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Effect of geological processes on coal quality and utilization potential: review with examples from western Canada

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Abstract

This review article compares the elemental and radionuclide concentrations in coals from western Canada, the vast majority of which are used for power generation in Alberta. The coals range in age from lower Cretaceous to middle Eocene, and in rank from subbituminous to high volatile bituminous. Some of the coals were deposited in deltaic lagoonal to marine settings while others formed under lacustrine conditions in intermontane graben settings or in alluvial plains. The role of source rock (provenance), depositional environment, tectonic regime and hydrologic conditions on elemental concentration and distribution will be discussed, with specific examples from western Canada. In addition, the effect of natural weathering, igneous intrusion and self-burning (spontaneous combustion) on the enrichment and/or depletion of elements will be presented. The emphasis throughout this review article will be on the fate of elements of environmental concern and interest (e.g. As, Ba, B, Cl, Co, Cr, Cu, Mn, Mo, Se, Th, U, V and Zn) and of radionuclides of the U and Th series upon coal utilization. This article is also intended for those not familiar with the geological or environmental sciences, particularly as related to fossil fuel utilization. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Trace elements; coal; environment; power stations; Alberta

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1. Introduction

Coals are known to concentrate elements as a result of interactions of organic matter with numerous processes [1]. The geochemistry of coal is strongly influenced by a number of geological parameters, including the nature of rocks near the coal basin and the rate of subsidence and uplift of the drainage area [2,3]. Other dominant factors that control the geochemical properties of coal are the following: the environment of coal deposition, the weathering of coal, the presence of intrusive rocks, and the self-burning of coal seams [4–6]. The depositional environment, the climatic and hydrological conditions and the nature of country rocks are more important in the early stages of coalification. Rank, tectonic setting and groundwater chemistry have greater influence on coal geochemistry in the later stages of coalification. It should also be noted that the original plant species that formed the coal could have taken up some elements by biological and physico-chemical processes [1]. Factors such as bacterial activity, pH and Eh of waters influence deposition of elements in coal.

The above processes can enrich, deplete or redistribute elemental concentrations in coal seams and coal-bearing strata. For example, country rocks influence the trace element concentration by controlling the composition of detrital minerals and aqueous species that are introduced into the coal-forming environment. They also influence the ionic composition of surface and groundwater that are responsible for epigenetic mineralization. Tectonism controls the composition and the amount of sediment input, whereas the hydrologic conditions influence the distribution of elements in the coal precursor (peat) through variations of pH and Eh. Finally, depositional environment controls the chemistry of interstitial waters.

The objective of this report is to examine the effect of geological processes on the concentration and distribution of elements of prime environmental concern and interest as well as selected radionuclides and polyaromatic hydrocarbons (PAHs) in coals from western Canada (Alberta and British Columbia). The processes that will be discussed, in some detail, include the following: type of country rocks (rocks that the peat is deposited on) and provenance (source of sediments), groundwater movement, tectonic activity, and depositional environment. Reference will be made to other processes, such as weathering, spontaneous combustion (self-burning) and igneous intrusions. The emphasis will be given on subbituminous coals that are currently utilized for power generation in Alberta. The coal deposits described in this study are shown in Figs. 1 and 2. The paper will also discuss the emission rates of potentially hazardous elements and radionuclides from power stations in Alberta, particularly those that are considered to be carcinogenic or toxic to humans and/or animals. Due to the confidential nature of the work with the power stations, the discussion will be limited to non-proprietary data.

2. Results and discussion

2.1. *Effect of country rocks, tectonism, and groundwater*

Coal deposits in Alberta are found in clastic sequences and have a simplistic layer cake stratigraphy. As a result, Alberta coals are not ideal to study the effect of igneous

