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# Effect of weathering transformations of coal combustion residuals on trace element mobility in view of the environmental safety and sustainability of their disposal and use. I. Hydrogeochemical processes controlling pH and phase stability



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## ABSTRACT

Coal combustion residuals (CCRs) are one of the most abundant high-volume waste materials disposed in impoundments worldwide. Some methods of CCR recycling, e.g. their use as structural fill for low lying areas or as soil amendment, also expose this material to atmospheric conditions. Combustion processes result in concentration of trace elements in CCRs at about an order of magnitude compared to coal. In order to assess an effect of long-term weathering transformations of CCRs on trace element binding/release, a study has been carried out. It is based on the chemical composition of real pore solutions extracted from the most abundant primary alkaline Class F bituminous CCRs, 0 to >40 years old, sampled from the surface layer and vertical profiles at four different impoundments. In this part of the study, results of a hydrogeochemical simulation of the saturation state of real pore solutions with respect to mineral phases of CCRs with use of the PHREEQC program, related to actual pH values reflecting the full cycle of weathering transformations, have been discussed. This study is the first geochemical proof of the general trend towards a progressive acidification up to pH < 4 of primary alkaline CCRs due to release of protons during internal processes of formation of gibbsite and aluminosilicate minerals, buffered by carbonates at the alkaline - near-neutral stages, and followed by parallel dissolution and buffering by aluminosilicates at pH < 7 after carbonate depletion, to the level up to pH-3.5–4.0. The intrinsic geochemical changes have resulted in the different susceptibility of trace elements to release and associated changes in risk to the environment at consecutive stages of weathering.

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## 1. Introduction

Coal combustion residuals (CCRs – often referred as coal ash or fly ash) are one of the most abundant high-volume wastes worldwide and their amount is steadily growing in parallel with increasing demand for coal-based power, in particular in developing countries. At the current coal production of 7823 Mt in 2013 (WCA, 2014), annual global generation of CCRs may be roughly estimated at over 780 Mt (Izquierdo et al., 2013) and will grow in

parallel with coal consumption.

The beneficial properties of CCRs make them suitable for a wide array of commercially and technically proven applications (Blissett and Rowson, 2012). However, the recycling of these materials is still far from complete use. While WWCCPN (2014) estimates CCR use at 41%, according to other sources the global average is close to 25% (Blissett and Rowson, 2012). As a result, the major portion of the total CCR stream is being disposed. The most widely used method of CCR disposal is sluicing coal ash (fly ash with an admixture of bottom ash) to surface impoundments in a form of slurry (coal ash: water mixture).

Being exposed to atmospheric conditions, CCRs are subject to weathering processes that may greatly influence the environmental behavior of these materials. These are relatively young

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products that appeared following common use of electrostatic precipitators in the 1950s, and thus the oldest identified coal ash impoundments are about 50 years old.

Recent environmental concerns pertaining to cases of ground-water pollution from leaching at CCR disposal sites and structural failures of impoundments (e.g. Ruhl et al., 2009, 2010; US EPA, 2010a,b; Earthjustice, 2014) have shown the need for a consistent approach to the potential environmental impact of CCRs exposed to atmospheric conditions, and for development of sustainable management criteria based on solid knowledge of CCR long-term environmental behavior. The awareness of existing gaps in knowledge is expressed by many authors (e.g. Pandey et al., 2009; Pandey and Singh, 2010; Lokeshappa and Dikshit, 2012; Ukwattage et al., 2013; Seshadri et al., 2013). These gaps originate mostly from the limited possibility of direct observations of weathering trends due to the mostly not yet much advanced weathering processes at CCR disposal sites. However, efforts in this field are steadily growing.

The properties of CCRs depend on the nature of the feed coal burned, the combustion technology and the pollutant emission controls applied. There are different classifications of coal fly ash based on its composition, properties and behavior (e.g. Roy and Griffin, 1982; Vassilev and Vassileva, 2007). The most widespread is the ASTM C618 classification, defining Class F and Class C fly ash. Class F fly ash originates mainly from bituminous coal and contains <10% CaO, thus has no cementing properties. This is predominantly an alkaline material. However, some Class F fly ashes with low Ca/S ratio, are primary acidic (e.g. Querol et al., 2001; Jankowski et al., 2006; Ward et al., 2009; Roy and Berger, 2011; Izquierdo and Querol, 2012). Class C fly ash originates mainly from subbituminous coals and lignites with higher CaO contents, generally CaO>20%, and typically displays cementing properties. In general, the prevailing type of CCRs worldwide is primary alkaline class F fly ash.

A large amount of research has been focused on the leaching behavior of these materials and assessment of possible environmental impact, (e.g. Sajwan et al., 1999, 2003, 2006). The majority of these studies are related to freshly generated CCRs and report relatively short-term and analytically limited experiments. Relatively long-term column or lysimetric studies typically cover less than a decade (Hjelmar, 1990; Fruchter et al., 1990; Janssen-Jurkovičová et al., 1994; Twardowska and Szczepańska, 2002; Wang et al., 2008; Izquierdo and Querol, 2012; Neupane and Donahoe, 2013; Mbugua et al., 2014).

