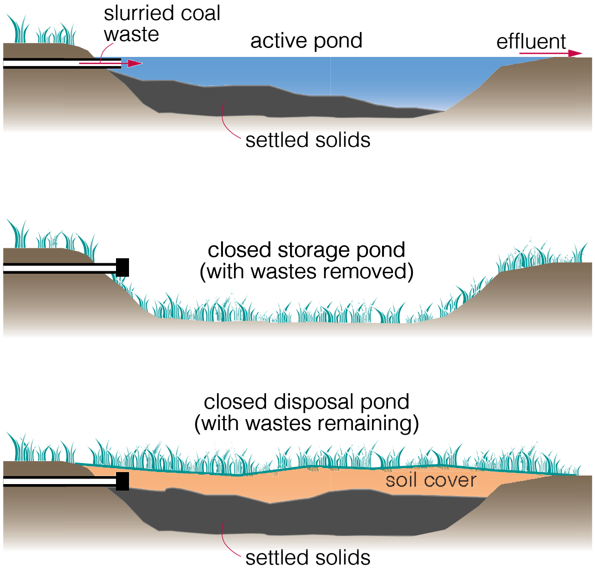

Figure 2 Stages in the life of a typical impoundment (EPA, 1988)

some state and local regulations concerning wet ponds have become more restrictive, requiring liners to prevent leakage and groundwater monitoring at these facilities. These types of restrictions will tend to increase wet ponding costs, making it less attractive as a disposal option.

Pond design varies widely depending upon local site conditions, the regulations that govern design of the impoundment, and whether the CCWs are to be disposed of and/or treated in the ponds. The total volume of an impoundment system depends on several factors, including the:

- total quantity of ash to be disposed of (both dry and slurried volumes);
- liquid and solid retention times;
- type and degree of treatment performed;
- desired quality of the discharge or effluent.

Utilities may use a single pond or a series of ponds to facilitate the settling of solids. Chemicals or different wastes can be added at different points in the ponding system to produce the desired chemical reactions, such as metals precipitation or neutralisation. The number of ponds in a system and the specific uses to which each is put can also influence the total volume required for wet ponding (EPA, 1988).

4.2 Landfills

Dry CCW is typically loaded onto trucks, wetted to prevent dust emission, and then transported to a dedicated landfill facility. The landfill may be located on the power plant property (on-site) or elsewhere (off-site). Landfilling CCW is a long-term proposition and it is usually placed on an area of land or an excavation for permanent disposal. The US EPA (2010a) defines a CCW landfill as:
. . . a disposal facility or part of a facility where CCRs are placed in or on land and which is not a land treatment facility, a surface impoundment, an underground injection well, a salt dome formation, a salt bed formation, an underground mine, a cave, or a corrective action management unit. For purposes of this proposed rule, landfills also include piles, sand and gravel pits, quarries, and/or large scale fill operations. Sites that are excavated so that more coal ash can be used as fill are also considered CCR landfills.

Landfills are used to dispose of CCW, such as fly ash, bottom ash, and FGD sludges, when they are produced or after they are dredged from surface impoundments that are used as interim treatment facilities. Most large landfills are divided into sections, or cells, of which only one or two may be active at any given time (*see* Figure 3). A landfill cell is defined as the area over which waste is placed to a depth ranging from 0.3 to 3 metres (industry practice refers to each layer of cells as a lift). Several lifts may be stacked on top of one another in the landfill. A cell may be open for periods ranging from a day to a few weeks, after which it is usually covered with 0.15 to several metres of soil. The waste and soils are often sprinkled with water throughout the fill operation to mitigate potential dust problems.

Excavation may be initiated in phases. For example, as one cell is filled, another is prepared for waste placement, while yet another is being excavated. Roads are built in order to provide access for waste hauling equipment as well as for the earth-moving and earth-compacting equipment that prepares the waste after it has been placed in the landfill cell. After a cell is filled, the access road frequently becomes part of the containment system as a wall separating one cell from the next.

Landfilling of coal ash and FGD sludges has increased over the past few years as the costs of wet ponding have risen and more stringent regulations have come into force. In the USA, most electric utilities that use landfills currently dispose of their high-volume wastes in Subtitle D (non-hazardous waste) landfills. Landfills in compliance with the RCRA Subtitle C standards may be used occasionally for disposal of small quantities of hazardous waste (EPA, 1988).

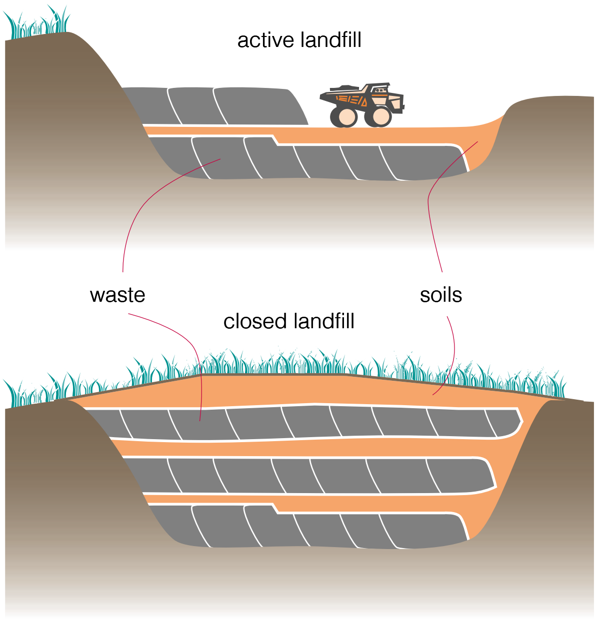

Figure 3 Active and closed landfills (EPA, 1988)

4.3 Comments

On-site surface impoundment has low delivery costs but may produce more leachate. It requires large quantities of water and about two and a half times more land than landfill, resulting in a significantly larger environmental footprint. After the catastrophic spill in Kingston, TN, USA (*see* Section 5.3), an even more pronounced trend towards dry ash disposal may be expected.

5 Environmental issues

CCW is generated in large volumes and has been accumulating at some sites for decades. Individual power plants may generate thousands to hundreds of thousands of tonnes of the waste each year, the majority of which is disposed of on-site. Based on the chemical properties outlined in Chapter 2, CCW is likely to contain certain hazardous constituents that pose a risk to human health and the environment if not properly managed, and adequate controls are not in place. As shown in Figure 4, the main hazards relate to:

- soil contamination;
- water contamination;
- dust dispersion;
- toxins entering the food chain.

Generally, there are two main potential risks associated with the disposal or use of CCW: surface or groundwater contamination, and a sudden release of liquid waste. There is also the danger of windblown ash. Radioactivity and mercury may come into contact with the general public when ash is dispersed in air and water or is included in commercial products. The primary concern regarding the management of CCWs usually relates to the potential for hazardous constituents to leach into the surface or groundwater, and hence contaminate drinking water, surface water or biota. A considerable amount of work has been done in this area, and therefore, leaching and water contamination will be discussed separately in the next chapter.

This chapter first summarises the US EPA's Human and Ecological Risk Assessment of Coal Combustion Wastes before discussing the factors affecting exposure and risk associated with CCWs disposal. Using the catastrophic US Kingston ash pond disaster as an example, this chapter discusses the potential for sudden release and its environmental impacts. The environmental impacts of windblown ash are also outlined. Mercury and radioactivity from CCWs will be reviewed as well.

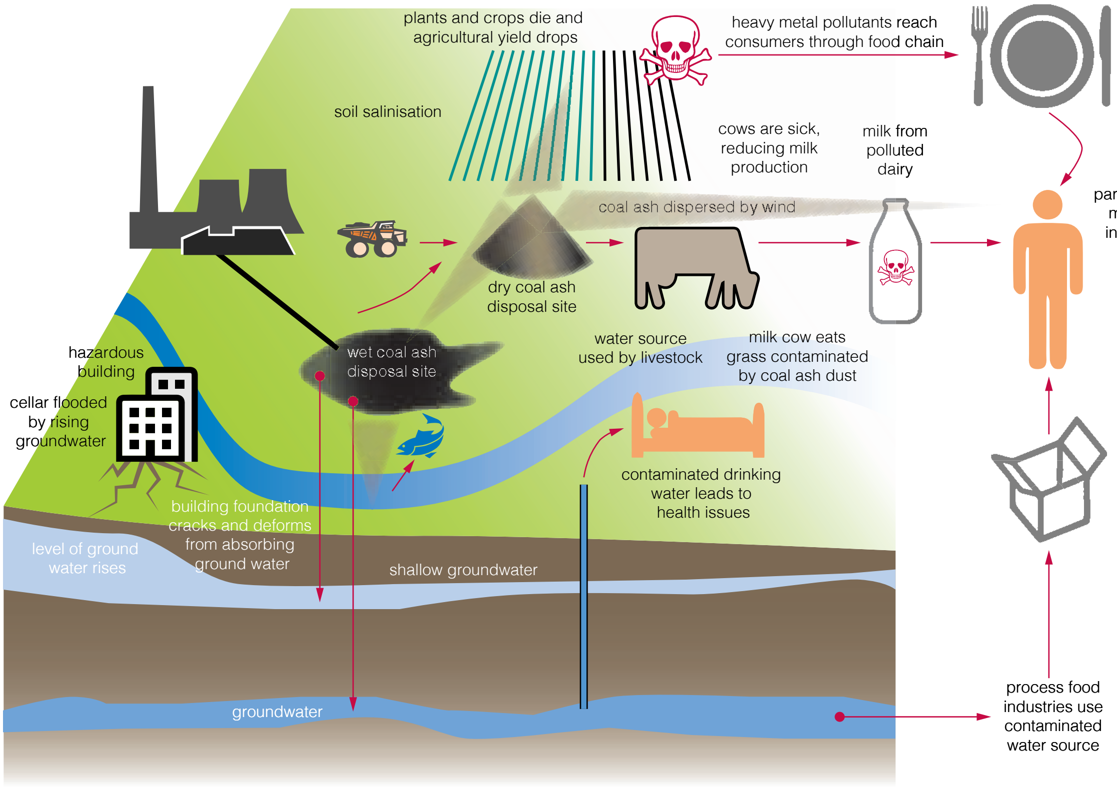

Figure 4 Potential hazards and contamination path ways (Yang and others, 2010)

5.1 US EPA risk assessment

Over the past several years, the US EPA has devoted significant resources to evaluate the human health and ecological effects from CCW management practices, including CCW disposal in landfills and surface impoundments. A US EPA risk assessment was first released in 2007 and updated in 2010.

In support of the 2007 risk assessment, the US EPA (2007b) assembled a constituent database that included leachate and total waste analyses for 41 CCW constituents taken from more than 140 CCW disposal sites around the country. The risk assessment of these wastes and leachates was carried out using the methodology shown in Figure 5. After hazard identification, the screening analysis looked at all probable exposure pathways from CCW management in landfills and surface impoundments, and identified 21 CCW constituents and three exposure scenarios to evaluate in the full-scale analysis (*see* Table 8). Exposure scenarios evaluated for people include contaminated groundwater being transported to drinking water wells from a CCW landfill or surface impoundment, and contaminated groundwater discharging into surface water and contaminating a nearby stream or lake where people catch and eat fish. The full-scale analysis also addressed ecological risks in the same water bodies. It was designed to characterise waste management scenarios based on two waste management options, namely disposal of CCW on-site in landfills and in surface impoundments. The risk assessment was also used to characterise waste management scenarios based on three liner types (unlined units, clay-lined units, and composite-lined units) and three waste types (conventional CCW, co-disposed CCW and coal refuse, and FBC wastes).