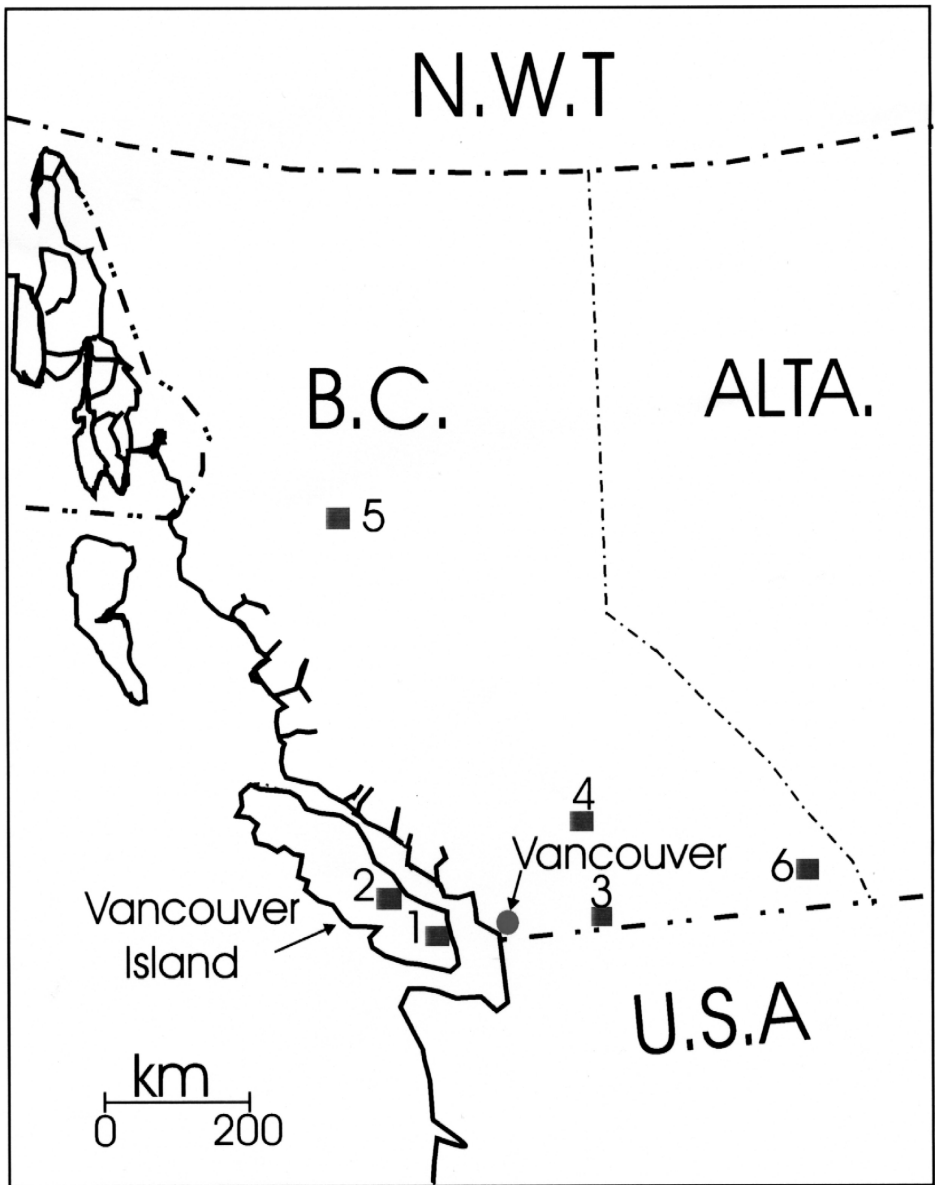


Fig. 1. Map of British Columbia showing the location of the coal deposits referred in the study (1 = Nanaimo coals, 2 = Comox coals, 3 = Tulameen coals, 4 = Hat Creek coals, 5 = Telkwa coals, 6 = Fording coals).

or metamorphic rocks and the effect of complex tectonic structures on trace element geochemistry. Therefore, in this study, examples on the effect of non-sedimentary rocks and tectonism will be given from coal deposits on Vancouver Island, British Columbia.



Fig. 2. Map of Alberta showing the location of the coal deposits referred in the study (1 = Montgomery coals, 2 = Vesta coals, 3 = Paintearth coals, 4 = Red Deer River Valley coals, 5 = Whitewood coals, 6 = Highvale coals, 7 = Genesee coals, 8 = Obed Mountain Coals, 9 = Coalspur coals).

Coal deposits on southern Vancouver Island are found in the Nanaimo and Comox subbasins, separated by a distance of about 100 km (Fig. 1). Although the coals are not being utilized currently, almost 67 Mt were produced from the 1850s to 1976 and exported to California [7]. These high-volatile bituminous coals of Upper Cretaceous age were formed in similar geological settings in terms of tectonics and depositional environment [8]. Briefly, the coals were deposited under coastal plain conditions during transgressive cycles [9]. The strata were folded and thrustured during the Tertiary [10], which resulted in the formation of highly sheared zones in some parts of the coalfields.

