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Coal characteristics, elemental composition and modes of occurrence of some elements in the İsaalan coal (Balıkesir, NW Turkey)

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ABSTRACT

This study uses coal petrography, micro-FTIR, mineralogical, and geochemical analyses to determine the coal characteristics and controlling factors of elemental enrichments in the İsaalan coal (NW Turkey). The obtained data show that this subbituminous coal has a relatively low ash yield (avg. 19%), high hydrogen content (avg. 5.4%) and total S content (avg. 2.4%), on an air-dry basis. The maceral composition, along with micro-FTIR data on ulminite, suggests that lignin- and resin-rich vegetation was predominant. The coal facies data imply the precursor peat was accumulating under, mesotrophic anoxic conditions where water level was high and fluctuating. The mineralogical composition of the bulk coal samples is composed mainly of quartz, pyrite, and clay minerals and lesser amounts of feldspars and carbonates.

The statistical analysis of the bulk compositional data shows the major elements, including Al, Fe, K, Na, Mg, P, and the vast majority of trace elements, have inorganic affinity, whereas B presumably has organic affinity. The trace element concentrations are variable, and especially As and elements including B, Ba, Cr, Cs, Ni, Sb, and V are enriched compared to most world coals. The enrichments and distributions of Cr, Cs, Ni, and V were controlled by clastic inputs into a palaeomire, whereas As enrichments were controlled mainly by epigenetic and, to a lesser extent, syngenetic factors. Introduced SO_4^{2-} and As-rich surface- and groundwater from the basin margin area, along with anoxic conditions in the palaeomire, resulted in syngenetic formation of As-bearing pyrite. The circulating intra-seam solutions later liberated As from syngenetically formed pyrite, which resulted in As- and Ni-rich overgrowths around framboidal and massive pyrite crystals. These solutions could also easily mobilize B that was absorbed by organic matter. Therefore, B enrichment appears to be related to epigenetic factors. Tectonic activities during coalification allowed hydrothermal solutions from neighbouring epithermal mineralisation to penetrate and circulate along with underground waters that leached overlying volcanoclastic-volcanic strata and supplied a number of elements to the studied coal. Thus, infilling As-bearing pyrite and silica were formed mainly during coalification. All these results suggest that As-enrichment in the study area is more likely a combination of epigenetic factors and syngenetic-detrital rather than volcanogenic contributions, as suggested earlier for some other Turkish coal deposits.

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1. Introduction

Elemental enrichments in coal are controlled by multiple factors during peat accumulation and coalification. Understanding these processes is very important because enrichments of trace elements can cause environmental, health, and technical problems upon coal utilisation. Arsenic is one of the most hazardous trace elements, and its enrichment in coal can cause severe environmental and health problems such as endemic arsenicosis or dangerously elevated As concentrations in natural waters (Black and Craw, 2001; Finkelman et al., 1999; Finkelman, 2004; Belkin et al., 2008). Because of this, the mode of occurrence of As and the controlling factors of its enrichment in coal have been widely investigated (Yudovich and Ketris, 2005a, and refers. therein). Arsenic enrichments in coal can result from synchronous volcanic activities, leaching from a nearby As-bearing ore deposit, and the penetration of epigenetic hydrothermal solutions into the coal deposit. Arsenic generally associates with pyrite and other sulphide minerals in coal, whereas As is rarely associated with clay minerals and organic matter

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(Finkelman, 1994; Belkin et al., 1998, 2008; Kolker et al., 2000; Ding et al., 2001; Yudovich and Ketris, 2005a; Dai et al., 2006; Mastalerz and Drobniak, 2007). In addition, different morphologies of pyrite crystals may contain different As concentrations. For instance, framboidal pyrite crystals, which are related to the early diagenetic stage, are generally poor in As, and epigenetic pyrite (e.g., infilling pyrite) can be enriched in As, Cu, and Ni (Eskenazy, 1995; Ruppert et al., 2005; Diehl et al., 2012; Kolker, 2012).