The results show that risks from clay-lined units are lower than those from unlined units, but 90th percentile risks are still well above the risk criteria for arsenic and thallium for landfills, and arsenic, boron, and molybdenum for surface impoundments. Composite liners effectively reduce risks from all pathways and constituents below the risk criteria (cancer and non-cancer) for both landfills and surface impoundments. Arrival times of the peak concentrations at a receptor well are much longer for landfills (hundreds to thousands of years) than for surface impoundments (most are less than a hundred years). Waste type has little effect on landfill risk results but in surface impoundments, risks are up to 1 order of magnitude higher for co-disposed CCW and coal refuse than for conventional CCW. The higher risks for surface impoundments than landfills are likely due to higher waste leachate concentrations, a lower proportion of

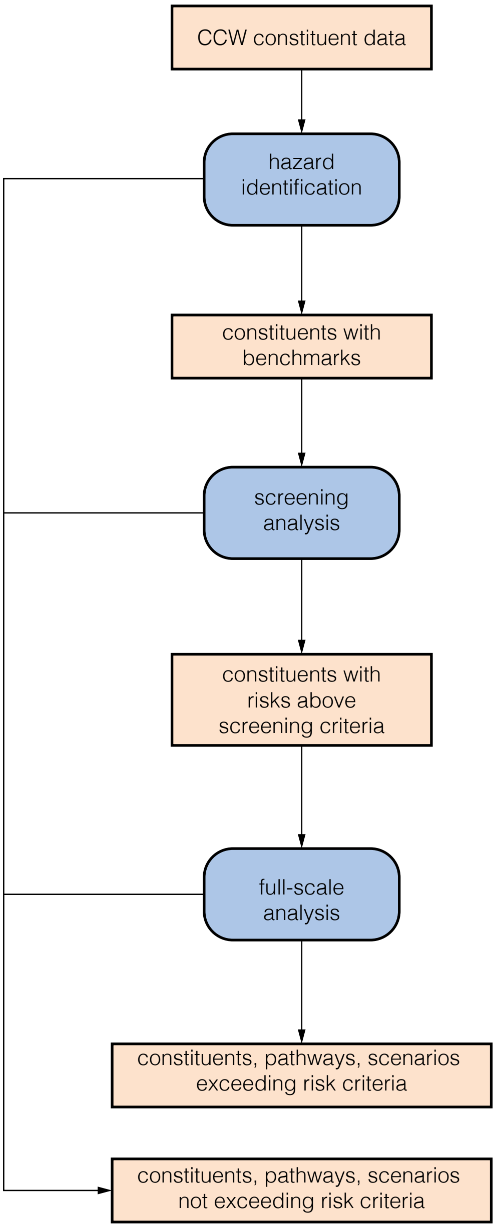

Figure 5 Overview of CCW risk assessment methodology (EPA, 2007b)

Table 8 Sources, releases, exposure pathways, and receptors evaluated in the CCW risk assessment (EPA, 2007b)

Release mechanism	Exposure pathway	Exposure mechanism	Receptor type*	Screening result
Landfills				
Leaching	Groundwater to drinking water	Residential well	Resident	Full scale analysis
	Groundwater to surface water	Stream or lake, uptake by fish; contact with water sediments	Recreational fisher; aquatic ecosystems	Full scale analysis
Water erosion	Overland transport to surface water	Stream or lake, uptake by fish; contact with water, sediments	Recreational fisher; aquatic ecosystems	Below screening criteria
	Overland transport to soil	Soil ingestion; uptake from soil by plants, beef, dairy	Subsistence farmer; terrestrial ecosystems	Below screening criteria
Wind erosion	Soil deposition	Soil ingestion; uptake from soil by plants, beef, dairy	Subsistence farmer; terrestrial ecosystems	Below screening criteria
	Fugitive dust	Inhalation	Resident	Below screening criteria
Surface impoundments				
Leaching	Groundwater to drinking water	Residential well	Resident	Full scale analysis
	Groundwater to surface water	Stream or lake, uptake by fish; contact with water, sediments	Recreational fisher; ecological receptors	Full scale

* Human receptor types include adults and children

lined units, and the higher hydraulic head from the impounded liquid waste. This is consistent with damage cases reporting wet handling as a factor that can increase risks from CCW management. For ecological receptors exposed via sediment, 90th percentile risks for Pb, As, and Cd exceeded the risk criteria for both landfills and surface impoundments because these constituents strongly absorb to sediments in the water body. The 50th percentile risks are generally an order of magnitude or more below the risk criteria.

However, the US EPA received many comments on the technical aspects of the analysis, the selection of models to estimate, and the characterisation of risks (available to review at: www.regulations.gov/#!docketDetail;D=EPA-HQ-RCRA-2006-0796). The American Utility Solid Waste Activities Group (USWAG) was one of the peers who were sceptical about the value of modelling CCW disposal risk (Roewer, 2008). Their comments on the EPA risk assessment included:

- The draft risk assessment is based on industry data submitted in 1995 that no longer accurately reflects current industry-wide management practices.
- The draft relies on the TCLP test to characterise the leaching potential from CCW disposal units. This test mimics the conditions of a municipal solid waste landfill, which result in an acidic environment mimicked by the TCLP. Utility CCW disposal units are generally monofills and tend to be alkaline.

- The risk assessment model has seriously mis-characterised the exposure pathway at CCW disposal units.
- The model makes some erroneous assumptions about exposure to CCW constituents from fish consumption. The model assumes that all persons living near disposal units are anglers and consume locally-caught fish and that all fish are contaminated by a groundwater plume from a CCW disposal unit.
- The draft risk assessment fails to demonstrate that it accounted for redox conditions of arsenic and other metals in assessing the transport of these metals in groundwater and the exposure risks associated with them.

In response to all the comments, the risk assessment was updated and re-released in 2010 but was still in draft form. The US EPA revised this risk analysis document to address comments on the analytical methodology, data, and assumptions used in the risk assessment (EPA, 2010b). A ‘Full- scale Risk Assessment’ was added to the ‘Problem Formulation’ section and a ‘Full-scale Modeling Approach’ was added to the ‘Analysis’ section. The new document added the finding that surface impoundments typically showed higher risks than landfills, regardless of liner type. For surface impoundments, co-disposal of CCW with coal refuse results in significantly higher risks from arsenic and certain other constituents than CCW disposal alone, while for other constituents, managing CCW alone results in higher estimated risks than co-disposal CCW. These risk results are in many cases consistent with damage cases compiled by the US EPA and others. For example, the full-scale modelling of selenium released from unlined surface impoundments into groundwater suggests that certain fish consumers may be exposed to relatively high levels of selenium, consistent with fish consumption advisories at some of the proven damage case sites. These results suggested that with a higher prevalence of composite liners in new CCW disposal facilities, along with practices to prevent co-disposal of coal refuse with CCW, future national risks from on-site CCW disposal are likely to be lower than those presented in this risk assessment.

The US EPRI (2011) still thinks that the risk assessment modelling will likely be of limited value in the rule making process. Their reasoning is that the modelling attempts to simulate all CCW types and sites in the USA, from leachate generation through groundwater transport to long-term health effects of potential receptors, over periods ranging from tens to thousands of years. Such an exercise is challenging for a single site risk assessment, let alone an aggregate of all CCW sites in widely varying geographic and hydro-geologic areas. Because of the large number and range of variables used to run the Monte Carlo simulations, the model results inherently have a high degree of uncertainty. The modelling performed to date established a higher risk associated with unlined sites compared to lined sites, which is a reasonable finding and is unlikely to change with additional model improvements. However, the crux regulatory question is designation of CCW as a hazardous waste under Subtitle C against non-hazardous waste under Subtitle D. Both the Subtitle C and D options in the US EPA proposal require similar composite liners for new or horizontally expanded facilities. Since the risk assessment modelling is largely driven by liner type, and the liners are the same under both the Subtitle C and D proposals, additional modelling will do little to distinguish between the two regulatory options. When considering the cost benefit in the Regulatory Impact Analysis (RIA), the calculated risks between the Subtitle C and D options do differ, but not because of the results of the risk assessment modelling. The difference is largely driven by the assumption that about half of the sites will not be upgraded to appropriate designs under Subtitle D due to lack of enforcement by the States. The assumption regarding state enforcement far outweighs any incremental improvements to the risk assessment modelling in terms of the US EPA risk-benefit calculation, again suggesting that spending more time on the risk assessment modelling is of little value in discriminating between the two options. Finally, the sites in the risk assessment modelling are not hypothetical. They have been in operation for periods ranging from a few years to more than 30 years. It would be of greater value to assemble existing field data from sites (some of which have been collecting groundwater data for more than 20 years), collect additional field data as warranted, and attempt to assess the existing transport modelling results, rather than continue to refine a hypothetical risk assessment model with significant inherent uncertainties.

5.2 Factors affecting the environmental impacts of CCW disposal

Apart from the constituents of CCW and its leachates, the impact that CCW disposal poses to the environment and human health is affected by a variety of factors (EPA, 1988). These factors are discussed below.

5.2.1 Environmental characteristics of CCW disposal sites

Environmental characteristics of CCWs disposal sites have a significant effect on the potential of the waste constituents to travel and reach receptor populations (EPA, 1988). Key environmental characteristics are:

- *Distance to surface water*: This is the distance between a coal combustion waste disposal site and the nearest surface water body. Proximity to surface water would decrease the possible health effects of groundwater contamination because there would be fewer opportunities for drinking water intakes before the groundwater reached the surface water body. Once the plume reached the surface water, contamination would be diluted. However, proximity to surface water would possibly increase the danger to aquatic life because less dilution of the contaminant plume would occur before the plume reached the surface water body.
- *Flow of surface water*: A high surface water flow will increase the dilution rate of coal combustion constituents that enter the surface water, thereby reducing concentration in the surface water.
- *Depth to groundwater*: A large depth to groundwater will increase the time it takes for waste leachates to reach the aquifer. It also allows more dispersion of the leachate before it reaches the aquifer, so that once the leachate reaches the aquifer, the metal concentrations would be lower.
- *Hydraulic conductivity*: A high hydraulic conductivity indicates that constituents will travel quickly through the groundwater and possibly more readily reach drinking water wells, although high conductivity also indicates a more rapid dilution of constituent concentration.
- *Net recharge*: This factor is a measure of net precipitation of a site after evapo-transpiration and estimated runoff is subtracted. Recharge is calculated in order to determine the amount of rainfall annually absorbed by the soil. A high net recharge indicates a short period of time for contaminants to travel through the ground to the aquifer, but will also indicate a higher potential for dilution.
- *Groundwater hardness*: Groundwater with over 240 ppm of $CaCO_3$ is typically treated when used as a public drinking water supply. This treatment of the hard groundwater has an indirect mitigative effect on exposure because treatment of the groundwater will tend to remove contamination from other sources.

5.2.2 Population characteristics of CCW disposal sites

Opportunities for human exposure to CCW constituents depend in part on the proximity of sites to human populations and to public drinking water. Many waste sites in the USA appear to be located on the outskirts of populated areas, with fairly low population density immediately adjacent to the site, but with significant populations within a 5 km radius. Usually the population density increases on average with distance from the disposal site. If CCW sites are close to public drinking water systems, there may be potential for human exposure through drinking water supplies.

5.2.3 Ecological characteristics of CCW disposal sites

If potential hazardous constituents of coal combustion waste do migrate and produce environmental contamination, they could affect species and natural communities that are particularly vulnerable, thereby lessening ecosystem diversity. As discussed earlier, depending on the proximity of CCWs disposal sites to surface water bodies, which provide animals with drinking water, there could be a high potential for species exposure to coal combustion constituents.

5.3 Ash pond sudden releases

Surface impoundment ponds hold liquid waste that has been sluiced from the power plant to the disposal area. It is generally held within the pond by depositing it in a natural or man-made depression in the ground or through the use of an earthen dyke. If part of the pond is breached, the liquid coal combustion wastes will be released into the environment.

The first widely recognised case of sudden release in America occurred in 1967 (Lemly and Skorupa, 2012). A dyke surrounding a fly ash settling lagoon collapsed spilling half a million cubic metres of caustic solution (pH 12) into the Clinch River, VA. Large numbers of fish were killed by the alkaline coal combustion wastes and benthic invertebrates were poisoned for 124 km downstream. The waste was eventually diluted, dispersed, and neutralised by natural physical/chemical processes. Two years after the spill, however, the river had still not fully recovered (EPA, 1988).

The most dramatic example of a sudden release case is the December 2008 spill at the US Tennessee Valley Authority's (TVA) Kingston coal-fired power plant. The retaining wall of its 50,000 m^2 ash pond ruptured, which released about 4 million m^3 of wet coal ash (fly ash with intermixed bottom ash), inundating the Emory River, its tributaries, and adjacent landscape near Harriman, TN. The ash spilled into the Emory River which joins the Clinch River, and then converges with the Tennessee River, a major drinking water source for populations downstream (*see* Figures 6 and 7). Hundreds of fish were found floating dead downstream from the plant. Water tests showed elevated levels of lead and thallium. Clean-up is expected to continue into 2014 and will cost $1.2 billion (Luther, 2012).