Despite the similarities, there are some differences in elemental concentrations and distributions within the coal seams. One of these factors is the nature of country rocks and the other is tectonism. The work of Van der Flier-Keller and Goodarzi [8] has shown that the Nanaimo coals contain more B, Ba, Cr, Rb and Sr than the Comox coals (Table 1) but the Comox coals are higher in As, Cu, Zn, Zr (Table 1) and the rare-earth elements (REE). The country rocks in the vicinity of the coal subbasins are Cretaceous volcanics, mainly ferrotholeiitic basalts and plutonic granodiorites. The increase in the proportion of acidic (Si and Al oxides being the main constituents) to mafic (Mg and Fe

Table 1

Mean concentration (mg/kg) of some elements in the Nanaimo and Comox coalfields

Element	Nanaimo	Comox	World
Arsenic	3.81	55.9	10
Barium	499	108.6	200
Boron	106	70	50
Chromium	28.9	19.1	20
Copper	12.2	18.2	15
Rubidium	12.8	10.4	15
Strontium	368	336.8	200
Zinc	9.6	11.3	50
Zircon	21	27.3	50
Europium	0.24	0.35	0.5
Cerium	8.65	10.34	20
Lanthanum	4.25	5.23	10
Samarium	0.91	1.1	2

Vancouver Island (taken from Ref. [8]). World data taken from Ref. [40].

oxides being the chief constituents) source rocks from the Nanaimo to the Comox coalfields (in a northern direction) is believed to be responsible for the variations in elemental concentrations between the two coalfields [8].

The influence of structural geology on the trace element geochemistry of coals is well known. Although the majority of faults on Vancouver Island are in the subsurface, one of the coal seams in the Nanaimo coalfield, the Wellington Seam, has been sheared and highly deformed. Elemental concentrations differ laterally between the undeformed and deformed parts of the seam and reveal that the distribution of trace element concentrations reflects the structural overprint. For example, Au is significantly enriched in the deformed seam, whereas B, Cl and Sr are depleted (Table 2). Other elements, such as

Table 2

Enrichment and depletion of selected elements (mg/kg) in the deformed Wellington Seam, Vancouver Island (modified after [8])

Element	A	B	C	Effect of deformation
Au	5	14	48	Highly-enriched
Co	4.1	2.8	9.1	Moderately enriched
Ni	13	10	18	Moderately enriched
Pb	1	NA	5	Moderately enriched
Sb	0.6	0.4	2.7	Moderately enriched
U	0.74	0.63	2.69	Moderately enriched
Th	1	1.1	5.3	Moderately enriched
Ce	8.7	6.5	22	Moderately enriched
Eu	0.24	0.16	0.55	Moderately enriched
Sm	0.9	0.51	2.11	Moderately enriched
B	106	90	26	Strongly depleted
Cl	128	60	10	Strongly depleted
Sr	368	380	130	Moderately depleted

A = average in Nanaimo subbasin coals, B = undeformed Wellington Seam, C = deformed Wellington Seam.

Co, Ni, Pb, Sb, Th, U and the REEs are moderately enriched in the deformed coal (Table 2). The interpretation given by van der Flier-Keller and Goodarzi [8] was that faults created permeability in the coal seam, which enhanced the movement of groundwater through the seam. Thus, certain elements present in the groundwater became available for absorption by the coal matrix (were leached) under suitable conditions.

Numerous studies of REE concentrations and distributions in western Canadian coals have shown that there is no relationship between REE concentrations and the age or rank of coal. Rather, source rocks, often modified by weathering, and the diagenetic history of the coal appear to control variations in REE [11]. Coal mineralogy is a function of source lithology (for detrital minerals). Many minerals in coal are thus related to the abundance of specific mineral forms in the mineral portion of the coal (the other portion being the organic). Source rocks control the initial detrital input (e.g. quartz) into the coal-forming environment. During this period the heavy detrital minerals and most of the clays are introduced into the system. Clay minerals modified by the coalification process mobilize the REE, particularly the HREE (heavy REE) that tend to be more mobile than the LREE (light REE) [12]. Diagenesis affects mostly calcite and iron-bearing minerals.

One of the best examples to describe the enrichment of heavy metals in coal due to groundwater activity is the Tulameen coal, Blakeburn Mine, in south central British Columbia.