Arsenic enrichment in Turkish coals is well-known and is described as "Turkish (volcanogenic) type" by Yudovich and Ketris (2005a). In this type, synchronous volcanic activities (e.g., volcanic ash deposition) during peat accumulation introduced S- and As-bearing inputs that elevated the As content in coal beds (Querol et al., 1997). Furthermore, As enrichments can be related to sulphide mineralization in cleats and fractures that precipitated from epigenetic solutions in coal seams (Karayiğit et al., 2000a). In specific cases, an organic form of As is also present in Turkish coals (Karayiğit et al., 2001; Palmer et al., 2004). Turkish coals are also characterized by the enrichments of B, Cr, Mo, Ni, U, and V in comparison with world coal averages (Karayiğit et al., 2000b; Tuncalı et al., 2002; Palmer et al., 2004). High Cr, Ni, and V contents are derived from clastic inputs from Cr- and Ni-rich serpentinites within ophiolites on the margins of Turkish coal deposits, whereas B and U enrichments are generally explained by the presences of synchronous volcanic activity along with the influence of leached water from volcanic and magmatic rocks into a palaeomire (Querol et al., 1997; Karayiğit et al., 2000b; Palmer et al., 2004).

Since the beginning of the Cenozoic, regional tectonic movements caused the development of regional volcanism and magmatic plutonic bodies along with fault-controlled sedimentary basins in NW Turkey (Köksoy and Ataman, 1980; Ercan et al., 1990; Yılmaz et al., 2000; Ersoy et al., 2014). Thus, basinal infillings (lacustrine and fluvial deposits) are intercalated with volcanic and volcanoclastic rocks. These basins host several economic coal deposits (e.g., Çan coal field) and coal horizons within lacustrine deposits (Köksoy and Ataman, 1980; Tuncalı et al., 2002). Regional volcanic activity and magmatic intrusions also caused the formation of several geothermal fields and metallic ore (e.g., epithermal) deposits in NW Turkey (Mutlu, 2007; Yiğit, 2012). As expected from such a geological setting, coal deposits in NW Turkey are characterized by high concentrations of As, B, Cr, Hg, Ni, and V (Karayiğit et al., 2000b; Tuncalı et al., 2002; Palmer et al., 2004; Gürdal, 2011).

The İsaalan coal deposit is located northeast of the town of Kepsut (Balıkesir Province, NW Turkey) and has been mined by surface techniques during the last decade. The studied coal deposit is smaller one among other deposits in NW Turkey, and no detailed coal-petrographical, geochemical, and sedimentological studies have yet been performed from the deposit. The geological data from surrounding areas are also limited, and mainly based on volcanic and regional tectonic studies (Ercan et al., 1990; Tetiker et al., 2015). The absence of coal-petrography and geochemistry data from the İsaalan coal and the high As concentrations in other coal deposits in NW Turkey motivated us to investigate this relatively unknown coal deposit. The main goal of this paper is to determine the coal-petrographical, mineralogical, and geochemical characteristics of the İsaalan coal, with the specific objectives of a) identifying elemental distributions; and b) evaluating controlling factors on elemental enrichments, particularly As.