Immediately following the Kingston ash spill, TVA began comprehensive environmental monitoring and initiated investigations to evaluate immediate, intermediate, and long term impacts of the ash spill on natural resources and public health. The studies covered a broad spectrum of disciplines and environmental media, and were conducted with rigorous quality assurance/quality control. Results showed no significant risks to human health or to populations of benthic invertebrates and low risks to riparian and aerial feeding birds that consume benthic invertebrates. However, there was potential for low to moderate risk to populations of benthic invertebrates and low risks to riparian- and aerial-feeding birds that consume benthic invertebrates (Carriker and others, 2013). These findings proved the earlier investigation (Ruhl and others, 2010) that the concentration of some leachable coal ash contaminants such as boron, arsenic, strontium, and barium increased slightly downstream of the spill relative to the upstream river concentrations, but remained below the EPA's maximum contaminant level. It is worth noting that the Kingston fly ash pond discharged water containing readily dissolved constituents to the Emory River for decades, so the impact of the ash spill on some constituents in the river was minimal (Feuerborn, 2013)

According to TVA's inspection report (TVA, 2009), TVA could have possibly prevented the Kingston spill if it had taken recommended corrective actions. TVA was aware of various 'red flags' that had been raised over a long period of time, signalling the need for safety modifications to TVA ash ponds. These 'red flags' were raised both by its employees and by hired consultants. Despite internal knowledge of risks associated with ash ponds, TVA's formal Enterprise Risk Management process, which began in 1999, had not identified ash management as a risk. In 1987, an internal memorandum stated that, 'great amounts of ash have resulted in expansions of ash ponds. In some instances the dikes that contain this water have become quite high with increasing risk and consequences of a breech. Because of the potential for harm to both surface and groundwater from the failure dike, greater attention and establishment of more specific inspection standards for these dikes should be examined'. This memorandum triggered internal discussion about whether the ash ponds should have

Figure 6 Aerial view of before and after Kingston ash pond release (Jones and others, 2013)

Figure 7 Photo of then and now of Kingston ash pond release recovery (TVA, 2012)

been managed under TVA's dam safety programme, which would have required substantially more rigorous inspections and engineering. Ultimately, TVA did not place the ash ponds under its dam safety programme. Subsequent to the Kingston ash spill, TVA management has begun to change the way it handles coal ash.

A more recent similar, but smaller, ash disposal site collapse occurred near Milwaukee at the We Energies Oak Creek Power Plant in October 2011, this time spilling coal ash into Lake Michigan (Lemly and Skorupa, 2012).

In the wake of the Kingston release, the US EPA declared its intent to move forward with CCW regulations to address the management of coal combustion wastes (Luther, 2010a). Regulations vary for each type of surface disposal unit, and differ significantly from State to State in America. A common regulatory element is that all disposal units are required to have some type of permit to operate. With regard to surface impoundments, states commonly regulate two elements of a unit, the structure itself (the State's dam safety requirements) and any discharges from the unit to surface or groundwater (the State wastewater or water pollution control requirements). Many states are likely to require groundwater monitoring to detect contamination from a disposal unit but fewer states are likely to have regulatory requirements to prevent groundwater contamination from occurring, for example, by installing liners. New surface impoundments are likely regulated under provisions of the state's dam safety programme to address the structural integrity and under the terms of a wastewater discharge permit programme.

In China, a number of dam rupture cases were reported (Yang and others, 2010). In June 2006, a 30 m deep ash dam failed at the Panxian Power Plant when a retaining wall gave away near the bottom. It was estimated that 300,000 t of ash slurry poured directly into the Tuozhang River, which is part of the Pearl River. In 2009, a large surface impoundment dam at the Jiangdian coal-fired power plant failed, causing the ash pond to collapse. In August 2010, a 100 m section of the ash dam gave way at the Shentou No 2 power plant. Toxic coal ash sludge spilled out, submerging about 1.2 km^2 of fields. The coal ash disposal sites of Fengcheng No 1 and No 2 power plants are both located within the flood zone of Fengcheng city. In the event of a flood, it is very likely that coal ash could pollute a substantial area of farm and residential land. China is aware of the potential danger of ash disposal sites and has some regulations for ash management. For example, apart from requirement for liners, according to Regulation 5 in the 'Standards for Pollution Control on the Storage and Disposal Sites for General Industrial Solid Waste', sites for a coal ash-disposal facility 'should be consistent with the locality's overall planning requirements, be located downwind of industrial and residential areas, and be at least 500 metres away from the nearest residential area'.

5.4 Windblown

A large proportion of CCWs are fine grained material that can become airborne if allowed to dry and become exposed to windy conditions. Thus, ash disposal sites are prone to wind erosion and serve as a significant source for dust resuspension. At these facilities, there is a need to address dust mitigation concerns in order to comply with environmental permits, maintain operational controls, and minimise disturbance to adjacent areas and the public. Dust mitigation is facilitated after water has evaporated, that is, after the lowering of the water table in the case of wet disposal and after evaporation of the remaining moisture in the case of dry disposal. However, ash-inherent pozzolanic (such as self-hardening) properties reduce ash dispersion over time (Dellantonio and others, 2010).

Although extensive literature is available on the quality and quantity of stack released particulate matter, little has been published about the environmental effects of dust generated at CCWs disposal sites. Dellantonio and others (2010) summarised Gangl's study on particulate matter resuspension around a managed, continuously irrigated disposal site in Austria. The study measured annual average dust deposition of 100–140 $mg/m^2/d$ in the immediate vicinity of the landfill, which is well within the legal limits of 210 and 350 $mg/m^2/d$ in Austrian and German legislation, respectively. But particulate matter fluxes derived from unmanaged disposal sites remain largely unknown, although it is frequently stated that they bear the potential to affect land and water resources, as well as human health and the aesthetics of landscapes.

It has been speculated that air-borne ash would be a significant health issue around the site due to the large surface area of exposed ash. As part of the clean-up and monitoring efforts, several air monitoring stations are in place around the Kingston facility following the 2008 disaster. Data from these monitors can be found on the TVA website (*see* www.tva.com/kingston). Results to date show that of the more than 200,000 air samples analysed since the spill occurred, none has exceeded the US EPA National Ambient Air Quality Standards (NAAQS). Furthermore, the US EPA has stated that there is no known threat to public health from air pollution exposure associated with the clean-up of the ash spill at the TVA Kingston Fossil Plant. Air monitoring at the Kingston ash spill site continues to confirm that the national ambient air quality standards for inhalable particulates (PM_{10}) and respirable particulates ($PM_{2.5}$) are being met. In addition, health monitoring has found no health issues related to the ash release among residents living in the area around the site (EPA, 2010a; EPRI, 2010).

However, China has reported a different scenario. Similar to stack release fly ash, windblown ash from disposal sites have the potential to reduce visibility and to degrade house gardens surrounding CCWs management sites. On 5 May 1993 at 2 pm, an extreme sand storm hit the Hexi Corridor in Gansu Province, China. It lasted until 8 pm and 160,000 m^3 of coal ash was picked up by the wind, leaving a 'black storm' collective memory among the locals. The coal ash disposal site belongs to the Yongchang Power Plant. On 16 March 2010, the local government of Gansu province issued a yellow dust storm warning. The storm arrived first in Minqin and then covered the whole province. At about the same time, strong winds swept up dust from the Mongolian plateau across the northwest and north of China (*see* Figure 8). The dust storm affected an area of about 1.8 million km^2, home to about 270 million people. On 18 March, Beijing suffered its first dust storm of 2011. Zhang and others (2011) collected dust samples before, during and after the March 2010 storm. They found that during the sandstorms, the concentrations of arsenic, lead, selenium, sulphur and other pollutants commonly found in coal combustion by-products were significantly increased. The particles had been picked up by winds from the ash disposal sites, mixed into the sandstorm and deposited on cities, and caused a deterioration in the regional air quality.

Figure 8 A heavy dust storm approaching Golmud, Qinghai Province, China at 17.51 pm on 14 May 2010 (Zhang and others, 2011)

Inhalation of dust resuspended from unmanaged CCWs disposal sites poses a severe risk to human health due to the presence of respirable particulate matter and toxic trace elements in FA. Studies of the potential health effects associated with ash dust have largely focussed on power plant workers, for whom exposure to dusts is more common than for the general public. However, comments the US EPA received on its 2007 risk assessment raise concerns about fugitive dust. Since there was anecdotal evidence that fugitive dust was often emitted from CCWs management sites, the EPA decided to examine the potential for uncontrolled emissions from dry handling and whether it leads to significant human health risks. The SCREEN3 model was used to estimate the concentration of fugitive dust in the air near a CCW landfill. SCREEN3 is a screening level air dispersion model that estimates maximum short-term (one hour) air concentrations for point, area, or volume sources to estimate total suspended particulate fugitive emissions from an open landfill (modelled as an area source). As the name implies, SCREEN3 is a screening level model that incorporates many simplifying assumptions. Results show that there is not only a possibility, but a strong likelihood, that dry handling would lead to the NAAQS being exceeded without fugitive dust controls. Yearly and monthly controls were also found to have the potential to lead to significant risks. However, with this screen, it was uncertain whether weekly controls would have the potential to exceed NAAQS limits. Even the most conservative evaluation of daily dust controls showed particulate concentrations well below the NAAQS. Thus, without further, more precise evaluation, only daily controls can definitively be said not to cause excess levels of particulate in isolation (EPA, 2010c).

Apart from inhalation of particles, scattered by wind across the land, coal ash can cause soils to turn alkaline, which damages agricultural production and ecology. Spilled coal ash can seep into nearby fields, reducing yields or even killing crops (Yang and others, 2010).

Therefore, controlling windblown ash is an operational consideration at CCW management sites. At disposal sites, windblown ash is generally controlled by periodic wetting of open ash areas, and by covering inactive areas with bottom ash (as it is heavier than fly ash), soil, or vegetation.

5.5 Radioactivity

Coal contains naturally occurring radioactive constituents, such as uranium (U) and thorium (TH) and their decay products, including radium and radon (*see* Chapter 2). Uranium and thorium are each typically present in coal at concentrations of 1 to 4 mg/kg (EPRI, 2009). During coal combustion, most of the U, Th, and their decay products are released from the original coal matrix, and are distributed between the gas phase and solid combustion products. The partitioning between gas and solid is controlled by the volatility and chemistry of the individual elements. Virtually 100% of the Rn gas present in coal is transferred to the gas phase and lost to stack emissions. In contrast, less volatile elements, such as Th, U, and the majority of their decay products are almost entirely retained in the solid combustion wastes (Zielinski and Finkelman, 1997). Therefore, the concentration of most radioactive elements in CCWs will be approximately ten times the concentration in the original coal. Since Rn, ^{210}Pb, and ^{210}Po are highly volatile, FGD installations may also reduce the release of these radionuclides into the atmosphere, as has been observed for other volatile species such as Hg, As and Se. However, increased radioactivity of FGD products may be the consequence (Dellantonio and others, 2010) .

Although the radionuclides are enriched in the fly ash in comparison to the coal itself, the US Geological Survey determined that the average radionuclide concentrations in ash are within the range of concentrations found in other geologic materials, such as granite and shale (EPRI, 2009). Generally, radionuclide activities in CCW are in the upper range of natural background concentrations for granites and shales; however, regional variation is larger. Activity concentrations determined in CCW from various sources showed a broad variation within the ^{40}K, ^{238}U, and ^{232}Th series, which ranged from 0.1 to 1.24, 0.006 to 2, and 0.003 to 0.3 Bq/g, respectively (Dellantonio and others, 2010). For the 25-year period 1977-2002, the increase of ^{226}Ra, ^{232}Th, and ^{40}K content in FA compared to coal ranged from 2 to 4.5 times, but was still more than an order of magnitude lower than the threshold level given in the permit for its disposal. Scarce data for India's FA showed an increase in the radioactivity level for ^{226}Ra in the range 6.2–4.2 compared to coal and 5–2.6 compared to soil. It exceeded the corresponding concentrations in coal from >2 to >5 times. Concentrations of ^{228}Ra exceeded the corresponding concentrations in coal from 8.9 to 4.5 times and in soil from 7.5 to 2 times, and average value for soil from >2 to >3.5 times. All the concentrations were within safety limits (Twardowska and Szczepanska, 2004). Gooding and others (2006) analysed samples of pulverised coal ash from five UK coal-fired power plants for the naturally occurring radionuclides of ^{238}U, ^{232}Th and ^{40}K. The exhalation rate of ^{222}Ra was also tested. The results were within the limits of EU directives and guidance and the UK legislation. The authors concluded that there is unlikely to be a significant increase in the radiological hazard from the use of pulverised coal ash in building materials. McOrist and Brown (2009) collected a suite of 55 CCP samples across Australia and tested the concentration of naturally occurring radionuclides present in each sample. The study showed that none of the fly ash samples would be considered radioactive from a regulatory perspective, since no single radionuclide is above the limit of 1 Bq/g or 1000 Bq/kg for radionuclides of natural origin or above 10,000 Bq/kg for ^{40}K.