Coals from the Tulameen coalfield (Fig. 1) were mined from 1919 to 1940 and almost 21 Mt were produced [7]. The coals are of Eocene age and are underlain by volcanic rocks. An ultramafic complex of Miocene basalts is located nearby. These high volatile C-B bituminous coals have been highly faulted, sheared, folded, and often tectonically thickened [7]. The platinum group elements Pt and Pd in the Tulameen coals are heavily enriched (up to 10000 ppb of Pt and 6100 ppb of Pd in the faulted coals compared to < 170 ppb of Pt and 40 ppb of Pd in the undisturbed coals) [13]. Otherwise, elemental concentration in these coals is within the range of other western Canadian coals. The ultramafic rock sequence near the deposit is the most likely source of the platinum group elements. The elements were leached by groundwater action from the ultramafic rocks and were transported to the site of the Tulameen coals some 30 km to the east. The intense folding enhanced coal permeability and its ability to absorb elements present in the groundwater. This is a clear case whereby groundwater removed elements from igneous rocks on one hand and deposited them in coal seams on the other.

2.2. Effect of depositional environment

Examples of the effect of depositional environment on elemental concentrations will be given from Alberta. The coals described include those from the Highvale, White-wood, Genesee (Upper Cretaceous-Tertiary), Vesta, Paintearth, and Montgomery mines (Upper Cretaceous), and those from the Red Deer River Valley (Upper Cretaceous) as well as those from the Obed Mountain deposit (Tertiary-Paleocene) (Fig. 2). All of the above coals are subbituminous to high volatile bituminous in rank. Coals from the

Highvale, Whitewood, Genesee, Vesta, Paintearth, and Montgomery mines are currently used for power generation in Alberta, while the Obed Mountain coals are exported to Ontario as thermal coals. The Highvale, Whitewood, and Genesee mines all mine the same coals (the seams can be stratigraphically correlated and are part of the same coalfield).

Goodarzi and Swaine [14] suggested that the degree of marine influence on coal could be inferred from its boron content. They proposed the following divisions: (a) < 50 mg/kg (ppm), (b) 50–110 mg/kg, and (c) > 110 mg/kg to indicate coals deposited under fresh water, slightly brackish water, and brackish water conditions, respectively.

The Highvale Mine coals have boron concentration in the 14–50 mg/kg range (mean is less than 50 mg/kg), while boron in the nearby (12 km away) Whitewood Mine coals is also less than 50 mg/kg (Table 3). These values are consistent with a freshwater influenced depositional environment. Evidence based on sedimentology and paleontology [15] points to deposition in a continental setting for the coal-bearing formation and for the overlying sedimentary strata. More specifically, the lower part of the coal-bearing formation represents alluvial fan deposits and the upper parts represent fluviolacustrine, lacustrine and swamp facies deposits [15].

On the basis of trace element geochemistry, Pollock et al. [16] identified two geochemical cycles during deposition of the No. 2 seam, Genesee Mine, located approximately 30 km southeast from the Highvale Mine. Boron concentration varied from 51 to 90 mg/kg in the coal (mean is 77 mg/kg; Table 3), although the

Table 3

Comparison of elemental concentrations (mg/kg) in subbituminous coals Alberta used for power generation with those in world coals (data for Canadian coals compiled from, Refs. 16,18,20,22,23). Word data taken from Ref. [40]

Element	Obed Mt.	Highvale	Whitewood	Genesee	Vesta	Paintearth	Montgomery	Hat Creek	World
Antimony	NA	0.7	0.7	0.8	3.3	0.08	0.08	0.5	1
Arsenic	4	3.8	4.7	0.8	0.9	1.3	2.4	12.7	10
Barium	490	700	450	537	450	467	815	450	200
Boron	50	< 50	< 50	77	208	237	291	40	50
Bromine	7.5	286	15.1	1.2	2	1.7	2.8	2	20
Chlorine	80.6	72.3	21.5	20.1	27	47	57	27	1000
Chromium	7.1	11.5	7	25.6	13.8	24	27	13.8	20
Cobalt	1.4	3.2	3.3	2	1.4	4.2	2.3	9.1	5
Copper	15.3	11.6	9.8	4.4	2	2.5	2	37.7	15
Manganese	31.8	100	110	3.2	27	23	113	28.8	70
Molybdenum	3.3	5	4.8	47.9	2.7	1.7	3.4	3.5	3
Thorium	1.9	5.6	6	2.4	1.8	6.7	5.8	2.4	4
Uranium	1.5	2.6	2.5	4.6	1.1	4.1	1.9	1.4	2
Selenium	1.1	1.7	1	0.6	0.8	0.8	1.2	1.1	1
Sulfur	0.4	0.5	0.6	0.5	0.45	0.5	0.5	0.5	NA
Vanadium	NA	17.1	19.3	1.1	3.7	8.1	7.9	122	40

Sulfur in wt.%.