A number of studies on weathering transformations of CCRs and their implications for trace elements release and mobility have been carried out. Since trace elements have been found to be mostly bound to glassy surfaces of fly ash particles (Adriano et al., 1980; Dudas and Warren, 1987; Evangelou, 1996; Gieré et al., 2003; Spears and Lee, 2004), temporal changes in susceptibility of trace elements to release have been associated with the share of their surface enrichment, devitrification of the glass phase, the role of pH, CO<sub>2</sub> partial pressure (Yan and Neretnieks, 1995; Evangelou, 1996; Spears and Lee, 2004; Spears and Martinez-Tarrazona, 2004; Kim et al., 2010), type of crystalline phases attached to the glass spheres (Gieré et al., 2003; Kim and Kazonich, 2004; Chancey et al., 2010) and formation of secondary minerals that are alleged to impact the trace elements mobility (Warren and Dudas, 1985; Fruchter et al., 1990; Donahoe, 2004; Twardowska and Szczepańska, 2002, 2004; Tiruta-Barna et al., 2006; Kim et al., 2010; Hareepsad et al., 2011; Akinyemi et al., 2011, 2012; Mahlaba et al., 2011, 2012; Izquierdo et al., 2013; Eze et al., 2013; Neupane and Donahoe, 2013; Mbugua et al., 2014). The prevailing methods for studies of trace element release induced by weathering are based on leaching tests (e.g. Warren and Dudas, 1985; Tiruta-Barna

et al., 2006; Kim and Kazonich, 2004; Spears and Lee, 2004; Hareepsad et al., 2011; Akinyemi et al., 2012; Neupane and Donahoe, 2013), and only very few works use actual porewater analysis either as the major (Twardowska and Szczepańska, 2002, 2004; Kim et al., 2010) or a supportive tool (Lee and Spears, 1995, 1997; Spears and Lee, 2004) in CCR weathering transformation studies. A relatively large data set on porewater analysis in CCR impoundments has been released by the US EPA (2010b), although without reference to the associated weathering transformations or pH values.

Due to the complexity of real-world temporal transformations of anthropogenic waste materials, only direct observations may reliably elucidate long-term environmental safety and sustainability of bulk waste materials introduced into the environment. Nevertheless, the weathering processes in CCRs and their temporal environmental consequences largely remain unclear.

In order to identify the trends in long-term environmental impact of primary alkaline Class F CCRs, as the most abundant and potentially the most problematic CCRs worldwide, and to evaluate trace metal mobility at the major stages of CCR weathering transformation, a study was performed on the chemical composition of pore solutions extracted from 0 to >40 year old material disposed at the impoundments. These data were utilized for trend analysis of hydrogeochemical transformations of disposed CCRs related to major and trace element release from the CCRs to solution at the different stages of weathering, and for elucidation of environmental implications of these trends on a long-term scale. Primary acidic Class F and alkaline class C residues were not included in the study. To our knowledge, this is the first real-time study on the full cycle of geochemical weathering transformations affecting major and trace element leaching trends over time in disposed CCRs, based on the analysis of actual pore solutions from materials of different age and weathering stages in different impoundments, and on hydrogeochemical simulation. The paper presents an analysis of the temporal mineral formation/dissolution in primary alkaline CCRs due to alterations in the saturation state of pore solutions with respect to solid phases, resulting in the relevant pH trends as a consequence of internal processes.

## 2. Materials and methods

### 2.1. Selected disposal sites

Criteria for the selection of objects for studies of CCR weathering transformations comprised: (i) representativeness of CCRs composition; (ii) adequately long and well-known history of a disposal site construction; (iii) easy access to the sections where material of different age was disposed. Four impoundments of a typical primary alkaline Class F coal ash located in different parts of Upper Silesia, Poland, were selected for the study: Halemba (HAL), Blachownia (BLA), Jaworzno (JAW), and Rybnik (RBK) (Appendix A, Fig. A1/A-D).

All impoundments were sited in unlined disused sand quarries in the late 1960s and early 1970s. They were constructed in subsequently filled sections, and had well-defined histories of construction. The time required to fill a section depended upon its capacity. The average thickness of CCR layers at the HAL and BLA sites in each section was 5–6 m, the typical duration of section filling was 5–10 years. At the JAW and RBK sites, the maximum thickness of the CCR layer was 20 m. At the HAL and BLA sites, a mound-type two-layer terrace constructions were formed with embankments above ground level, while the JAW and RBK sites retained a pond-type character. The detailed characteristics of the RBK site have been presented elsewhere (Twardowska and Szczepańska, 2002). Of all four sites, three sites (HAL, JAW and

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