It has been shown that ash from the co-combustion of coal (70%) and biomass (peat 30%) contains 163 and 89 Bq/kg of ^{226}Ra and ^{232}Th respectively. For ash, produced in the co-combustion of coal (50%) and biomass (50%), these values are 123 and 68 Bq/kg. For ash, with higher amounts of biomass (90%), the concentrations of ^{226}Ra and ^{232}Th in the ash were 140 and 57 Bq/kg respectively. According to these values, it can be deduced that the addition of biomass to coal before combustion results in the ash showing a decrease in natural radioactivity of Ra and Th (Kalembkiewicz and Chmielarz, 2012).

In fly ash, uranium is more concentrated in the finer sized particles. If during coal combustion some uranium is concentrated on ash surfaces as a condensate, then this surface bound uranium is potentially more susceptible to leaching. However, no obvious evidence of surface enrichment of uranium has been found (Kalembkiewicz and Chmielarz, 2012).

Chemical conditions are likely to influence the leachability of uranium, barium (a chemical analogue for radium), and thorium from fly ash. For example, leachability of radioactive elements is critically influenced by the pH that results from the reaction of water with fly ash. Extremes of either acidity ($pH<4$) or alkalinity($pH>8$) can enhance solubility of radioactive elements. Acidic solutions attack a variety of the mineral phases in fly ash. However, neutralisation of acid solutions by subsequent reaction with natural rock or soil promotes precipitation or sorption of many dissolved elements including uranium, thorium, and many of their decay products. Highly alkaline solutions promote dissolution of the glassy components of fly ash that are an identified host of uranium. This can, in particular, increase uranium solubility as uranium-carbonate species. Fortunately, most leachates of fly ash are rich in dissolved sulphate, and this minimises the solubility of barium (and radium), which form highly insoluble sulphates (Zielinski and Finkelman, 1997).

5.6 Mercury

Increased control of mercury emissions from coal-fired power plants may change the amount and composition of CCPs. Such changes may increase the potential for release of mercury to the environment from either landfilling or the use of CCPs. Mercury volatilisation from CCWs in landfills and/or surface impoundments is expected to be small because of the low temperatures involved and the existence of a relatively small surface area per unit volume of residue. Since the residues are typically alkaline and the mercury is adsorbed on alkaline particles, acid leaching potential is expected to be minimal. Fly ash and FGD residues typically have higher mercury concentrations than bottom ash and boiler slag (Kilgroe and others, 2000). The condensation of mercury on fly ash particles as the water-soluble form $HgCl_2$ may pose a risk of contaminating groundwater. However, of particular note is the low or non-detectable concentrations of mercury leached by TCLP or water-based procedures for a large number of Australian and American fly ash samples (Izquierdo and Querol, 2012). Bhangare and others (2011) found that mercury in fly ash and bottom ash is minimal compared to the level in coal indicating that mercury escapes in flue gas. Research by the US EPA, EPRI and others has all shown that mercury is stable on fly ash at ambient temperatures, with very little potential for leaching or volatilisation (EPRI, 2009). The European Association for Coal and Lignite decided not to classify ash from coal and lignite power stations as a mercury waste source (Euracoal, 2013). After an intensive literature search, Izquierdo and Querol (2012) concluded that mercury leaching in coal fly ash is not a major environmental concern. There is a consistent lack of correlation between the total mercury concentration and leachable mercury. Mercury leaching appears to be controlled by adsorption from the aqueous phase with strong interaction between adsorbed mercury molecules. For FGD waste, tests performed by Babcock & Wilcox indicate that the form of mercury is normally stable and re-release to the environment is not expected. The extremely low concentration of mercury contained in the gypsum is insoluble, thermally stable and not expected to adversely impact its use for disposal in a landfill (Bielawski and others, 2001). Based on the limited data available, Kilgroe and others (2000) concluded that mercury levels in the leachate are non-detectable.

6 Leaching

The presence of hazardous components in waste does not, by itself, mean that they will contaminate the surrounding air, soil, groundwater, or surface water. Extensive testing has shown that coal ash rarely, if ever, exceeds hazardous waste criteria contained in the Toxicity Characteristic Leaching Procedure (TCLP) promulgated under the US Resource Conservation and Recovery Act (EPRI, 2009). According to Szczygielski (2012), there is no negative impact on the aquatic environment from coal ash landfills. In the vicinity of moist ash landfills, there tends to be a minor, local increase in mineralisation of groundwater and surface water. Impacts on the local aquatic environment are limited to the immediate vicinity of the given landfill. An increase in the content of trace elements and substances especially dangerous to the aquatic environment has not been found. However, the US EPA thinks differently. It was convinced that CCW could pose a risk to human health and the environment if it is not properly managed, and there is sufficient evidence that adequate controls may not be in place. Disposal or uses that involve direct applications of waste to the ground may allow hazardous constituents in the waste to leach from the material, migrate, and contaminate groundwater or surface water and, ultimately, living organisms (Luther, 2010b). The US EPA (2010a, b) conducted a risk assessment to investigate the matter and the results indicated that CCW disposal in unlined landfills and surface impoundments do present substantial risks to human health and the environment from releases of toxic constituents (particularly arsenic and selenium) into surface and groundwater.

Given the wide range of coal characteristics, such as rank, ash yield and chemical and mineralogical composition (inherited from the depositional setting), and different temperatures used in power stations and the different particulate control devices used worldwide, one might expect ash products from different countries to have very little in common. However, in an extensive literature review it is striking to see that some general trends in the leaching behaviour of elements do emerge. The leached concentrations, expressed in terms of absolute value, may certainly differ, but the leaching behaviour appears to follow relatively common patterns. This suggests that the factors that control the leaching of elements from fly ash do not differ significantly, regardless of the composition and characteristics of the ash. Understanding the reasons underlying the mobility of an element is a key question for better control over environmental impacts in the wide variety of utilisation and disposal practices. It may also assist in cost-effective design of management strategies and in the decision making of regulatory and governmental agencies (Izquierdo and Querol, 2012).

As mentioned above, the absolute leaching value varies greatly from place to place, reflecting the type of coal ash that is stored, its concentration and acidity, and the nature of the disposal site. As a result, the concentration of leachate differs from site to site. The rate of leaching may be affected by a number of factors (Gottlieb and others, 2010):

- the size of the disposal pond, pond depth, and the amount of pressure the waste creates;
- the underlying geology (the types of soil and rock that lie underneath);
- the gradient or slope of the land;
- how far an aquifer or underground stream might lie beneath the pond or bottom of the landfill.

This chapter summarises the leachate chemistry and assess the environmental impact of CCW disposal. Some prevention methods are also discussed.

6.1 Leaching chemistry

Leaching can be described as the fluid extraction of a compound or element from a solid. It is an interactive process in which both leachant and solid variables control the results. Leachate is typically defined as a liquid that has percolated through waste or drained from waste. Leachate by this definition includes both an infiltration component and runoff (contact water) component. Figure 9

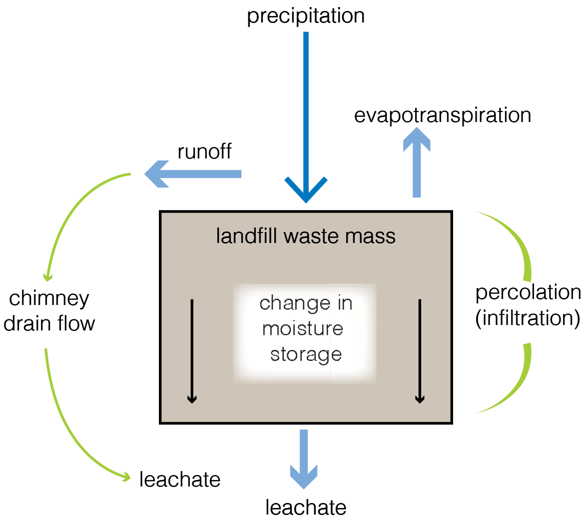

Figure 9 Leachate generation balance diagram (Baucom and Ruhl, 2013)

shows the balance diagram of leachate generation (Baucom and Ruhl, 2013).

Leaching of trace metals from CCWs is a slow process and can be complicated by secondary reactions, such as precipitation, adsorption, or the formation of complexes (Kim, 2002). Laboratory studies have demonstrated that the process is dependent on a number of factors, primarily chemical speciation of the compound, solution pH, and availability or release potential of the constituent for leaching. Availability for leaching depends on whether the element resides on the surface of the ash particle, in the outer glass hull, or within the interior glass matrix (EPRI, 2009). Weathering of ash releases metals and therefore also has an impact.

As Izquierdo and Querol (2012) summarised, a large number of elements are tightly bound to fly ash and may not be easily released to the environment, regardless of the nature of the ash. Also fly ash is a heterogeneous material as the elements are not evenly distributed throughout. The mode of occurrence of a given element in the parent coal was found to play an important role in the leaching behaviour of fly ash. The mineral fraction in coal undergoes different transformations during and after combustion, namely decomposition, volatilisation, fusion, agglomeration or condensation. As the flue gas cools down after combustion, volatile elements such as As, B, Hg, Cl, Cr, Se and most prominently S, condense on the surface of the fly ash particles, forming compounds with a variable solubility (commonly on the high side) and essentially combined with Ca. This results not only in a strong gradient of element concentrations within fly ash particles, but also in a gradient of leachability. The surface layer of fly ash particles, only microns in thickness, contains a significant amount of readily leachable elements that, with the exception of S, stand out for being inherently more toxic at low levels than major elements. Alongside the above listed elements, Cd, Cu, Mo, Sb, V and Zn are also preferentially enriched on the surface, whereas Ba, Co, Cr, Mn, Ni and Pb tend to be more evenly distributed between the surface and the matrix. The elements enriched in the core of the fly ash particles are not directly exposed to leaching, and therefore their releases are diffusion controlled and are also dependent on the dissolution rates of the surface layers. Surface-associated elements are therefore more susceptible to leaching in an aqueous environment.

As discussed in Chapter 2, cofiring with biomass changes ash chemical composition. Diversified types of biomass cause an increase (Al, Co, Cu, Fe, Ca, K, Mg, Mn, Na, Ni, Zn, Ag, As) or decrease (Ca, Cr, K) in the mobility of metals in relation to co-combustion fly ash (Kalembkiewicz and Chmielarz, 2012).

When fly ash interacts with water, the principal processes affecting the leaching behaviour are dissolution of primary solids and precipitation of secondary solids, as well as redox conditions, sorption and hydrolysis reactions, which are bound to control the mechanisms of mobilisation of toxic metals in the water. The concentration of metals in fly ash and the incorporation of the metals into secondary solid phases as a result of weathering reactions also determine the rates and amounts of metals released into the water column during leaching. The notable feature of ash leachate is the large variation in pH range, 4.5 to 12. The pH of fly ash is usually determined by the amounts of acidic and alkaline components in the soluble fraction of the fly ash. For example, a high pH of fly ash leachate is due to the dissolution and hydrolysis of alkaline and alkali metal oxides present on the surface of the fly ash (Lokeshappa and Dikshit, 2012). The amount of calcium in fly ash exerts a dominant influence on the pH of the ash-water system. The mobility of most elements contained in ash is markedly pH sensitive. The alkalinity of fly ash attenuates the release of a large number of elements of concern, such as Cd, Co, Cu, Hg, Ni, Pb, Sn or Zn among others, but at the same time, it enhances the release of oxy-anionic species such as As, B, Cr, Mo, Sb, Se, V and W. The precipitation of secondary phases, such as ettringite, may capture and bind several pollutants, such as As, B, Cr, Sb, Se and V. Ettringite requires Ca, Al, SO_4^{2-}, excess water and pH>11 to form (Izquierdo and Querol, 2012).

In addition, subsequent chemical interactions and secondary mineral formation can further modify leaching characteristics of the ash. For example, because arsenic typically condenses on the surface of the fly ash particle, it may initially be available for leaching. However, the presence of calcium in the ash can limit the release of dissolved arsenic by the formation of calcium-arsenic precipitates. Weathering and formation of iron hydroxide compounds can also serve to sequester arsenic (EPRI, 2009).

The leaching behaviour of fly ash in ash ponds appears to display three basic stages of leaching (Twardowska and Szczepanska, 2004):

- wash-out;
- dissolution;
- delayed release.

In the first two stages, the contamination potential of fly ash is governed predominantly by the release of macro-components at pH values ranging from alkaline to close to neutral. Hence the leaching of the major trace elements is low, except of oxy-anions of a wide stability field in solution (Mo, As, Cr(VI)).