NA = Not available.

concentrations were higher in the carbonaceous shale and mudstone near the top of the seam (91 and 119 mg/kg, respectively). The partings are removed by selective mining because they are present near the top of the seam. The shale and mudstone intervals located beneath coal intervals had lower B concentrations (21 and 31 mg/kg). Using the classification of Goodarzi and Swaine [14], it can be inferred that the Seam 2 coal was most likely deposited in a mildly brackish water influenced depositional environment. However, this is not in agreement with the sedimentological and paleontological evidence, which points to secondary enrichment of boron. The absence of boron enrichment in the nearby Highvale and Whitewood coals strongly suggests that the B enrichment in the Genesee coals is a localized phenomenon. The Genesee coals were deposited during a period of tectonic quiescence but experienced higher detrital input than the Highvale or Whitewood coals. Thick deposits of sandstone are present in the vicinity of Genesee that are absent at Highvale or Whitewood [17]. This indicates a greater fluvial influence for Genesee that was also responsible for the higher input of detrital minerals. The detritus included clay minerals rich in boron, which were incorporated in the geochemical cycles during deposition of Seam 2. Mean elemental concentrations in the Highvale, Whitewood and Genesee coals (Table 3) are comparable to world coals, with a few exceptions. For example, the high concentrations of Cr and Ni in the above three coals are related to high Cr–Ni–Fe oxides (chromite and magnetite) present in the coaly siltstones. The above oxides were derived from ultrabasic diatremes, almost 300 km to the southwest of the mine area, in British Columbia [16].

Boron concentration varies greatly in coals from the Red Deer River valley in the central Alberta Plains (Fig. 2). Boron concentration ranges from 10 to 628 mg/kg, which reflects drastic changes in water chemistry during the deposition of the coal. The study of Gentzis and Goodarzi [18] showed that there is a decrease in B concentration from bottom to top of the 15-seam succession. The lower coal seams were deposited under strongly brackish water because of their stratigraphic position in relation to the formation underneath (Bearpaw Formation) which was deposited in a marine environment. As the depositional environment changed from marine to deltaic (lower to upper delta plain) and finally to freshwater (interdistributary bays, floodplain), boron concentration decreased accordingly. Boron concentration also decreased with increasing inertinite content in the coal (a maceral group) from bottom to top of the sedimentary succession [18]. Goodarzi and Swaine [14] were able to correlate lateral changes in B concentration of one of the coal seams in the area to changes in depositional environment over a distance of almost 17 km. The results of Pollock et al. [16] also suggest that as the inertinite content in the Genesee coals increased from cycle I to cycle II (bottom to top of succession) the concentration of boron decreased in the coals. Therefore, there appears to be a negative relationship between primary boron concentration and inertinite content in coal.

Coals from the Vesta Mine (Fig. 2) are also enriched in boron (200–215 mg/kg; mean is 208 mg/kg) (Table 3), indicating interaction of the coal with brackish waters during the early stages of coalification. The Vesta coals were deposited along the western margin of the Bearpaw Sea, most likely in a fluviodeltaic setting [19], therefore, no secondary enrichment of B is likely. The Vesta Mine coals have a Ba concentration of 450 mg/kg [20], whereas the milled coals that are fed to the Battle River power

station have B concentration of 550 mg/kg [21]. Coals from the nearby Paintearth Mine (Fig. 2) have mean B concentration of 237 mg/kg (Table 3). The same is true for the Montgomery Mine coals (Fig. 2), which have a mean B concentration of 291 mg/kg (Table 3). These values are higher than the mean concentration in world coals (200 mg/kg). The only other element that is present at high concentration is Sb (3.3 mg/kg vs. 1 mg/kg in world coals); this element is not considered to be of environmental concern or interest.

Boron concentration in the Obed Mountain coals located at the boundary between the Alberta Plains and Foothills (Fig. 2) ranges from 27 to 100 mg/kg (mean is 50 mg/kg; Table 3), indicating fresh water to slightly brackish water conditions during peat deposition. This is corroborated by evidence from sedimentology and organic petrology, which points to coal deposition in a distal fluvial floodplain [22]. The recent study of Gentzis and Goodarzi [22] showed that in three vertical sections through two coal seams, boron is enriched in the coal intervals and is systematically depleted in the sedimentary partings above the coal (12–29 mg/kg). The interpretation given is that B, a soluble element, is mobilized by groundwater and carried from the shale and siltstone partings downward where it is assimilated in the coal organic matrix. Another example of very low B concentration in coals deposited under fresh water conditions is the Hat Creek subbituminous coals from intermontane British Columbia (Fig 1) [23]. Boron concentration averages 40 mg/kg (Table 3). These Eocene-age coals have been deposited under freshwater lacustrine conditions in an intermontane graben that was created by block faulting.

