

Fractionation of metals and metalloids by chemical bonding from particles accumulated by electrostatic precipitation in an Argentine thermal power plant

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Abstract

A study was undertaken to evaluate the distribution of Al, As, Cr, Cu, Fe, Mn, Ni, Pb, Ti, V and Zn in fly ashes collected in the electrostatic precipitator of a thermal power plant in San Nicolás (Argentina). Five samples were collected during one week of operation. For the fractionation, the scheme applied consisted in extracting the elements in four fractions namely (i) soluble and exchangeable elements; (ii) carbonates, oxides and reducible elements; (iii) bound to sulfidic metals; and (iv) residual elements. Metals and metalloids at $\mu\text{g g}^{-1}$ level were determined in each fraction by inductively coupled plasma optical emission spectrometry (ICP OES). For validation, a standard reference material (SRM 1633 coal fly ash) from NIST was subjected to the same chemical sequential extraction procedure that the samples. X-ray diffraction powder (XRD) analysis and scanning electron microscopy (SEM) were used to characterize the major minerals present in the matrix. Total analyte concentration (in $\mu\text{g g}^{-1}$) varied from 10.6 for Pb to 17,622 for Al. Minimum and maximum concentrations (in $\mu\text{g g}^{-1}$) found in individual samples in the four fractions were: Al, 92.7–9668; As, <0.3–143; Cr, 2.0–10.4; Cu, <0.2–35.6; Fe, <0.3–4992; Mn, <0.1–128; Ni, <0.3–139; Pb, <0.5–9.1; Ti, <0.3–2243; V, 17.0–112.9; and Zn, <0.1–68.2. The leachability of the 11 elements under study proved to be different. Low percentages of Al (1%), V (7%) and Cr (8%) were detected in the most bioavailable fraction. Arsenic was found to be most abundant in the non-silicate phase, represented by the second and third fractions, while Cr, Fe, Ni, Pb and Zn were mostly associated to the residual fraction.

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1. Introduction

The major use of coal today is the generation of power through combustion. The coal combustion by-products in thermal power plants are significant sources of potentially toxic elements into the environment. A 1000 MW power plant has a normal consumption of 12,000 t day⁻¹ of sub-bituminous coal

[1] and coal ashes are generated in a very aggressive combustion process. The amount and nature of metals and metalloids released are related to the composition of the coal burned and the technology employed in the power station. Coal contains many elements (As, Cd, Cr, Cu, Hg, Mn, Ni, Pb, Se, Sn, etc.) and most of them have been reported to be human carcinogens [2]. After combustion, these elements are typically found in the bottom ashes, the fly ashes and the gaseous phase. Electrostatic precipitators can remove more than 95% of the fly ashes but particles with aerodynamic diameters less than 10 μm are likely to be emitted directly to the atmosphere. Studies on the mobility of metals and metalloids during coal combustion

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have demonstrated that their volatility depends on their affinities and concentrations, and on the physical changes and chemical reactions of these elements with sulfur or other volatile elements [1].

A percentage of coal fly ash (~ 10–20%) is used in the concrete and cement manufacturing industries [3,4] and in other construction areas. However, ashes are more often discarded in sites such as lagoons, settling ponds or landfills [5], requiring investments for the non-productive use of land resources and long-term maintenance. As a consequence, alternative uses for fly ash other than the cement and construction industry have been and still are sought. Iyer and Scott [6] have reviewed such possible uses that include synthesis and application in materials, adsorbents and waste management, materials recovery and agriculture. In this context, knowledge of the chemical composition, physical characteristics and fractionation by chemical bonding of the ashes are of prime importance to assess the environmental impact and health risks. The leaching and consequent mobilization of the elements present in the ashes into waters and soils constitutes a serious risk that has to be evaluated.

Studies based on chemical sequential extractions provide useful information about solubility, origin, mode of occurrence, biological and physicochemical availability, mobilization, fate and transport of trace metals into the environment. Recently, Smichowski et al. [7] have reviewed metal fractionation of atmospheric aerosols via sequential chemical extraction procedures. The same authors also reviewed the role of atomic spectrometric techniques in the determination of chemical elements in atmospheric aerosol, including fractionation studies [8]. It is important to remark that the information gained by employing chemical sequential extraction procedures is operationally defined. Thus, the term “operational” is meant to indicate that the data obtained are dependent on the experimental approach adopted. Their operationally defined nature may be considered a weakness. In spite of this, sequential extraction methods are important approaches and the knowledge gained using these schemes would considerably improve our understanding of the risks for human health and the environment.

Different schemes have been developed and applied to the chemical fractionation of fly ashes from different origins both for research and regulatory controls [9–11] developed a five-stage leaching procedure that was applied to fly ash samples collected at several emission sources in Austria, Hungary and Switzerland. The analysis of the extracts by atomic absorption spectrometry (AAS) and graphite furnace atomic absorption spectrometry (GFAAS) showed different leachability of Cd, Cr, Cu, Pb, V and Zn exhibiting different distribution patterns. Cadmium distribution in coal fly ash, soil and sediment was assessed by sequential extractions of three certified reference materials and subsequent determination of the metal in the extracts by GFAAS [12]. Smeda and Zyrnicki [13] applied a modified BCR extraction scheme [14] for studying the partitioning of metals in fly ashes.

In this study, fractionation of ashes by chemical bonding was performed using a four-step chemical sequential leaching pro-

cedure similar to that developed by Fernández Espinosa et al. [15] to establish the distribution of 11 elements between (i) soluble and exchangeable metals, (ii) bound to carbonates, oxides (reducible elements), (iii) bound to oxidizable and sulfidic elements and (iv) environmentally immobile fractions (included in the crystal lattice).

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2. Experimental

2.1. Instrumentation and reagents

A Perkin-Elmer (Norwalk, CT, USA) ICP Optima 3100 XL (axial view) simultaneous inductively coupled Ar plasma optical emission spectrometer provided with a Model AS 90 autosampler was used for trace elements determination. Instrumental details and operating conditions are summarized in Table 1.

Welding Ar from Indura (Buenos Aires, Argentina) was used for ICP OES determinations. Deionized distilled water (DDW) was produced by a commercial mixed-bed ion-exchange system Barnstead (Dubuque, IA, USA) fed with distilled water. All reagents were of analytical grade. Hydrochloric, perchloric and nitric acids and hydroxylamine chloride, ammonium acetate and hydrogen peroxide (Merck, Darmstadt, Germany) were used for sample treatment and preparation of the standards. Commercially available 1000 mg l⁻¹ standard solutions (Merck) of the elements

Table 1
Instrumental details and operation conditions for the determination of metals and metalloids in fly ashes by ICP OES

Instrument	Perkin-Elmer Optima 3100 XL
Forward rf power	1300 W
Frequency of rf generator	40 MHz (axial view)
Coolant gas flow rate	15 l min ⁻¹
Auxiliary gas flow rate	0.5 l min ⁻¹
Sample gas flow rate	0.8 l min ⁻¹
Solution delivery	1.0 ml min ⁻¹
Automatic sampler	Perkin-Elmer AS90
Nebuliser	Cross-flow with Scott type expansion chamber
Polychromator	Echelle grating, cross-dispersed, wavelength range (nm): 165–403
Detector	Segmented-array charge coupled device
Measurement mode	Continuous nebulisation
Signal processing mode	3 pixels peak area
Background correction	2 points
Wavelengths (nm)	As, 188.979; Al, 308.215; Cr, 267.716; Cu, 324.742; Fe, 238.204; Mn, 257.610; Ni, 232.003; Pb, 217.000; Ti, 337.279; V, 290.880; Zn, 206.200

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