6.2 Testing methods

Detailed leaching studies under controlled conditions are used to elucidate the mechanisms controlling constituent release, and these provide the best indication of the long-term potential for release and environmental risk. The tests are used as tools to estimate the release potential of constituents from fly ash over a range of possible waste management activities, including recycling or reuse, for assessing the efficacy of waste treatment and disposal processes.

There are three types of leaching methods, namely:

- regulatory – those promulgated and approved by a regulatory agency to generate specific information for submission in a legal context;
- standard methods – those adopted by a standards organisation (for example ASTM, ISO) for a specific set of conditions and sometimes for specific materials;
- research methods developed for a particular objective.

With exact duplication of regulatory or standard methods, there is a 60–80% probability that tests conducted under the same protocol by different laboratories will have comparable results. However, compliance tests and standard methods are not necessarily appropriate for leaching tests to simulate natural processes, obtain data on reaction mechanisms, or unravel complex solubility relationships. For these, it is necessary to select or develop a procedure that addresses the experimental objective, meets the procedural constraints, and produces data that are directly or functionally related to the problem being studied. Data obtained by different laboratories using standard or regulatory tests are probably comparable. Studies performed utilising different leaching methods do not necessarily produce comparable data, although researchers employing different methods may arrive at similar conclusions. But the results of the individual studies can be correlated sufficiently to discern general trends (Kim, 2002).

There are two distinctly different approaches to leaching tests: (i) attempting to mimic specific field conditions in laboratory testing and (ii) measuring intrinsic leaching characteristics such as liquid-solid partitioning as a function of pH and mass transfer rates and using appropriate mass transfer models to estimate leaching under field conditions (Kosson and others, 2002). A series of criteria must be met for a leaching test to be used to determine the potential for environmental impact of coal combustion ash and to stand up to legal and scientific scrutiny (Hassett and others, 2005):

- The test must take into account any reactivity or unusual properties of the material being leached.
- The test must as closely as possible utilise a leaching solution that mimics the solution most likely to contact the material in a natural disposal setting.
- If the ash is reactive with water, the test must take this into account by allowing the hydration reactions to occur during the course of the leaching test. This necessitates the use of long-term leaching (LTL).

According to Kim (2002), leaching methods are categorised as *static* or *dynamic* methods. *Static* (single addition) methods are divided into two categories:

- batch: a sample is placed in a given volume of leachant solution for a set period of time;
- monolithic: a particle of regular geometry and known surface area is immersed in a volume of leachant solution.

Dynamic (renewed) methods include:

- column: simulating the flow of percolating groundwater through a porous bed of granular material;
- serial batch: a sample of waste is leached successively with fresh aliquots of the same leaching fluid;
- sequential batch: a single sample that is leached by a series of different leaching fluids.

The most commonly used batch leaching methods are:

- Toxicity Characteristic Leaching Procedure (TCLP 1311);
- Extraction Procedure Toxicity Test (EPTOX);
- Synthetic Precipitation Leaching Procedure (SPLP);
- Standard Test method for Shake Extraction of Solid Waste with Water (ASTM D3987).

The dynamic serial batch method is intended to eliminate the effect of concentration on solubility and to simulate long-term exposure to the leachant solution. These methods include:

- EPA's Multiple Extraction Procedure (MEP);
- Standard Test Method for Sequential Batch Extraction of Waste with Acidic Extraction Fluid (ASTM D5284);
- Standard Test Method for Sequential Batch Extraction of Waste with Water (ASTM D 4793).

Among the variety of available leaching procedures, four types are commonly used in the literature to assess the leaching behaviour of coal ash (Hassett and others, 2005; Izquierdo and Querol, 2012):

- batch leaching tests using de-ionised water or similar as leachant (no pH buffering) to obtain information on the leachability of elements when the pH is governed by the ash itself;
- column/flow-through tests to provide data on the release pattern of elements under uncontrolled pH conditions;
- Toxicity Characteristic Leaching Procedure (TCLP), which was designed by the US EPA and mandated for use in determining the hazardousness of materials to be disposed of. The US EPA does not specifically recommend this test for coal combustion products, as it does not provide an accurate assessment of the material. However, the TCLP appears to be the worst-case test most often employed. The test is conducted using buffered acetic acid solutions with $pH=2.88$ or $pH=4.93$ depending on the pH of the material;
- acid extractions, performed under aggressive conditions ($pH<2$). These are difficult to relate to field conditions, but are aimed at gaining an in-depth knowledge of the ash chemistry.

In America, the most common methods employed to determine the environmentally 'hazardous' characteristics of solid waste materials for RCRA purposes are TCLP and SPLP (Kim, 2002). Kim and Hesbach (2009) at the US National Energy Technology Laboratory (NETL) of the US Department of Energy compared five leaching methods. Four of the procedures were developed specifically to determine the potential environmental effects of fly ash use and disposal; the fifth, toxicity characteristic leaching protocol (TCLP), is a standard toxicity measure. The procedures selected were the serial batch leaching procedure (SBLP) developed at NETL, the synthetic groundwater leaching procedure (SGLP) from the Energy and Environmental Research Center at the University of North Dakota, the mine water leaching procedure (MWLP) by the West Virginia Water Resources Institute, the 3TIER integrated framework leaching protocol (3TIER) by Kosson and others at Vanderbilt University, and the TCLP from the US EPA. The study indicated that several of the leaching procedures produce comparable data, but differ in the time and analytical resources required. Although none requires prohibitively expensive equipment or unusual reagents, the time required varies from 1 (TCLP) to 60 (SGLP) days. The number of leachate samples to be analysed per replicate varies from 1 (TCLP) to 20 (3TIER) or more (MWLP). The differences in resources required may influence the choice of leaching procedure, but type of data desired and how closely the procedure approximates the intended usage should be the primary factors in the selection of a leaching method.

Nevertheless, the above tests, when applied to CCWs, have not always considered field conditions known to influence leaching. Furthermore, assessments have sometimes considered initial but not final site conditions, although changes at a site over time could change the propensity of a material to leach. Therefore, the US EPA initiated the review and validation process of a new generation of leaching tests, namely the Leaching Environmental Assessment (LEAF) methods – EPA/600/R-10/170 (published in Nov 2010) (*see* Table 9). The LEAF test methods are a suite of four leaching tests which include three batch tests and one dynamic test. These four methods are the next generation of leaching methods and are a considerable shift from the current EPA SW-846 approach of TCLP and SPLP (McIsaac and others, 2012). As of October 2012, two of these methods (method 1313 and 1316) have been included in the new methods section of US EPA's Method Compendium-Test Methods for Evaluating Solid Waste, Physical/Chemical Methods, commonly referred as SW-846. Inter-laboratory validation testing of methods 1314 and 1315 have been completed and the results are being evaluated in anticipation of inclusion in SW-846. One of the main drawbacks of using LEAF is the large volume of data provided by the tests, and the potential of the results being improperly used. Additional guidance and research is needed before they can be used to make and/or influence site specific decisions about leaching (Hattaway and others, 2013).

Nelson and Morrison (2008) reviewed the status of leaching tests and concluded that the TCLP was limited in its usefulness as a regulatory tool in Australia. Previous experimental and modelling work in the Black Coal CRC and CCSD had demonstrated that no single leaching test is totally appropriate, and that a range of tests, together with hydro-geochemical modelling, is an effective way to assess trace metal release under different PH conditions.

Lokeshappa and Dikshit (2012) used three synthetic ponds fabricated in perspex sheets to test fly ash leaching potential over the course of eleven months. The results show that water discharged from silica-rich coal fly ash ponds could contribute to leaching of As and Cr to surface water and groundwater in addition to contaminating the nearby soils. Though the concentration of Se, Zn, Pb and Ba leached from Ca-rich ashes in the ash pond reduced with time, it was still above 150 ppb in most of the cases. Therefore, the practice of fly ash disposal in unlined ash ponds could be considered as environmentally unsafe.

TCLP-1311 and ASTM D3987 are commonly used methods for batch tests in Turkey. Akar and others (2012) carried out a short-term leaching behaviour test for Fe, Ca, Cu, Co, Cd, Mn, Ni, Pb, Zn and Cr in ash from Yenikoy coal-fired power plant.

Table 9 US EPA’s leaching test methods (McIsaac and others, 2012)

Leaching test	Reference documents	Summary description	Comments
Toxicity Characteristic Leachate Procedure [TCLP]	US EPA SW 846 Method 1311	Samples are preliminarily evaluated for solids and particle size. The liquid to solid ratio is 20:1. The sample is then leached with appropriate fluid. A pH 2.9 acetic acid is used for moderate to high alkaline material and pH 4.9 acetate buffer is used for all other materials. The total time for the leachate generation is 18 hours.	This is a single point leachate test. Predicts the mobility of both organic and inorganic analytes in landfills. It is used to classify material as hazardous or non-hazardous for purposes of disposal in a landfill.
Synthetic Precipitation Leachate Procedure [SPLP]	US EPA SW 846 Method 1312	Samples are preliminarily evaluated for solids and particle size. The liquid to solid ratio is 20:1 and the samples are then leached with appropriate fluid. The extraction fluid is based on the region of the country where the sample is located. For samples east of the Mississippi River, the extraction fluid pH is 4.2, and for materials west, the pH is 5.0. The total time for the leachate generation is 18 hours.	This is a single point leachate test. Predicts the mobility of both organic and inorganic analytes into ground and surface waters. SPLP fluid simulates precipitation.
Liquid-Solid Partitioning as a Function of Extract pH for Constituents in Solid Materials using a Parallel Batch Extraction Procedure	US EPA Background Information for the Leaching Environmental Assessment Framework [LEAF] Test Methods EPA/600/R-10/170 LEAF or pre EPA Method 1313	This is a pH dependent batch leaching procedure. Ten parallel extractions of a particle sized reduced solid material in dilute acid or base and reagent water. Series of eluate having pH values ranging from 2-13 as well as natural condition. Liquid solid ratio is 10:1. Eluate is centrifuged and filtered; then analysed for constituents of concern. Total time to generate the eluate is five days for material with 85% greater solids; 8 days for material with less than 85% solids.	This method is designed to provide aqueous extracts representing the liquid-solid partitioning [LSP] curve as a function of pH for inorganic and non volatile organic in solid materials
Liquid-Solid Partitioning as a Function of Liquid-Solid Ration for Constituents in Solid Materials using an Up-Flow Percolation Column Procedure	US EPA Background Information for the Leaching Environmental Assessment Framework [LEAF] Test Methods EPA/600/R-10/170 LEAF or pre EPA Method 1314	This is a dynamic leaching procedure. Eluate is introduced into a column with packed particle sized reduced solid material in an up-flow pumping mode. Flow rate is maintained at 0.5–1.0 L/day. Eluant is collected at predetermined times, filtered and analysed for constituents of concern. Total time to generate the eluate is approximately 14 days.	This method is designed to provide the liquid-solid portioning [LSP] of inorganic constituents and nonvolatile organic in granular solid material as a function of liquid to solid [L/S] ratio under percolation conditions.
Mass Transfer Rates of Constituents in Monolithic or Compacted Granular Materials using a Semi-Dynamic Tank Leaching Procedure	US EPA Background Information for the Leaching Environmental Assessment Framework [LEAF] Test Methods EPA/600/R-10 /170 LEAF or pre EPA Method 1315	This is a hybrid batch and dynamic leaching procedure. Leaching of continuously water saturated monolithic or compacted granular material in an eluant-filled tank with periodic renewal of the leaching solution. LS ratio of 9 ml eluant per cm^2 of surface area. Eluant is collected at predetermined times and analysed for constituents of concern. Eluate is centrifuged and filtered for constituents of concern. Total time to generate the eluate is approximately 63 days.	This method is designed to provide the mass transfer [release rates] of inorganic analytes contained in a monolith or compacted granular material. Under diffusion controlled release conditions, as a function of leaching time.
Liquid-Solid Partitioning as a Function of Liquid-Solid Ratio for Constituents in Solid Materials using a Parallel Batch Extraction Procedure	US EPA Background Information for the Leaching Environmental Assessment Framework [LEAF] Test Methods EPA/600/R-10/170 LEAF or pre EPA Method 1316	Five parallel extractions of a particle size reduced solid material in reagent water over a range of L/S values from 0.5 to 10 ml eluant/g dry material. Depending on particle size, sample is tumbled between 24 and 72 hours. Eluate is centrifuged and filtered for constituents of concern. Total time to generate the eluate of test is two days.	This method is designed to provide the liquid-solid partitioning [LSP] of inorganic and non volatile organic at the natural pH of the solid material as a function of liquid to solid ratio [L/S] under conditions that approach liquid-solid chemical equilibrium.