2.3. Effect of spontaneous combustion (self-burning)

Coal seams are susceptible to self-burning as a result of an external source of ignition, such as forest fire or lightning strikes. Information on elemental mobilization during self-burning of coal seams is scarce. Goodarzi [24] published information on the variation of elements in a self-burning high-volatile bituminous C coal seam of Paleocene age from Coalspur, Alberta (Fig 2). He reported on the vertical distribution of major, minor and trace elements in three clearly identifiable zones (carbonization, combustion and oxidation from bottom to top) within an 80-cm thick interval. The elements Fe, Mg, Ca and Mn were concentrated in the lower half of the distillation zone, where temperatures reached 500–550°C. There was a noticeable decrease toward the upper part of the carbonization zone (300°C) before their concentration increased drastically in the combustion zone (600–650°C). This behavior is anticipated based on models from coal power plants; the above elements do not volatilize during coal combustion but are concentrated in the bottom ash and fly ash [25].

Elements that are associated with the clay mineral matter in coal, such as Al, Cs, K, Rb, Ti and the REE, showed a variation that correlated well with ash variation. These elements which were concentrated in the lower half of the carbonization zone but were depleted considerably in the upper part of the carbonization zone just beneath the combustion zone. Their concentration again increased in the combustion and oxidation zones because of clay dehydration in the hottest part of the coal seam. Concentrations of

the halogens Br and Cl decreased steadily throughout the carbonization zone before increasing slightly in the combustion zone. These two elements are highly volatile and are present in the vapor phase during coal combustion. Goodarzi [24] attributed the high concentration of Cl at 650°C to the nonporous and compact nature of the combustion chars from this non-coking coal. Chlorine became depleted again in the oxidation zone. Unlike the porous coke that allows volatiles to escape, the blocky and compact char formed from this low rank coal trapped the volatiles (and the halogens) and prevented their escape. The variation of other elements (As, Mo and Sb) was not clear. These elements showed a gradual increase in concentration within the combustion zone following depletion in the upper part of the distillation zone.

2.4. *Effect of igneous intrusions*

Although the effects of igneous intrusions on the organic matter in coal have been studied thoroughly, very little information exists in the published literature on the effects of intrusive bodies on the inorganic geochemistry in coals. Goodarzi and Cameron [26] reported on the trace element distribution in thermally altered coals from the Telkwa deposit in north-central British Columbia (Fig. 1). They found that Br, Cl, H, O and N were susceptible to depletion and were absent in the coal seam at a distance of 65 cm from the intrusion contact. The elements Ba, Cr, K, Sc, V and Zn showed a sudden increase in concentration in the coke zone near the intrusion and became progressively depleted away from the intrusion. The above elements, along with Br and Cl were released because of rapid volatilization of the coal. They were mobilized and trapped in the porous coke at some distance from the intrusion. The elements As, Fe, Mo, S and Sb showed a sudden increase in concentration in the coke at a distance of 25 cm from the intrusion and were associated with sulfides. The elements Ca, Cu, Mg, Mn and Sr were enriched in the coke, while the elements Br, Cl, Hf, Fe, Mo, Rb and Se were depleted because of their volatilization. Calcium and Mg showed the highest concentration at the intrusion contact, indicating an introduction of these elements from the alkali basaltic (an extrusive igneous rock having large proportions of alkalis of Na and K) dike into the coke. The relative enrichment of Cr, Co, Cu, Mg, Mn, Na, Sr and V at the dike contact was the result of elemental input from the basalt into the pyrolyzed residues.

The recent study of Finkelman et al. [27] on a western US bituminous coal showed that the volatile elements, Cl, F, Se and Hg, were not depleted, as expected, in the zone close to the dike contact that experienced the highest temperatures. Rather, they were enriched following volatilization of the elements inherent in the coal by secondary processes. The uniform distribution of Na, B and Cl was attributed to equilibration with groundwater, whereas the high concentrations of Ca, Mg, Fe, Mn, Sr and CO₂ in the coke region were attributed to the reaction of CO with CO₂ generated during coking of the coal with fluids from the intrusion. High As, Hg, Cu, Zn and Fe concentrations in the coke zone were most likely the result of sulphide precipitation. Numerous elements were concentrated at the contact between coke and metamorphosed coal, such as Ga, Ge, Mo, Rb, U, Al, K, Si and the REE, all of which are known to be associated with clays or with organic matter in coal.

