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Effect of weathering transformations of coal combustion residuals on trace elements mobility in view of the environmental safety and sustainability of their disposal and use. II. Element release





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ABSTRACT

This paper is the second one of two companion papers. It presents results of a study aimed at assessing the effect of real time weathering transformations of Coal Combustion Residuals (CCRs) on trace element binding/release and its environmental implications. The study is based on the chemical composition of pore solutions extracted from primary alkaline Class F CCRs, 0 to >40 years old, sampled from the surface layer and vertical profiles at four selected typical CCRs impoundments. The long-term weathering transformations were found to lead to gradual acidification to pH < 4 of this primary alkaline material, due to internal processes of mineral formation/dissolution. Direct analysis of the pore solutions and a statistical analysis have shown different susceptibility of many trace elements to release during internal acidification processes occurring at consecutive Wash-out I (pH > 8), Dissolution II ($8 \ge pH \ge 7$) and Delayed Release III (pH < 7) stages of weathering compared to that at external sources of pH. The elements occurring in the CCRs are represented by three major groups showing the highest release to pore water: (a) within the acidic pH range (Na, K, Zn, Fe, Cd, Mo, Cr, B, Mn, Be and Ni; (b) within the nearneutral pH range (Al, V, Ba, Cu and Ag) and also Sb, Hg and Co not analyzed at pH < 7; (c) within the alkaline pH range (Ca, Mg, Pb, As, Se, Tl). Elements whose concentrations exceeded the threshold values for good chemical status of groundwater (TVs) at all weathering stages over the entire pH range studied were K, Al, B, Cr, Mo, V, As, Se, Sb and Hg, while Na, Zn, Fe and Cd showed particularly high delayed release at pH < 7, thus confirming the need of a precautionary approach to CCRs uncontrolled disposal and bulk reuse as common fill in view of long term environmental safety and sustainability.

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1. Introduction

The study of real-time hydrogeochemical transformations in primary alkaline Class F coal combustion residuals (CCRs) representing material from 0 to >40 years old disposed at four different impoundments (Stefaniak et al., 2015) has shown that the general weathering trend is toward the acidification of the primary alkaline material to pH < 4, resulting from simultaneously occurring internal processes of mineral formation/dissolution. Due to the different

affinities of elements to incorporation in primary, newly forming and decomposing mineral phases and the known strong influence of pH on the mobility of elements, these processes affect their leaching behavior and the environmental sustainability of their disposal and use. The short period associated with CCRs disposal practice from a geochemical perspective and the generally low rates of weathering transformations have often led to underestimation of the potential environmental and health risks from CCRs exposed to environmental conditions and to neglect of precautionary principles. A number of beneficial properties that in many cases allow using CCRs as a substitute for virgin materials encourages reuse of these otherwise troublesome bulk residuals for many purposes, also connected with the environmental exposure. A good example was promotion by the US EPA since 2001 of CCR use without following accepted and standard practices in risk

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assessment. This resulted in increase of CCR use from 32% in 2001 to 44.6% in 2008; 22.7% of the generated CCRs were used for explicitly non-point uncontrollable applications (structural fills/ embankments, soil modification/stabilization, snow and ice control and agriculture). After the 2008 catastrophe of CCRs impoundment in Kingston and many other cases of environmental damage and contamination (Source Watch, 2012), the US EPA released in 2010 proposal to regulate CCRs and stopped the program promoting their beneficial use (US EPA, 2011). In some developing countries, environmental awareness concerning possible adverse impact of CCRs appears to be still rather low – the results of numerous shortterm studies on freshly generated CCRs are presented to prove that this is "a resource and not a waste product", a commodity which should be more extensively used in applications connected with uncontrolled CCRs exposure such as agriculture or structural fill (e.g. DVC, 2004; TIFAC, 2004; Jala and Goyal, 2006; Dhadse et al., 2008; Singh et al., 2010; Aktar, 2011; Singh, 2011). A number of researchers consider utilization of the acid neutralization capacity of CCRs for the amelioration of soil acidity or Acid Mine Drainage (e.g. Surender and Petrik, 2005; Gitari et al., 2008; Yeheyis et al., 2009; Manoharan et al., 2010; Kusuma et al., 2013). However, the majority of recent publications, also from developing countries, besides the advantages of CCR use for such purposes, indicate potential risks and impacts of element leaching from disposed or used CCRs on soil and aquatic ecosystems (e.g. Jankowski et al., 2006; Manoharan et al., 2007; Dellantonio et al., 2008; Jo et al., 2008; Pandey et al., 2009; Basu et al., 2009; Pandey and Singh, 2010; Singh et al., 2010: Neupane and Donahoe, 2013: Ukwattage et al., 2013: Park et al., 2014). A number of studies discuss the relation of leachability of elements with regard to their distribution in CCRs (Manoharan et al., 2007; Jegadeesan et al., 2008; Wang et al., 2008; Li et al., 2012), the origin and some properties of the CCRs such as geochemical features, initial acidity/alkalinity or sulfur content of the burned coal (Jankowski et al., 2006; Ward et al., 2009; Izquierdo et al., 2011; Izquierdo and Querol, 2012; Jones et al., 2012; Akinyemi et al., 2012; Neupane and Donahoe, 2013), coal co-firing with waste fuel (Izquierdo et al., 2008), the influence of magnetic fractions (Warren and Dudas, 1989; Kukier et al., 2003; Lu et al., 2009), compaction (Tiruta-Barna et al., 2006) or effect of bacterial activity (Tiwari et al., 2008). Many studies investigate effects of several major external and internal experimental parameters likely to occur under environmental conditions of CCRs disposal or bulk non-point use – particle size, pH, alkalinity, CO₂ partial pressure, liquid-to-solid ratio (L/S), temperature, freezing-thawing cycles, and leaching time (e.g. Otero-Rey et al., 2005; Wang, 2007; Jo et al., 2008; Kim et al., 2010). A large literature is related to the effect of CCR interaction with soils on mobility of elements (e.g. Su and Wong, 2004; Moon and Dermatas, 2007; Pandey and Singh, 2010; Riehl et al., 2010). Also, the effects of CCR treatment (e.g. Bhattacharyya et al., 2009) or of application of novel coal incineration technologies (e.g. Seshadri et al., 2013) to reduce mobility of trace elements are reported. These studies, although they significantly add to the knowledge on CCR leaching behavior under the influence of different factors, do not provide an insight on the realworld effect of long-term weathering transformations of the material exposed to environmental conditions on the mobility of different elements occurring in CCRs and on CCR sustainability. The up-to-date information presented in a critical overview of available data on leaching behavior of CCRs (Izquierdo and Querol, 2012), some long-term leaching experiments coupled with geochemical analysis (e.g. Bednar et al., 2010; Izquierdo et al., 2011; Neupane and Donahoe, 2013) and rare studies on the environmental effect of CCR weathering processes (Twardowska and Szczepańska, 2002, 2004; Donahoe, 2004; Tiruta-Barna et al., 2006; Dellantonio et al., 2008; US EPA, 2010; Hareeparsad et al., 2011; Akinyemi et al., 2011, 2012;