6.3 Field studies

While laboratory studies are used to define long-term leaching mechanisms, field studies provide the best information on leachate quality under actual environmental conditions. In 2006, EPRI, with support from the US DOE, completed an extensive characterisation of field leachates at over 30 coal combustion product disposal facilities (Hensel and others, 2006). This study provides the most comprehensive database available for ash leachate characteristics representative of typical environmental conditions at disposal sites (*see* Figure 10). The data in the figure represent initial concentrations in the management facility, not the concentration to which the public is exposed.

Since 2000, the US EPA have been identifying the 'damaged cases' caused by CCWs. In 2007, an assessment identified 67 CCW management sites with groundwater or surface water impacts that were categorised as 'proven'(24) or 'potential'(43) damage cases (EPA, 2007a). The location of the 67 damage cases identified by US EPA are shown in Figure 11. The US EPRI carried out a further evaluation of these cases, including additional data obtained from power companies and public sources. Their review indicates that the damage cases are largely older, unlined facilities that began operation prior to enactment of current disposal regulations. The constituents identified most often were sulphate and boron, and impacts were generally contained on-site. Although results suggest that unlined sites can be a source of groundwater impacts, such sites are not representative of modern non-hazardous waste disposal designs that include liners and leachate collection systems (Hensel, 2010a,b).

However, scientists at the Environmental Integrity Project (EIP) Earthjustice (2010) think that EPRI and some state agencies are not strict enough with coal ash. They have identified 31 new coal combustion waste sites that have contaminated groundwater, wetlands, creeks, or rivers in 14 states. At 15 of the 31 sites, contamination has already migrated off the power plant property (off-site) at levels that exceed drinking water or surface water quality standards. The remaining 16 show evidence of severe on-site pollution. As off-site monitoring data at 14 of these 16 sites were not available, damage may be more severe and widespread than indicated in their report. The damaged sites include

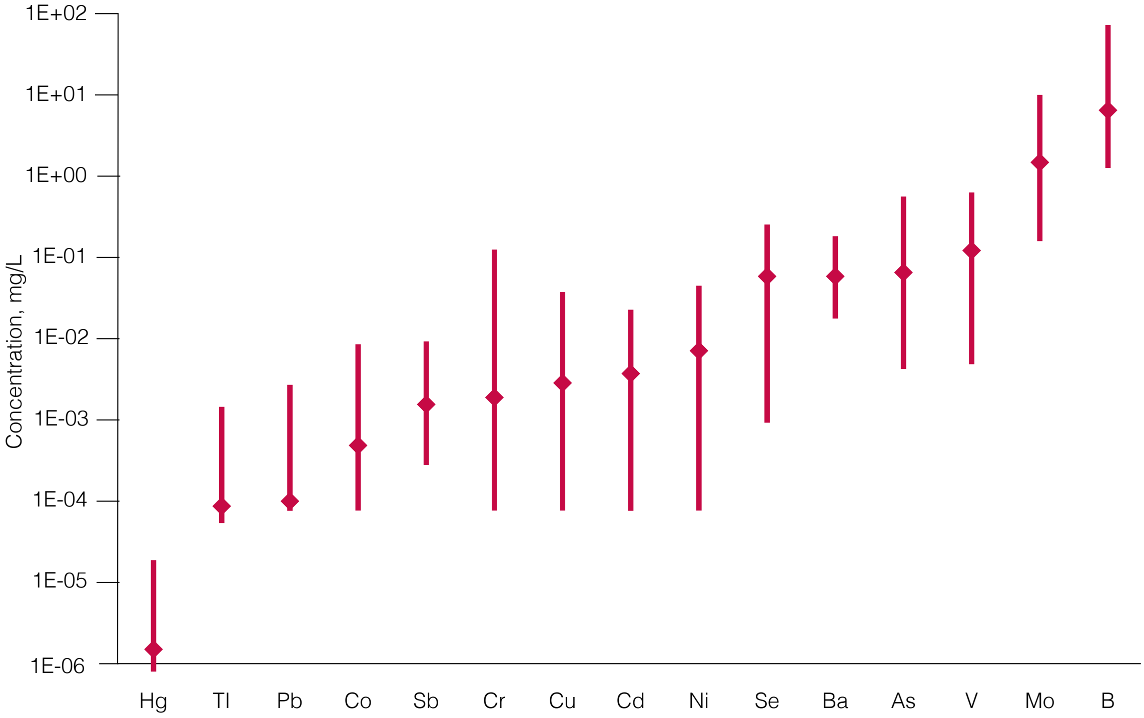

Figure 10 Field leachate concentrations for coal ash disposal sites (EPRI, 2009)

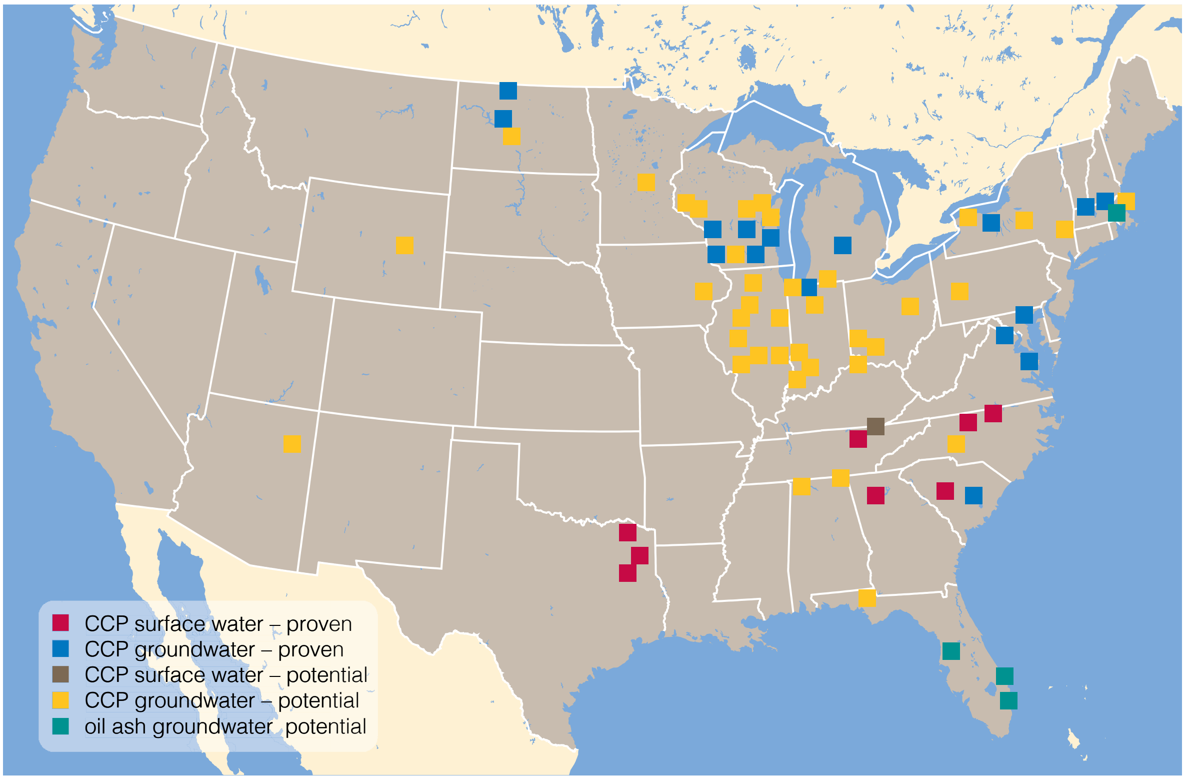

Figure 11 Location of 67 damage cases sites in USA (Hensel, 2010b)

several high hazard ash ponds. This report brings the total number of damaged sites in the USA to more than 100, with still more to be investigated. Monitoring data for eight of the disposal facilities identified in this report show significant pollution has migrated off-site into the underlying aquifer in excess of maximum contaminant levels (MCLs). At the following coal ash dump sites contaminated groundwater has travelled beyond site boundaries: Big Bend Station (Florida), Gibson Power Plant (Indiana), Karn and Weadock Generating Facility (Michigan), Colstrip Power Plant (Montana), Swift Creek Landfill (North Carolina), Reid Gardner Generating Facility (Nevada), Orion's Phillips Power Plant (Pennsylvania), and Trans Ash Inc's CCW Landfill (Tennessee). Several of the other sites with limited data may also have polluted groundwater moving off-site. At least 26 of these sites report on-site groundwater contamination that exceeds one or more primary drinking water criteria. Concentrations of toxic pollutants at many of these sites are high.

In December 2011, EIP identified an additional 19 sites where coal combustion waste appears to have contaminated groundwater with arsenic or other pollutants at levels above primary Safe Drinking Water Act MCL. All but two have also measured concentrations of other pollutants (such as boron, molybdenum, and manganese) above the limits recommended by the US EPA in its Health Advisories for children or adults. In addition, the report included new information about seven previously recognised damage cases, including evidence of groundwater more toxic than hazardous waste leachate (Boulding and others, 2011). Posted in June 2013, but as of 18 September 2012, the US EPA (2012) confirmed 18 more CCW disposal sites which have polluted groundwater or surface water at concentrations that threaten human health and the environment. Fifteen of the 18 polluted sites were identified by the Environmental Integrity Project, Earthjustice (2010, 2013). Moreover, the EPA's new list does not address the 19 damage cases identified in EIP's December 2011 report (Boulding and others, 2011).

A joint report by Sierra Club, Kentucky Wasterways Alliance, and Global Environmental, LLC

(Quarles and Segall, 2010) agrees with Environmental Integrity Project Earthjustice (2010). They investigated all 44 ponds in Kentucky and were able to obtain information on only eight sites. Based on the investigation on the eight sites, their conclusion was that CCW ponds are clearly leaking into the groundwater beneath them, as every pond for which data were available was associated with contamination.

Although not investigated as comprehensively as in the USA, other countries, such as India and China, have also made considerable efforts in reporting CCWs leaching impacts on the environment. Some examples are discussed below.

In early 2013, news emerged of groundwater pollution in a small village, Ganyugou, China. Ganyugou is the nearest village to the Lijiayu ash pond which belongs to the Tangshan Douhe power plant. The power plant has eight units with a total of capacity of 1550 MWe. The first units began operation in 1976 and the last unit went online in 1986. Typically, the plant produces 0.6 Mt of fly ash a year. Lijiayu ash pond has a capacity of 68 million m^3 without liners. The nitrate concentration in its under-groundwater was 36% higher than the standards for drinking water (South Reviews, 2013). Greenpeace had previously found water pollution from coal ash disposal (Yang and others, 2010). They collected samples from under-groundwater wells near the ash disposal sites of eight power stations. The samples were tested for the presence of heavy metals and other harmful pollutants (*see* Table 10). Of these samples, three contained concentrations of pollutants that exceeded levels set by the ‘Sanitary Standards for Drinking Water’. At the Douhe Power Plant, the concentration of nitrates (27.12 mg/L) was 36% over the maximum; at Chifeng Thermal Power Plant, boron (898 µg/L) was found in concentrations 80% over the maximum; and at Yuanbaoshan Power Plant, boron concentrations (1850 µg/L) exceeded the maximum by 270%, molybdenum concentrations (142 µg/L) by 103%, nitrate concentrations (34.72 µg/L) by 74%, and fluoride concentrations (2.8 mg/L) by 180%.

Choi and others (2002) investigated the leaching behaviour of disposed fly ashes in South Korea and assessed the potential influence of the fly ash landfill on the groundwater composition. The methods adopted in the study included batch leaching tests in the laboratory and the analysis of field samples, including surface water, groundwater and fly ash slurry collected from the fly ash lagoon at Yongdong coal-fired power plant. The Yongdong ash lagoon is lined with bottom ash, underlaid by a silt and sand layer which contains some marine clays. Such lining systems are believed to be permeable to some extent and the leachate from the lagoon might be migrating into the groundwater below. The ash lagoon in YD power plant has been in operation for some 20 years and the presence of measurable amounts of heavy metals in water samples could be attributable to the dissolution of fly ash in the lagoon.

A surface pond for the disposal of coal ash slurry from the Maharashtra State Electricity Board (MSEB) at Chandrapur was sited in the Erai River basin, India in 1983-84 . No specific lining had been provided to insulate the pond from the aquifer. A survey (Twardowska and Szczepanska, 2004) showed significant adverse changes of water quality in the shallowest unprotected alluvial aquifer within and down-gradient of the pond. This was revealed mainly by an increase of macro-constituent concentrations typical of the leachate from the power plant ash (TDS, chloride, sulphate, Ca, Mg). Increased concentration of trace elements (B, Cd, Cr(VI), Pb, Hg), fluoride and nitrate were also found compared to the background concentrations. Contamination of the deeper aquifer has not yet been observed. However, in the long run, the hydraulic connection of aquifers poses a serious threat due to downward development of contamination.