2.5. *Effect of weathering*

The study of Goodarzi [28] on a suite of fresh and weathered coals from Fording Mine, southeastern British Columbia (Fig. 1) showed that the concentrations of the halogens Br and Cl were lower in the weathered coals. In contrast, concentrations of Al, As, Ca, Cr, Fe, Mn, Rb, Sc, U, V and Zn increased in the weathered coals. Other elements such as Mg, K, Ta, Th and Ti were higher in the fresh coal, which is not in accordance with the anticipated behavior. It is suspected that the action of surface water has removed these elements from the severely weathered coals in the area.

It is generally accepted that the REE in sedimentary rocks do not undergo fractionation during weathering because of the uniformity in their chemical composition [29]. During weathering of source rocks, the behavior of REE depends on pH, Eh and the presence of organic and inorganic ligands [30]. Fractionation of REE into resistate HREE (heavy) and hydrolysate LREE (light) takes place in acidic coal-forming environments [31]. Eskenazy [32] also stated that coalification can result in the fractionation of REE, causing an anomaly in Ce and Eu. A negative Eu anomaly is usually indicative of the presence of kaolinized volcanic ash deposits (tonsteins) [33]. Therefore, certain patterns in REE distribution in coal seams may provide an indication about the extent of weathering or provenance of the REE.

2.6. *Impact of elemental and radionuclide concentrations on coal utilization in Alberta*

This section deals with the utilization potential of Alberta subbituminous coals for power generation, in terms of their elemental and radionuclide concentrations and potential effect on the environment. The Alberta coals will also be compared with other world coals so that the reader appreciates the “cleanness” of coals from Alberta.

Alberta subbituminous and high volatile bituminous coals are low in sulfur (S content is 0.24–0.36 wt.%), all of which have been described in earlier sections. The values of Th and U in the low ash (< 20 wt.%) coal samples at the minesites range from 1.3–4.4 mg/kg to 0.4–1.8 mg/kg, respectively (Table 4). The concentration of these two elements in the higher ash samples (22–88 wt.%) in the minesites is 3.1–14 mg/kg for Th and 1.6–5.2 mg/kg for U (Table 4). These concentrations are within the range of other Canadian coals and coals from the United States that contain < 20 wt.% ash (1–4 mg/kg) [1]. Thorium in coal is associated mainly with the minerals monazite, xenotime and zircon, which are found in higher concentrations in roof rocks, partings within the coal seams, and in tonsteins (altered volcanic ash layers) [34]. In contrast, U is mostly associated with coal macerals [1] but it can also be present in various minerals [35]. Swaine [1] stated that “radioactivity from coal burning is generally only a small fraction of background radiation and is below permissible units; it would be unwise to burn coal with more than 30 mg/kg of U”. The results presented in Table 4 clearly show that the U content for low-sulfur Alberta coals is within the normal range and it is not expected that emissions of Th and U from coal-fired power stations in Alberta would alter the background radioactivity in the vicinity of the stations.

The average concentration of U and Th in milled coals (coals mixed at various ratios from numerous seams that comprise the feed to a particular power station) in Alberta is

Table 4
Concentration of thorium (mg/kg) and uranium (mg/kg) in subbituminous coals from Alberta

		Obed Mt.	Whitewood	Highvale	Vesta	Paintearth	Montgomery
<i>Low ash</i>							
Ash	Range	2.9–19.6	7.0–19.9	7.0–18.8	5.0–16.4	7.1–18.4	4.7–17.4
	Average	9.7	15.3	12.6	8.2	11.8	10.7
Thorium	Range	0.2–3.2	2.4–12.5	1.1–6.8	0.4–5.3	0.6–6.3	0.7–10.8
	Average	1.3	4.4	3.5	1.3	3.1	4.1
Uranium	Range	0.1–2.7	0–3.7	0.7–3.3	0.9–3.0	0.4–4.1	0.4–9.3
	Average	0.7	1.5	1.5	0.4	1.6	1.8
<i>High ash</i>							
Ash	Range	24.8–70.2	20.7–37.5	22.6–33.9	22.0–81.3	32.8–83.8	2.3–13.8
	Average	36.6	25.8	28.9	42.5	47.1	36.2
Thorium	Range	1.3–5.2	3.5–12.5	4.4–20.2	5.1–12.3	5.1–19.4	2.3–13.8
	Average	3.1	6.4	8.7	8.8	14	6.1
Uranium	Range	0.3–2.9	0–5.9	1.3–8.1	0.04–7.1	1.3–7.5	1.3–5.7
	Average	1.6	2.7	3.2	3.5	5.2	2.8