2. Geological setting

The Miocene İsaalan coal deposit is located in a NE-SW trending graben at 512 m above sea level in NW Turkey. The northern margin and the basin basement (Fig. 1a) are mainly composed of ophilotic rocks of the Yayla Mélange and Permian-Trissaic Karakaya complex (basalts, marble, spilite), whereas in the southern margin Neogene volcanic and volcanoclastic rocks crop out (Akyüz and Okay, 1996; Ercan et al., 1990; Okay et al., 1996; Tetiker et al., 2015). Mesozoic metamorphic rocks (mainly marble and schist) with host hydrothermal alteration zones are present in the western margins (Yıldırım, 2002). The coal-bearing basin-infillings commenced with the deposition of alluvial fan sediments (Figs. 1b and 2). These units are overlaid by fluvio-lacustrine deposits that host coal beds (Fig. 1b) and are composed of interbedded claystone, marl, coal, and tuff strata. The tuff layers developed from synchronous volcanic activities during the Miocene in western Anatolia (Ercan et al., 1990; Ersoy et al., 2014). The period following coal-bearing sequence deposition was characterized by a regional compressive tectonic regime that caused development of the İsaalan syncline; thus, the coal-bearing sequences were folded and faulted. Similar deformations are also reported from other Neogene coal deposits in NW Turkey (Köksoy and Ataman, 1980). The coal-bearing sequences are overlain by Pliocene volcanics consisting of basalts and agglomerates. Finally, Quaternary units, mainly composed of alluvial deposits, unconformably overlie the basinal sequence (Fig. 1b).

3. Samples and methods

Applying channel sampling techniques, a total of 19 coal samples from the working coal seam were collected from a surface mine (Fig. 2). The sampling profile was 23.5 m thick in total, and the cumulative thickness of the coal beds is 12.25 m. The standard proximate-ultimate analyses and gross calorific value of coal samples were conducted using LECO TGA-601, SC-144DR, TruSpec, and AC 350 at Hacettepe University, Turkey, according to ASTM standards (ASTM D3174, 2004; ASTM D3175, 2004; ASTM D3302, 2004; ASTM D5373, 2004; ASTM D5865, 2004). Polished blocks from coal particulate (Ø < 1 mm) samples were prepared from crushed coal, according to ISO 7404–2 (2009) for maceral analyses. The microscopic studies were performed using Leica DM 2500P and Zeiss RS-III microscopes at the Indiana Geological Survey, USA. The identification of macerals followed the Stopes–Heerlen nomenclature as it is modified in the ICCP System 1994 (ICCP, 1994; 2001; Sýkorová et al., 2005; Pickel et al., 2017). Random reflectance measurements of eu-ulminite B were measured according to ISO 7404–5 (2009) from selected samples (CB-309, -310, -311, -313, -316, -320, -324, -326, and -327). In addition, random reflectance measurements were also collected on corpohuminite in one sample (CB-310) to determine differences in reflectance among huminite macerals. Coal facies indices, as noted by Karayiğit et al. (2016), were calculated based on maceral composition. The Tissue Preservation Index (TPI), the Vegetation Index (VI), the Groundwater Influence (GWI) and the Gelification Index (GI) (Calder et al., 1991; Diessel, 1992) were based on the formulas of Kalkreuth et al. (1991) and modified by Kalaitzidis et al. (2004) for low-rank Cenazoic coals. Micro-FTIR analysis of seven samples (CB-309, -312, - 315, -316, -319, -325, and -327) were conducted according to the standard methods, applying the same techniques and equipment as described by Chen et al. (2012) and Bechtel et al. (2016). Selected semiquantitative ratios from the absorbance peak integration areas were calculated to characterize the chemical properties of ulminite and resinite. Final average values for the macerals were calculated from the average values of each sample.

The mineralogical compositions were determined in all the samples using X-ray powder diffraction spectrometers with CuKα radiation (4–60° 2θ) at the Institute of Environmental Assessment and Water Research (IDAEA), Spain. Semiquantitative mineralogical compositions (%) of coals were calculated on a whole coal basis, as described by Li et al. (2016). Furthermore, 12 polished coal blocks (CB-309, -310, -311, -313, -315, -319, -320, -321, -322, -325, -326, and -327) were coated with carbon and examined under a Carl Zeiss EVO-50 EP type SEM–EDX at Hacettepe University. The elemental composition of the whole coal samples was determined by means of inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS) at IDAEA. A special two-step sample digestion method for the analysis of potentially volatile elements in coals as explained by Querol et al. (1995) was used to dissolve the samples prior to analysis. The international reference material SARM 19 was

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