Eze et al., 2013), mostly based on accelerated simulation of leaching behavior, standard leaching tests, or on quantitative alterations of element contents in bulk CCRs. This study is the first real-time research focused on the evaluating the environmental risk from release of elements to pore solutions in the course of a full cycle of primary alkaline Class F CCR weathering processes, along with elucidation of relations between mineral formation/dissolution and leaching behavior of elements.

2. Material and methods

2.1. Data source and processing

The description of selected CCRs impoundments, material sampling, and techniques of pore solution extraction and analysis has been presented in a companion paper (Stefaniak et al., 2015). In total, 43 samples of pore solutions extracted by centrifuging from primary alkaline material 0 to >40 years old, representing different stages in the advancement of weathering transformations, and with a resultant pH from 11.80 to 4.29, were analyzed using ICP-OES, ICP-MS and IC techniques, and four indices and the concentration of 41 elements were determined. The distribution of elements was computed with use of statistical software packages IBM SPSS Statistics v. 19.0 and presented as boxand-whisker plots (SigmaPlot v. 10.0, Jandel Scientific, Corte Madera, CA). For interpretation purposes, the outcome of the study of weathering transformations resulting in temporal mineral formation/dissolution and pH alterations in the studied primary alkaline Class F CCRs, presented in the companion paper (Stefaniak et al., 2015), were utilized. Relevant data on chemical composition and mineralogical analysis of the freshly generated and weathered CCRs, as well as on the elemental speciation and assessment of the saturation state of pore solutions (SI values) obtained by use of the PHREEQC hydrogeochemical computer program are also provided in that paper.

2.2. Evaluation of element concentrations

The extent of weathering transformations of the primarily alkaline class F coal ash (CaO <10%) is best characterized by the resultant pH alterations caused by hydrogeochemical processes occurring in such CCRs exposed to environmental conditions over time (Stefaniak et al., 2015). Thus, concentrations of elements in pore solutions were evaluated with respect to the trends in leaching behavior related to pH ranges occurring at the consecutive stages of weathering: alkaline (pH > 8), moderately alkaline/nearneutral (8 \ge pH \ge 7) and acidic (pH < 7). For these ranges, a potential to negatively affect groundwater quality by exceeding Threshold Values (TVs) for good groundwater chemical status based on the criteria set out in the EC Water Framework Directive (EC WFD, 2000) and EC Groundwater Directive (EC GWD, 2006) was assessed. Threshold values (TVs) indicate the maximum contaminant levels in groundwater bodies of acceptable quality, in which values of physico-chemical parameters are elevated as a result of natural processes or a weak impact of anthropogenic activities. According to the EC environmental legislation, these values mark out the borderline between the good and poor groundwater chemical status (EC FWD, 2000; EC GWD, 2006).

2.3. Threshold values as evaluation criteria

Since groundwater is the most sensitive and the largest body of freshwater in the EC, and a main source of public drinking water supply in many regions of the EC and also in the world, measures to prevent or limit inputs of pollutants should ensure that under the Download English Version:

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