Ramya and others (2013) assessed the impact of ash ponds on groundwater quality in the sub-watershed surrounding the ash ponds in the vicinity of Koradi near Nagpur in Maharashtra, India. The ponds do not have liners. The analysis indicated that the sulphate concentration is very high (>1,000 mg/L) in samples close to the ash pond and in its downstream direction. The fluoride concentration exceeded the Bureau of India Standards limits in one sample.

Table 10 Heavy metals and other compounds found in water samples collected near coal ash disposal sites in China (Yang and others, 2010)

Power station	Sample location	Heavy metals and compounds detected
Douhe	Lijiayu coal ash disposal site (surface water)	Barium, boron, iron, molybdenum, titanium, vanadium, zinc, nitrates, chlorides, fluorides
	Ganyugou village (well water)	Barium, boron, zinc, nitrates, chlorides, fluorides
Chifeng Thermal	Dongjiao Badui coal ash disposal site (surface water)	Barium, boron, manganese, molybdenum, zinc, nitrates, chlorides, fluorides
	Dongjiao Badui village (well water)	Barium, boron, molybdenum, zinc, nitrates, chlorides, fluorides
Fengzhen	Fengzhen ash disposal site (surface water)	Aluminium, barium, boron, cadmium, calcium, chromium, copper, iron, lithium, magnesium, nickel, potassium, sodium, strontium, zinc, sulphates
	Jiuquan village (well water)	Aluminium, barium, boron, calcium, chromium, iron, magnesium, mercury, molybdenum, potassium, sodium, strontium, zinc, sulphates
Yuanbaoshan	Xinglongpo village (well water)	Barium, boron, molybdenum, zinc, nitrates, chlorides, fluorides
Shentou No 2	Shuimotou village (well water)	Barium, calcium, chromium, copper, magnesium, potassium, sodium, strontium, zinc, sulphates
Datong No 2	Dangliu village ash disposal site (surface water)	Aluminium, barium, boron, cadmium, calcium, chromium, copper, lithium, magnesium, nickel, potassium, sodium, strontium, zinc, sulphates
Xuanwei	Miaohou village (well water)	Barium, calcium, magnesium, potassium, sodium, strontium, sulphates
Jiujiang	Phase 2 Weijiachong coal ash disposal site (surface water)	Aluminium, barium, boron, calcium, chromium, copper, iron, magnesium, manganese, nickel, potassium, sodium, strontium, zinc, sulphates
	Yujiahe village (well water)	Aluminium, barium, calcium, magnesium, potassium, sodium, strontium, zinc, sulphates

The mobility patterns and mineralogical associations of major elements in weathered dry disposed ash dumps aged 1, 8, and 20 years old from a coal-fired power station in South Africa were investigated by Akinyemi and others (2011) using a modified sequential extraction scheme. The results show a clear mobility of elements, which is influenced by the heterogeneity of the ash dump, inhomogeneous continuous brine irrigation, and chemical interaction of ash cores with ingressed CO_2 from the atmosphere and percolating rain water.

6.4 Technologies used to prevent leaching

While new CCWs facilities are likely to be built with liners and groundwater monitoring systems, the majority of facilities currently in use are not new. For example, in the USA, EPA determined that 75% of surface impoundments in use today are more than 25 years old with 10% being more than 50 years old. EPA estimated that with an average life expectancy of approximately 31 years, those older disposal sites would likely continue to operate without necessary protections in place well into the future (Luther, 2012). These old units that lacked liners and groundwater monitoring therefore posed a potential risk to the environment. US EPA (2010a) reported that for landfills in the USA, only 57% of the units had groundwater monitoring, while for surface impoundments, only 26% of the units had liners and 38% of the units had groundwater monitoring. EPRI (Hensel, 2010a) assessed 63 damage case facilities and found that only six of these were built with liners. Ten other lined facilities were either retrofits or partially lined. The rest were unlined. It is clear that what most determines the amount of leaching is not the coal but the robustness of the storage site.

Leachate runoff and infiltration to groundwater can be controlled by a variety of standard engineering practices employed at disposal facilities. Depending on site-specific conditions, these practices may include the use of liners, leachate collection systems, diversion ditches, caps, and vegetation. Monitoring networks are used to ensure the performance of the engineering controls in protecting groundwater and surface water resources.

Summarised from varies publications (Elcock and Ranek, 2006; EPA, 1988, 1999, 2010a; Gottlieb and others, 2010, Hardin, 2011), the technologies used to reduce the potential environmental risk will be discussed in the following sections:
- liner system;
- leachate collection systems;
- groundwater monitoring systems;
- pre-treatment technologies, by altering physical and chemical properties.

6.4.1 Liner system

Most countries require the installation of a liner system for new ash disposal sites. Liners are being retrofitted at old sites, where the CCWs are removed then a liner is added and ash management resumes. This method is often used at impoundments. Some old units are partially lined where the original landfill or impoundment cells were unlined with lined cells added later.

A liner is a barrier placed underneath a landfill or on the bottom and/or sides of a surface impoundment. Depending on their construction, liners can slow or prevent the release of the leachate from a landfill or liquids from a surface impoundment to underlying soils and groundwater. Liners can consist of compacted soil, compacted clay, a synthetic material or membrane, or a combination of barrier types. In general, single liners consist of one type of liner, composite liners consist of a membrane combined with a clay liner, and double liners consist either of two single liners, two composite liners, or a single and a composite liner. Some units employ multiple types of liners. For example, a single unit with double, synthetic, and compacted clay liners would be reported as having multiple types.

Clay/soil liner

The installation of a clay liner in a surface impoundment or landfill entails several steps:
- First, the site must be excavated or graded to a level below the design elevation of the facility floor. Many facilities take advantage of natural low areas or abandoned ponds to minimise excavation costs. The excavated earth can be used to build up the dyke walls for the impoundment or to build containing berms within the landfill. Occasionally, soil must be brought to the construction site to raise the dykes to the design height.
- Once the floor and dykes or berms have been prepared, the clay liner is laid in 15 to 23 cm lifts; its final thickness will be between 0.3 m and 2.4 m. Each lift is individually compacted before the next one is laid, thereby providing effective compaction and minimising leakage potential. Field testing of the clay for permeability and other pertinent characteristics is sometimes performed during construction to provide quality assurance. Before the impoundment or landfill can be used, the liner is visually inspected for flaws; non-contaminated water may also be piped to the pond to assure that the liner is sufficiently impermeable.

One of the primary concerns about the use of clay liners is whether the entire clay liner meets thickness and permeability requirements. If weather conditions during liner construction are arid and hot, the liner may dry out and crack, causing localised areas of leakage. If conditions are wet or the clay is too moist, clay compaction may never be sufficient to achieve the necessary low permeability. The clays used as liner materials vary in the degree to which they are compatible with the wastes placed in the facility. Laboratory tests, in which the proposed liner material is exposed to the wastes intended for management, should be conducted for each facility to ensure that components of the waste material will not unduly alter the permeability of the clay used as liner material. If the chemical characteristics of the generated waste were to change over time, then the tests would need to be repeated to determine what effect the altered waste stream would have on the clay liner.

An advantage of clay liners is their potential for chemical, particularly cation, attenuation. The chemical structure of clay allows its use as an exchange site for metallic cations and other ions that might gradually seep out of the facility. Such exchange further reduces the opportunities for migration of waste constituents to groundwater. For facilities with fairly ready access to clays, the capital and construction costs associated with the use of these liners, even one that is several feet thick, may be substantially lower than those associated with the use of a synthetic liner.

The US EPA has reservations on the reliability of clay liners, especially in surface impoundment. They (EPA, 2010b) found that leaching risks are lower from clay-lined than unlined units when using landfill to dispose CCWs or CCWs with coal refuse. However, risks of leaks for arsenic and thallium from clay-lined landfills still exceed the risk criteria for excess cancer risks adopted by EPA. Risks of arsenic, boron, and molybdenum leaking from impoundments are also above the criteria. Quarles and Segall (2011) also believe that compacted clay liners are ineffective at preventing migration from dry waste disposal in landfills.

Synthetic liners

The most common materials of construction for these liners include polyvinyl chloride (PVC) and high-density polyethylene (HDPE), although several other impermeable synthetics have also been used. Liners may be reinforced with fibres to increase strength and decrease the likelihood of punctures. Most liner installations will include protective geotextile fabric above and/or below the impermeable synthetic liner to minimise further the potential for puncture.

Preparation of a site prior to installation of a synthetic liner is similar to that which occurs before clay liner construction. However, more care must be taken to smooth out the surfaces to eliminate any peaks and cavities on the disposal facility floor that could cause a puncture of the liner material. Consequently, surface preparation costs are greater than those for clay liners. Excavation costs are usually less, however, because the thinner synthetic liners allow shallower excavation. For example, the additional excavation required to install a clay liner that is 2 m thick can be avoided if a much thinner synthetic liner is installed.

The liner itself, which comes rolled or folded in large pieces, is laid in the field and sealed along the seams by heat or solvent fusion techniques. The seams may be field tested at spot checkpoints. The liner is usually covered with 30 cm or more of soil to protect it from puncture and to keep it in place during construction of the disposal facility. The edges of the liner at the tops of the dykes or landfill cell walls must be well secured to prevent the liner from pulling out and shifting due to the mass of the wastes placed in the impoundment or landfill. Some facilities are double lined and often contain a leachate collection system located in a soil or sand layer between the two liners.

Among the limitations to the use of synthetic liners is their susceptibility to tear and puncture. This is of particular concern in a single-lined impoundment because of the opportunity for liquids to seep through a single tear. Synthetic liners are also susceptible to degradation by certain waste materials. Acidic wastes, for example, can degrade some synthetic liner materials. As with clay liners, waste/liner compatibility testing should be performed to ensure that the disposed wastes will not weaken or permeate the liner. Additionally, because the seams of a synthetic liner are frequently weaker than the liner itself, they may pull apart under stress, such as large mass loadings or wave action. Finally, dredging synthetically-lined impoundments must be done cautiously, sometimes at very significant expense.

Synthetic liners, unlike clay liners, are impermeable. Another advantage is the ease of repairing an exposed, damaged impoundment liner. A tear or puncture can be patched and seamed, and an impoundment put back into service, relatively quickly. To repair subsurface damage, however, the impoundment must be wholly or partially drained. Another advantage of using synthetic liners is that because of manufacturer quality control, a facility owner can be fairly certain that each liner sheet is as impermeable as the next. Clay is expensive to transport and in areas of the country where clay soils are scarce, a synthetic liner system may prove to be the less expensive option.

Composite liners

Many waste management facilities in industries are installing liner systems that combine both clay liner and synthetic liner technologies. The majority of the new facilities built after 1995 use composite liners. Most commonly, an impoundment or landfill will be lined with 0.6–1.2 m of impermeable clay, which is then prepared for placement of a synthetic liner (*see* Figure 12). The synthetic liner may be covered with 0.3–0.6 m of sand to serve as drainage for a leak detection system. Some facilities may then add another 0.3–0.6 m layer of clay, which is again prepared for placement of the upper synthetic liner. In landfills, another leachate collection system is usually placed above this upper liner.

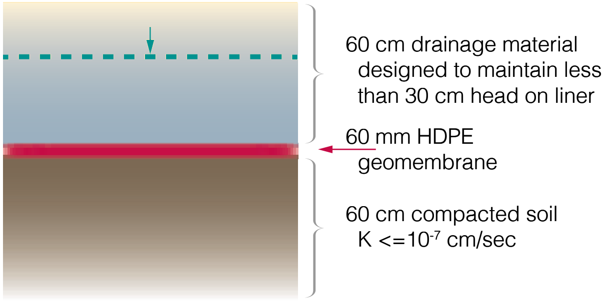

Figure 12 Single composite liner (Hardin, 2011)

The composite synthetic/clay liner system offers a combination of advantages over single-material liners. A composite liner has some of the advantages provided by synthetic liners, such as factory quality control and ease of repair (for the upper liner), as well as the advantage of clay's propensity for attenuating escaped ions. Furthermore, use of multiple-liner materials reduces the likelihood that waste material will leak into the groundwater because of chemical incompatibility between a waste and a single liner material. In general, the more layers of impermeable liner material that are used, the more efficient the containment of liquids will be, thus reducing the likelihood of a release of waste material.

