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Geochemistry and nano-mineralogy of feed coals, mine overburden, and coal-derived fly ashes from Assam (North-east India): a multi-faceted analytical approach

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A B S T R A C T

In order to address the scarcity of information on the nature and mode of occurrence of minerals and elements in coal, coal mine overburden and coal ashes from North-east India, and also the relations between coal mineralogy and ash chemistry, the petrology, mineralogy and nano-mineralogy, and ash chemistry of some industrially important high-sulfur coals, mine overburden materials and fly ash samples from Assam (India) have been evaluated. A combined approach, using petrography, low-temperature ashing plus X-ray diffraction (LTA-XRD), field emission scanning electron microscopy (FE-SEM), high resolution transmission electron microscope (HR-TEM), Raman spectroscopy, and inductively coupled plasma mass spectrometry (ICP-MS), has provided new information on the mineralogy and nano-mineralogy of these sub bituminous coals and associated mine overburden. The presence of rare earth elements and yttrium (REY) in Northeast Indian coal, mine overburden and fly ash samples is also reported for the first time. The identification of these components may be significant from an economic point of view. Some Northeast Indian fly ashes, with REE oxides up to 1580 ppm on an ash basis, might possibly represent sources for recovery of rare earth elements.

1. Introduction

Coal has been and remains one of the major cornerstones of modern industrial society. The development of clean coal technologies is a priority area for research in many countries, with goals such as increased thermal efficiency and decreased pollutant emission. In addition to atmospheric emissions such as CO_2, coal burning produces large amounts of solid waste products annually, including fly ash, bottom ash, and flue-gas desulfurization sludge, some of which may contain Hg, U, Th, As, and other trace elements (Clarke and Sloss, 1992; Dai et al., 2010, 2014; Kukier et al., 2003; Meij, 1994; Ribeiro et al., 2011). Depending in part on the coal characteristics, utilization of coal may be associated with a number of environmental challenges, including soil erosion, dust, noise and water pollution, and impacts on local biodiversity.

The emplacement of overburden rocks and other reject materials generated from opencast coal mines in some areas may represent a major source of ecological and environmental degradation, especially if the materials also contain pyrite (which may form acid leachates) and high concentrations of potentially toxic trace elements. An area larger than 1000 hectares in the North-eastern coalfields in India has been filled to date with mine overburden waste materials. Due to its high sulphur content (2–12%), part of which is pyritic, the mine overburden from the North-eastern coalfields also tends to produce highly acidic leachates (pH 2.0–3.0) (Baruah et al., 2006; Dowarah et al., 2009).

Fly ash (FA) is the main solid waste product from pulverized coal combustion and is collected by the emission control equipment (usually electrostatic precipitators or fabric filters) in thermo-electric power stations. Depending on the feed coal, the combustion conditions and the ash capture technology, those fly ashes may contain significant concentrations of trace elements which, if mobilised into the environment, could represent a potential pollution source. The coal-based thermal plants in North-east India produce approximately 37,000 t of fly ash per year (Pandey et al., 2011). In order to provide a technical framework from which to address some of these environmental issues, a multi-faceted analytical approach has been carried out, applying advanced level characterization of the feed coals and the associated mine overburden and fly ashes, with special attention to key aspects indicating the geochemical, mineralogical and elemental composition of the different types of material.

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1.1. High sulfur in the northeast Indian coals

The coal samples were taken from the Ledo (ALD) and Tikak (ATK) collieries in the Makum coalfield, Assam, India (Fig. 1). The coal beds mined at these sites are of Oligocene age and occur in the Tikak Parbat Formation, the uppermost member of the Barail Group. The Tikak Parbat Formation, 300-600 m in thickness, is made up of well-bedded and massive, fine-grained sandstones, sandy shales, shales, and claystones, with thick beds of coal in the basal part (Ahmed, 1996). The coals have low ash (<20%) and high volatile matter yields (>35%), and high hydrogen (>5%) and sulfur contents (>2%) in comparison to other Indian coals (Baruah et al., 2006; Saikia et al., 2014). They have lower moisture and oxygen contents than similar low rank coals from the well-known Raniganj coalfield (Chandra et al., 1984). The northeast Indian coals are considered abnormal because their physico-chemical characteristics and behaviour are not commensurate with properties adopted for rank classification (Iyenger et al., 1960), and are mostly classified as subbituminous in rank. North-east Indian coals are mostly medium to high sulfur coals and generally produce Acid Mine Drainage (AMD) problems (Baruah et al., 2006). The Ledo and Tikak collieries are among the most AMD-affected mines in the Northeast coalfield, Assam.

