



## A mineralogical and geochemical study of three Brazilian coal cleaning rejects: Demonstration of electron beam applications



César M.N.L. Cutruneo<sup>a</sup>, Marcos L.S. Oliveira<sup>a,b</sup>, Colin R. Ward<sup>c</sup>, James C. Hower<sup>d</sup>, Irineu A.S. de Brum<sup>e</sup>, Carlos H. Sampaio<sup>e</sup>, Rubens M. Kautzmann<sup>a</sup>, Silvio R. Taffarel<sup>a</sup>, Elba C. Teixeira<sup>f,g</sup>, Luis F.O. Silva<sup>a,b,\*</sup>

<sup>a</sup> Laboratory of Environmental Researches and Nanotechnology Development, Centro Universitário La Salle, Mestrado em Avaliação de Impactos Ambientais em Mineração, Victor Barreto, 2288 Centro, 92010-000 Canoas, RS, Brazil

<sup>b</sup> Environmental Science and Nanotechnology Department, Catarinense Institute of Environmental Research and Human Development, IPADHC, Capivari de Baixo, Santa Catarina, Brazil

<sup>c</sup> School of Biological, Earth and Environmental Sciences, University of New South Wales, Sydney, NSW 2052, Australia

<sup>d</sup> University of Kentucky Center for Applied Energy Research, 2540 Research Park Drive, Lexington, KY 40511, USA

<sup>e</sup> Universidade Federal do Rio Grande do Sul, Escola de Engenharia, Departamento de Metalurgia, Centro de Tecnologia, Av. Bento Gonçalves, 9500. Bairro Agronomia. CEP: 91501-970, Porto Alegre, RS, Brazil

<sup>f</sup> Programa de Pós-Graduação em Sensoriamento Remoto e Meteorologia (UFRGS), Porto Alegre, RS, Brazil

<sup>g</sup> Fundação Estadual de Proteção Ambiental Henrique Luis Roessler, Porto Alegre, RS, Brazil

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### ABSTRACT

The background and anthropogenic levels of hazardous elements in the surface soil of a coal mining area depend on the geological setting of the region and the underlying soil material, but may also be influenced by water-borne or aeolian transport of sediment from adjacent coal-related waste piles. Very few studies have focused on the chemical and mineralogical composition of Brazilian coal cleaning rejects (CCRs), which may represent significant sources of soil or water contamination. In this study, we have investigated the quantitative distribution of minerals and potentially hazardous elements in CCRs and a run-of-mine coal from the Brazilian states of Rio Grande do Sul and Santa Catarina. The major minerals, identified by X-ray diffraction (XRD), high-resolution transmission electron microscopy (HR-TEM), and field-emission scanning electron microscopy/energy dispersive X-ray analysis techniques (FE-SEM/EDS) are kaolinite, quartz, mixed-layer illite-smectite, pyrite, jarosite, melanterite, gypsum, rutile, and calcite, while minor minerals include barite, hematite, siderite, sphalerite, and goethite. Galena, magnetite, zircon, and many other species may also occur as accessory/trace minerals. Pyrite and jarosite are relatively abundant in some cases, making up to around 4% or 5% of the mineral matter, with jarosite, melanterite, and gypsum probably formed by complex interaction of oxidation products from Fe-sulfides and clay or carbonate components, initiated by exposure and storage of the host material. Such atmospheric exposure promotes sulfide oxidation that releases substantial sulfate loads as well as  $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ , and  $\text{Al}^{3+}$ . Metals with the most severe discharges were Zn, Cu, Mn, Co, Ni, and Cd. Most of the trace pollutants in the CCRs displayed a pH-dependent solubility, being immobile in near-neutral samples but mobile under the low-pH conditions associated with oxidized material. The results highlight the complex interactions among mineral matter components of the CCRs during storage, and the potential for release of potentially hazardous elements in association with longer-term exposure and storage.

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### 1. Introduction

Brazil is the largest consumer of coal in the South America (Oliveira et al., 2012, 2013). However, with the increasing use of coal there is growing concern about the environmental and human health impacts from potentially hazardous trace elements released in the course of coal mining, cleaning, transportation, and combustion.

According to the Santa Catarina State Coal Industry Syndicate the average run-of-mine (ROM) coal production is 6 Mt/year, from which 3.5 Mt/year are rejected and disposed of in landfills (Marcello et al., 2008). The Rio Grande do Sul and Santa Catarina coal mining region (Fig. 1) was classified as a national environmental endangered area by a 1980 Federal Decree. Accordingly, the area obtained special government assistance to address polluted soil and water quality concerns. This assistance allowed the mining sector to meet Brazilian demands for steam coal while protecting the environment. In response to a court ruling by the Federal Prosecution following a public lawsuit, and in accordance with associated agreements, the coal mining companies and the federal government are devoting special attention to

\* Corresponding author.

E-mail addresses: [luis.oliveira@unilasalle.edu.br](mailto:luis.oliveira@unilasalle.edu.br) (M.L.S. Oliveira), [felipeqma@hotmail.com](mailto:felipeqma@hotmail.com) (L.F.O. Silva).



Fig. 1. Location of coal mines from which the samples were obtained and used in this study. **1.a:** Cambuí. **1.b:** Criciúma. **1.c:** Fine Copelmi and Coarse Copelmi.

environmental projects in areas degraded by past coal mining operations. For this reason, the need for research on the behavior and potential use of coal cleaning rejects (CCRs) has become more economically and environmentally relevant.

The impacts on human health and regional environment caused by coal use are mainly related to the type and abundance of the minerals present in the coals and, especially, to the levels of potentially toxic elements contained within the minerals (Dai et al., 2004a; Finkelman, 1993; Finkelman et al., 2002). Coal cleaning processes are designed to remove the minerals occurring in the coal prior to use. An understanding of the mode of association of minerals with the coal matrix is important before application of coal cleaning technology. Large amounts of coal-derived minerals become concentrated in the residues from these coal-cleaning processes, especially from high-ash coals such as Brazilian coals. Knowledge of the mineral characteristics is also very important in understanding the formation of ultrafine and nanoparticles during coal cleaning, and to develop the necessary control steps. Brazilian CCRs are

often disposed of, however, with few attempts to understand their potential impacts on the environment.

Brazilian coal mining companies have valid environmental operating licenses for mine exploitation and preparation plants, and substantial efforts are taking place to meet environmental regulations regarding site reclamation. A number of measures are applied to reduce the environmental impact of coal mining and coal cleaning activities, such as restricting truck traffic at night, watering roads to reduce dust formation, and covering trucks to prevent spilling. However, these measures have proven to be insufficient to prevent damage caused by mining activity in the Santa Catarina coal mines.

With the exception of secondary products associated with pyrite oxidation, the minerals in coal cleaning residues are usually similar to those found in coals (Dai et al., 2008a,b; Depoi et al., 2008; Kalkreuth et al., 2006, 2010; Oliveira et al., 2012; Querol et al., 2008; Ribeiro et al., 2013a,b; Silva et al., 2009; Suárez-Ruiz and Ward, 2008; Yossifova, 2014). Most of the minerals themselves do not present

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