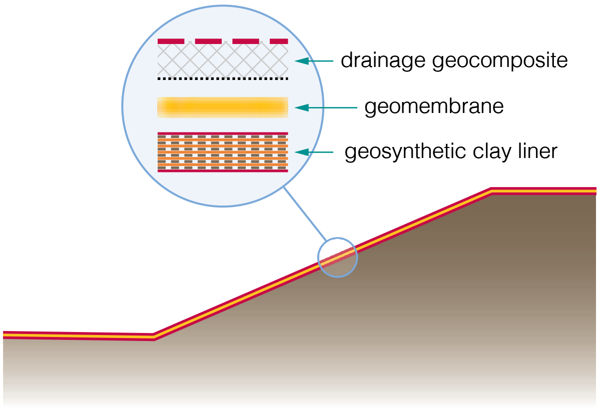

Figure 13 Conceptual layout of a single composite liner system (Zimmel and others, 2013)

The biggest drawback of the composite synthetic/clay liner system is the cost of installation. Utility waste landfills are very large (up to 0.4 km^2 or more), and a liner large enough to cover such a area can be very expensive. In areas where labour costs are high, and clay is unavailable locally and must be transported long distances, these costs would be magnified.

In recent years, composite liner systems have been innovated to improve performance. Zimmel and others (2013) introduced a geo-membrane with a geo-synthetic drainage layer and geo-synthetic clay liner composite liner system (*see* Figure 13). Tests show that the system meets the design requirements for CCW disposal sites.

6.4.2 Leachate collection and groundwater monitoring

It is a requirement in most countries to have a leachate collection system at lined waste management facilities and any facility (lined or unlined) may be equipped with a groundwater monitoring system. A leachate collection system is a series of drains placed beneath a disposal unit. Leachate collection systems are used to prevent the migration of contamination from a landfill or impoundment. Both systems can be used to monitor the rate and extent of contaminant migration. Groundwater monitoring systems consist of one or several wells drilled in the vicinity of a disposal unit. Samples from these wells are periodically collected and analysed. In China, it is suggested to establish at least three water quality monitoring wells near a coal ash storage and handling facilities (Yang and other, 2010). Groundwater monitoring is not strictly an environmental control but rather a warning system. Groundwater samples that display contamination may trigger regulatory requirements to mitigate or eliminate the source of contamination. The design and placement of groundwater monitoring and leachate collection systems should take into account the manner in which a landfill or impoundment might potentially interfere with natural groundwater flow and usage patterns.

In surface impoundments, the leachate collection system can be placed below the entire liner system or it can be placed between any two liners. It typically consists of a drainage media (coarse sand and/or gravel) and perforated pipes (called riser pipes) that slope toward a collection sump. The collected leachate is pumped out via these riser pipes to the surface for treatment and/or disposal. If the riser pipes through which the leachate is pumped perforate the synthetic or clay liner, tight seals are necessary to ensure that the leachate does not escape through the perforation.

In landfills, leachate control systems can be installed below all liners (this is usually called a pressure relief system), between liners (the inter-liner leachate control system), and/or above the upper liner. The floors of a landfill cell are designed to slope to the leachate collection sumps, and are usually covered with a drainage media such as sand or gravel. Each leachate control system has its own collection sump, which is emptied through riser pipes so that the leachate can be treated or disposed of appropriately. As with impoundment liner systems, riser pipes, if they pierce the liners, must be sealed to prevent leakage.

Groundwater monitoring wells are placed at strategic locations to facilitate early detection of any contaminants that escape the facility and migrate to the groundwater. The design and placement of the monitoring wells is based on site-specific hydro-geological assessments, soil chemistry, specific regulatory directives, and other physical and chemical factors. Typically down gradient wells are used to monitor the extent of contamination arising from a facility, and up gradient 'background' wells are installed to serve as controls.

6.4.3 Pre-treatment

Facilities employ a variety of waste treatment processes to alter the physical or chemical characteristics of wastes so that they should be compatible with the disposal method used. Treatment methods may also be employed to comply with regulated effluent limitations. The evaluation of leachate treatment options is driven by costs, which in turn might be driven by estimated leachate generation rates. For instance, higher leachate generation rates might justify constructing an on-site wastewater treatment facility, whilst lower leachate generation rates might justify constructing leachate storage tanks and off-site leachate treatment (Baucom and Ruhl, 2013) .
Generally, the pre-treatment methods include:

- sludge de-watering;
- reagent addition;
- low-volume waste treatment.

Sludge de-watering

The most commonly used pre-disposal treatment process is sludge de-watering. This process is often necessary so that the sludge can be more easily handled and is of a consistency suitable for landfill disposal. This procedure can also be utilised for any wet coal ash or combined coal ash/FGD sludge wastes. Most frequently, sludge de-watering is accomplished by sedimentation of the suspended solids in surface impoundments or, in some cases, in clarification tanks. This type of de-watering is carried out at 80% of the utilities in the USA.

After the waste solids have had sufficient time to settle, the water layer is drawn off the tank or impoundment and is either discharged or recycled back to the plant as sluice or cooling water. The sludge layer containing the solid ash and other particles is allowed to accumulate for several months (or longer), and is finally dredged after the pond is drained. With this process, the solids content (initially 5–15% by weight) can be increased to 30–60%. The final solids content in the sludge is affected by the sedimentation impoundment or tank design, the initial solids content, the liquid and solids retention times, and the physical and chemical characteristics of the solid particles.

Even after de-watering, the settled sludges often have a mud-like consistency and still contain so much free liquid that they are inappropriate for landfill disposal. In this case, the sludge may be further de-watered by natural or mechanical processes.

Reagent addition

Most FGD sludges, and some other wet sludges, can be rendered less chemically reactive and/or more structurally stable by adding solidification, stabilisation or fixation reagents. This practice is not widespread.

Solidification agents, such as sawdust or soil, absorb the liquid but do not chemically react with the sludge. Stabilisation and fixation reagents chemically react with part of the sludge, either the water, dissolved solids, particulate solids, or some combination of the three, and in some cases may render potentially hazardous material non-hazardous as a result. All of these processes result in an increased volume of waste that contains less free water and is easier to handle than the original waste stream. An additional benefit is an increase in the structural integrity (shear stress and load-bearing potential of the waste material) so that it may be placed in deeper disposal facilities and covered with more material.

Low-volume waste treatment

The major methods available for low-volume waste management and treatment include:

- co-disposal;
- contract disposal;
- evaporation;
- incineration;
- neutralisation;
- physical/chemical treatment;
- recycle/reuse.

The type of waste management method used most often depends on the type of low-volume waste stream.

Following the US Steam Electric Power Effluent Guidelines, additional treatment may be necessary for leachate generated from CCWs landfills. Cook and others (2013) explored treating landfill and impoundment leachate with the following options:

- precipitation; an agent is added to aid the coagulation of particles, which then rise to the surface or settle to the bottom of a tank. Solids are collected and typically de-watered prior to disposal. Examples include direct chemical precipitation, co-precipitation, lime softening, electro-coagulation, and zero valent iron;
- biological treatment; where bacteria and/or vegetation are used to consume waste and other organic materials from wastewater. Examples include surface flow wetland, gravel media biofilter, and anaerobic biological treatment;
- adsorption; a process in which molecules of a substance (such as a fluid) are absorbed on the surface of another substance (such as a solid). Examples include ion exchange, activated alumina, and activated carbon;
- membrane technologies; which force wastewater under pressure through a semi-permeable membrane, removing constituents in the process;
- evaporation methods: where the wastewater is heated and the resulting water vapour is condensed for collection and discharge.

7 Conclusions

While coal remains a major fuel for energy generation, large amounts of coal combustion products (CCP) will be generated. Worldwide production of coal combustion products (CCPs) was approximately 780 Mt in 2010. 415 Mt or 53% of total production was utilised effectively and this varies widely between countries. The remaining CCPs are considered to be wastes and required to be stored or disposed of. There is a suspicion that the utilisation rate was exaggerated due to pressure from environmental protection groups as utilising CCP could enable power plants to lower their carbon footprint and minimise the effects on the environment. For example, in China, it was stipulated that 'by 2010, the rate of comprehensive utilisation of solid industrial waste should exceed 60%.' In reality, the utilisation rate of CCPs is far below this value. The exaggeration of CCP utilisation results in under estimation of the environmental impacts of CCWs.

CCWs are disposed of in surface impoundments, landfills, mine and quarry fills, and oceans. Ocean disposal is not commonly carried out today. Mine and quarry fill is a way of using the material but only takes a small proportion of CCP production. CCW is generally managed in two ways: landfills or surface impoundments. More than half of the coal-fired power plants manage their CCW on-site, mostly using surface impoundments. Landfills are commonly used off-site. On-site surface impoundment has low delivery costs but requires large amounts of water and more land than landfill disposal. It also has a bigger potential for generating leachate. After the catastrophic ash spill from the US Kingston power plant, more attention has been put into dry ash landfills.

CCW management and its environmental impacts are influenced by its characteristics. Depending on the mineral components of coal, the combustion technique, and the pollution control technology used, one or more types of CCPs, such as fly ash, bottom ash, boiler slag, FBC ash, or FGD residues, are produced. The physical and chemical properties of each type of ash are influenced by the coal source and its quality, combustion process, cofired materials, air emission control methods, degree of ash weathering, and so on. The texture, particle size, colour, and bulk density are different for different type of ashes. While the type of ash and its characteristics vary, all CCW is likely to include certain amounts of toxic constituents, primarily heavy metals such as aluminum, arsenic, beryllium, boron, calcium, cadmium, chromium, cobalt, iron, lead, manganese, mercury, molybdenum, selenium, and silicon. The waste may also include a certain level of toxic organic compounds, such as dioxins and PAH. These toxic substances pose a risk to human health and the environment when CCWs are disposed of without proper management.

The US EPA put forward a risk assessment to characterise waste management scenarios and found that the majority of the damage and highest potential health risks associated with CCW involved its deposition into unlined units and/or with no groundwater monitoring systems. With a liner and groundwater monitoring system in new CCW disposal facilities, along with practices to prevent co-disposal of coal refuse with CCW, future risks from on-site CCW disposal are likely to be lower.

The primary environmental concern for CCW disposal is leaching which may contaminate surface and groundwater. Leaching is a complex process, which depends on many factors, namely chemical speciation of the constituent, solution pH, availability of the constituent for leaching, and age (weathering) of the ash. In addition, subsequent chemical reactions and secondary mineral formation can further modify leaching characteristics. There is a variety of laboratory leaching test procedures. When these are applied to CCW testing, field conditions known to influence leaching and initial and final site conditions need to be considered. The US EPA has validated a suite of four leaching tests (LEAF methods) in order to provide a comprehensive and sound laboratory protocol for evaluating leaching under a variety of conditions. The seriousness of the environmental impacts from leaching is still being debated. The US EPA, together with the US Environmental Integrity Project and Earthjustice, identified over 100 damage cases in America. However, EPRI thinks that these cases are primarily at older, unlined facilities. Retrofitting activities are either ongoing or completed for these sites. The real cases for damage may not be as high as previously thought. There are reports of water contamination from China and India. Liner, leachate collection systems, groundwater monitoring systems or pre-treatment technologies should be employed to prevent potential environmental risk posed by leaching.

The US Kingston disaster provides a ‘wake-up call’ about the danger from the catastrophic release of CCW due to the structural failure of surface impoundments. Dam safety and planning requirements must be applied to regulate the construction, operation, and maintenance of surface impoundments.

Fine dry fly ash can be airborne under windy conditions. However, little has been published about the environmental effects of dust generated at CCWs disposal sites. Following the US Kingston release, several air monitoring stations were installed as part of the clean-up and monitoring efforts. Results from tests of the air samples collected from these stations showed that none have exceeded the US air quality standards and there was no known threat to public health. However, more severe cases have occurred, such as a sandstorm in China. At disposal sites, windblown ash can be controlled by spraying water onto the open ash areas and covering inactive areas. Although there are concerns about the risks from radioactivity and mercury, no significant damage has been reported.

CCWs management needs to be guided by legislation. There are no finalised regulations currently in place up to now although lively debates have taken place over the years in the USA, Europe and South Africa about regulating coal ashes as hazardous waste. At the moment, CCPs are generally classified as non-hazardous waste. While the decision on whether CCPs are hazardous or not is very important, for CCW management, the impact is not as big as for CCP utilisation because the liner requirements for hazardous or non-hazardous wastes (Subtitle D and Subtitle C) are identical. From a technical perspective, both classifications demand the same technologies in preventing leaching.

Over the last decade, a number of changes have occurred globally in the coal-fired power generation sector that have affected quality and quantity of CCPs. The regulations for CCW management which are currently under discussion will also have an impact on the cost of CCW disposal. CCWs management is facing new challenges.

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