



Direct identification of hazardous elements in ultra-fine and nanominerals from coal fly ash produced during diesel co-firing



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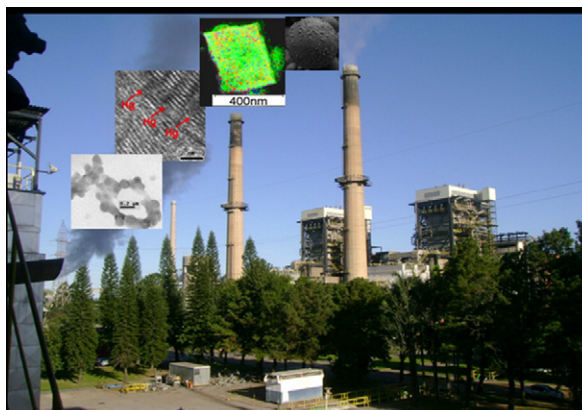
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HIGHLIGHTS

- Environmental and human health risk assessment was performed.
- Further exposition risk assessment required if future expansion is pursued.
- The advanced methodology has been applied to investigate elements occurrence and nanomineralogy properties.

GRAPHICAL ABSTRACT



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ABSTRACT

This study has provided an initial assessment of the environmental impacts and potential health effects associated with coal fly ash produced during diesel co-firing. Many hazardous elements that are typically detected by multifaceted chemical characterization by XRD, petrology, FE-SEM/EDS, and HR-TEM/SEAD/FFT/EDS in ultra-fine compounds and nanominerals from the co-fired coal fly ashes (CFAs). It provided an in-depth understanding of coal ash produced during diesel co-firing. Several of the neoformed ultra-fine compounds and nano-minerals found in the coal ashes are the same as those commonly associated with oxidation/transformation of aluminosilicates, carbonates, sulphides and phosphates.

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1. Introduction

Approximately 11% of the electricity generated in Brazil is produced by seven coal-fired power plants in the states of Rio Grande do Sul, Santa Catarina, and Paraná (ANEEL, 2006). These plants produce 3 Mt of ashes per year, which consist of 65–85% fly ash and 15–35% bottom ash (Levandowski and Kalkreuth, 2009). In addition, the existing coal-fired energy park in Brazil is expected to increase with another five power plants. When fully operational, this new scenario will triplicate the current coal ash production to around 12 Mt per year (Rohde and Silva, 2006).

Ultra-fine particulate matter emitted from coal power plants may contain potentially harmful constituents that include a wide range of hazardous elements and metalloids, polycyclic aromatic hydrocarbons (PAHs), amorphous silica, fluorides, elemental carbon and is known to impact adversely on environmental quality and human health (Finkelman, 2004; Ando et al., 1998; Quispe et al., 2012; Silva et al., 2011a, 2011b, 2011c, 2011d). Many of these particles are ultrafine (<100 nm) and nanometric (<50 nm) in size, thus with an enhanced potential for easy entry into the human body via the skin, digestive system or respiratory tract (Hochella et al., 2008; Nowack and Buchelli, 2007; Fubini and Fenoglio, 2007; Hoet et al., 2004). Given the growing interest in the toxicology of ultrafine particles, especially those containing hazardous elements (Ispas et al., 2009; Brian et al., 2009; Meng et al., 2007; Khan et al., 2007; Chen et al., 2006, 2005), this study reports on the direct observation of metal-bearing CFA ultrafine and nano particles using advanced electron beam.

Coal fly ash is typically polymineralic and chemically complex. The distribution, mode of occurrence and concentration of elements in the ash particles are crucial in determining their bioreactivity and biodurability. Potentially toxic trace elements homogeneously disseminated as impurities hosted in insoluble minerals pose a lower health risk than occurring as major constituents in individual, metal-rich ultrafine or nanoscale particles. Ultra-fine and nanoparticles are known to have a greater reactivity compared to larger-size particles of the same composition due to an increased surface-to-volume ratio (Navrotsky, 2001). For example, in particles smaller than 20 nm in diameter, at least 40% of the atoms are present on the surface of the nanoparticle (Banfield and Navrotsky, 2001). The effects of this reactivity in ultrafine and nanoscale materials are more likely as distances between functional groups and facets on nanoparticle surface decrease and quantum effects become important (Auffan et al., 2008). To date, such nanoscale effects and their influence on metal absorption or complexation as a function of particle size have not been studied in detail.