1.2. Makum coalfield

The North-Eastern Coalfield (NEC) is located in the States of Assam, Meghalaya, Nagaland, and Arunachal Pradesh, with coal mining activity located in the first three States. The Oligocene coal deposits of Northeast India are spread along a linear belt of over-thrust known as the 'Belt of Schuppen' (Misra, 1992), where Tertiary strata have been folded and dispersed into a number of thrust slices, including the Makum coalfield. The Makum coalfield, largest of all the coalfields of North Eastern region, is located in the Dibrugarh District of Assam, with a small part extending into the Tirap District of Arunachal Pradesh. The adjacent coalfields to the east and west are the Namchik-Namphuk and Dilli-Jaypore coalfields, respectively. The Makum coalfield is located along the outermost northern flank of the Naga-Patkai Range, which is characterized by highly dissected topography caused by numerous rivers, streams, and water channels. The major coal mining activities in the north-eastern region of the country are located in this coalfield.

1.3. Acid mine drainage from high sulfur coals and mine overburden

Acid Mine Drainage (AMD), recognized as one of the most serious environmental problems in the world (Akcil and Koldas, 2006), occurs in the Makum coalfield. Acidic drainage occurs as a result of natural oxidation of sulfide minerals, mostly pyrite (FeS_2), contained in the coal, reject materials, and mine overburden, which are exposed to air and water (Egiebor and Oni, 2007). The problem of AMD has also been widely felt in the coal mining industries of the North Eastern Region of India. The high-sulfur subbituminous coals of this area (2–12% total sulphur) are typically dominated by the organic sulphur fraction (75-90 % of the total sulphur), and thus have only relatively low proportions of pyritic sulphur (Baruah and Khare, 2007; Baruah et al., 2006; Saikia et al., 2014). Even so, the formation of acidic drainage in the coal mines is very much pronounced. In order to understand the origin and extent of AMD formation in these mines, a detailed assessment of the mineralogical composition of the coal and associated mine overburden, including the Fe-mineralogy, needed to be carried out.

The nature and modes of occurrence of various minerals present in coals, mine overburden, and fly ash are important considerations in the design of control strategies for reducing their release and potential impact. The research covered by this investigation therefore included evaluation of the mineralogical and geochemical compositions of typical coals, fly ashes, and mine overburden from the Makum coalfield, and the occurrence of trace elements, including rare earth elements (REEs) as well as potentially hazardous components, in those materials. Such an understanding may also be helpful in assessment and mitigation of similar environmental degradation in other nearby coalfields. The relevant information has been obtained from a multi-faceted analytical approach using a combination of basic and advanced techniques, and is intended to open up an avenue that may assist with environmental management in the region.

2. Methods and materials

2.1. Sampling program

Run-of-mine samples were collected from two most active and industrially important coal mines in the Makum coalfield, the Ledo (ALD) and Tikak (ATK) collieries, using standard methods (ASTM

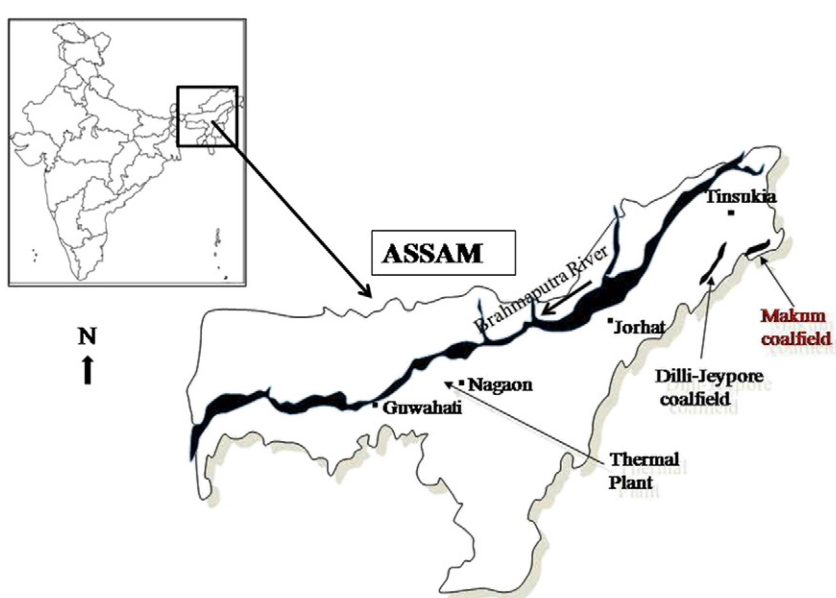

Fig. 1. Location map of the Makum coalfield and the thermal power plant (not to scale).