With the growing concern about the impacts of exposure to hazardous elements, there is a need for a better understanding of the relationship between geochemistry, nanomineralogy and the effects associated with both short-term and chronic inhalation exposure to coal ash. Therefore, we report on hazardous elements present in ultrafine and nanoparticles sourced from CFA, with emphasis on Al, Cr, Fe, Ni, Ti, Zr, which are the most abundant in the studied samples. Such metal-rich ultrafine and nanoparticles are very common in CFA and, given the fact that around 40% of global electricity is coal-generated (IEA, 2005), they are produced in large quantities (Silva et al., 2009a; Ruhl et al., 2009; Hower et al., 2008; Giere et al., 2006; Chen et al., 2004). An estimated 780 Mt of coal ash are produced every year (WWCCPN, 2012). Typically risk assessment methodologies have been widely applied for the management of contaminated sites. However, their application to element and coal power plants sites affected by tailing contamination is more limited (Moreno-Jimenez et al., 2011; Quispe et al., 2012). In a first-tier approach, an ecological risk assessment is based on geochemical analysis, where concentrations measured in soil are compared to established thresholds. These accepted levels are based on the worst possible scenarios: toxicity data on the most

sensitive species and the application of the most protective safety factors. Under these assumptions, the risk may be overestimated and may result in unnecessary remediation (Ollson et al., 2009). A risk assessment based on geochemical analyses is highly simplified and does not take into account factors such as the bioavailability of a contaminant or the simultaneous presence of different contaminants, which can affect toxicity and exposure estimates (De Zwart and Posthuma, 2005; O'Halloran, 2006). These facts are of the utmost importance for toxic hazardous elements. The availability of hazardous elements in the soil is highly dependent on CFA properties. Moreover, hazardous elements can be present in different geochemical forms with different geochemical and toxicological properties that can be transformed from one into another due to environmental changes. To have a more complete impression of the degree of pollution and toxicity of a contaminated site, it is recommended to consider alternative approaches that more accurately reflect specific site conditions. Direct toxicity assessment, conducted with CFA samples taken at the site, allows the measurement of the toxicity of complex mixtures of contaminants and can enhance the realism and certainty of the risk assessment. Although these techniques have an increasingly important role, they are not generally available in existing guidelines for ecological risk assessment on contaminated sites. Therefore, the ecotoxicological risk assessments based on direct toxicity assays need to be validated through field studies performed with natural samples.

The fact that coal-fired power plants can be a source of airborne particulate matter with elevated concentrations of toxic hazardous elements is well established in areas with coal generation plants (e.g. Iordanidis et al., 2008; Bhanarkar et al., 2008). Coal combustion ash has particulate sizes ranging from less than 1 μm to 100 micrometers in size (Teixeira et al., 1992; Blaha et al., 2008), with the inhalable sizes (<10 μm) being of most direct concern to human health. The most readily bioavailable particles of all will be those in the sub-micronic, ultrafine and nanometric size-ranges, although given the difficulty involved in their study relatively few publications have so far specifically characterized such materials.

In this work we aim to provide a multi-analytical approach to understand the implications of coal ash materials. A key constituent of this study was the identification and analysis of nano-particles in the different CFA, an area of research with potentially great environmental significance but about which very little is currently known. At the time of sampling, two of the units were being co-fired with a mixture of fuel oil and diesel oil as part of the boiler start-up procedure. The opportunity was, therefore, taken to investigate the influence of oil co-firing on the resulting coal combustion products. Little attention has been paid to the effects of co-firing with oil during start-up, even though this process may also have a significant impact on fly ash characteristics (Navarrette et al., 2004).

2. Methods

2.1. Sampling

Fly ash samples were obtained from the largest coal-fired power plant in Brazil, located in Santa Catarina State, which generates 857 MW/h (Quispe et al., 2012; Oliveira et al., 2012). The incineration temperature in the combustion chamber is ca. 1000–1500 °C, and about 98.5% of the fly ash is captured in the electrostatic precipitators (ESP).

A total of 7 CFA samples were collected from the ESP over a five-day period. The CFAs from Units 3 and 4 (Samples FA15 and FA16) were taken while oil was co-fired with coal as a means of starting up the respective boiler systems. Representative samples of around 15–20 kg of fly ash were collected from each unit. Samples of fresh dry fly ash were also collected from the current station output immediately prior to emplacement at the dry disposal site. The CFA characteristics have changed little over time due to the essentially constant geological source of the coal supplied (Depoi et al., 2008; Silva et al., 2009a,b).

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