Low ash (< 20 wt.%), high ash (> 20 wt.%)

1.5–2.7 mg/kg and 3.7–5.7 mg/kg, respectively [36]. These values are low to moderate compared to world coals (0.5–10 mg/kg) [1]. Both U and Th show little variation in milled coals in Alberta, and both concentrations compare well with the range for other Canadian milled coals (U = 1.18–3.2 mg/kg; Th = 2.3–5.8 mg/kg) [37] and US milled coals [38]. The low-sulfur Alberta subbituminous coals have lower concentrations of U and Th than all geological material except diabase (a mafic igneous rock). Their concentration in milled coals is lower than in soils, in shale [1], and also lower than the Earth's crust or granite [31]. The Alberta coals have almost nine times less Th than granite and two times less than shale.

In a comprehensive study of the chemical characteristics of milled coals from the Vesta Mine that feeds the Battle River power station, Goodarzi [21] showed that most elements of “prime environmental concern” are low in concentration compared to mean concentrations in western Canadian or world coals. The same was true for elements of “environmental interest”. During a 3-day “snapshot” study, it was shown that the electrostatic precipitators (ESPs) removed most of elements of prime environmental concern, such as Cr, Co, Mo, Ni, Pb, U, Th, V and Zn. Other elements that do not condense on the ESP fly ash (e.g., Br, Cd and Hg) were found in the hopper close to the stack, which collected the finer fly ash particles. The above study also calculated that the total emissions of hazardous air pollutants (Cd, Cr, Hg, Mo, Ni, Pb and Se) was much less than the 25 t/yr for all these elements allowed by the US EPA or the 10 t/yr allowed for individual elements [21]. More specifically, total emission of toxic to humans heavy metals (Cd, Hg, Pb, Se) at Battle River was only 0.05 t/yr. Regarding other elements of environmental concern (Be, Ba, Co, Cr, Mo, Mn, Ni, V), the total amount emitted was estimated to be 15.5 t/yr. For B, it was about 15 t/yr. These emission rates are very low and clearly demonstrate how “clean” these coals are and how efficient the ESPs are in capturing the fine particulates.

The concentrations of U and Th remained low in the ashes (bottom and fly ash) of the Vesta Mine feed coal [21]. It was estimated that 2.6 t/yr of U and 6.4 t/yr of Th were emitted from the stack. Furthermore, the activities of radionuclides in the milled coals and in coal ashes were within the world average values. The activities of ^{235}U and ^{40}K were below detection limit, while those of ^{226}Ra , ^{210}Po and ^{212}Pb increased from milled coal to stack gas [21]. The total emissions of radionuclides of the U and Th series were estimated to be 17 kg/yr. Finally, the emissions of polycyclic aromatic hydrocarbons (PAHs) from the power station were low, and only three PAHs were detected at low concentrations in the stack gas (phenanthrene, fluoranthrene and pyrene) [21].

The total emissions of heavy metals toxic to humans from the Genesee power station were 0.09 t/yr [36]. High concentrations of Cr (9–2620 mg/kg) and Ni (1–1440 mg/kg) were measured in non-coaly intervals at the Genesee Mine [16]. However, the average Cr concentration in the milled coals was only 11.6 mg/kg and that of Ni is 6.3 mg/kg [36]. The non-coaly intervals in the minesite were removed by selective mining, which reduced the overall concentration of these toxic metals considerably. The above study also estimated that Cr and Ni had the highest emission rates of any toxic metal (595 and 753 kg/yr), followed by Hg (34 kg/yr) and Cd (15 kg/yr). The above values are far below the permitted rates by the US EPA (10 t/yr for individual elements) [36], indicating that the Genesee coals are “clean” by world standards. Other elements of environmental interest (Be, Ba, Co, Mn, Mo, V and Zn) are also emitted at low rates (total of 2.7 t/yr) [36].

Furthermore, Goodarzi [36] estimated that 4.9 t/yr of U and 12.9 t/yr of Th are emitted from the Genesee power station, well within the world average range. The emission of all radionuclides of U and Th series is about 12 kg/yr, one third of which is due to the U series. These emissions are very low and numerous radionuclides were below detection. As Goodarzi stated characteristically, “background radiation from natural sources, such as cosmic radiation for K, Th and U and radon and its daughter products is higher than the input from the Genesee station”. Finally, only two PAHs were detected at low concentrations in the stack gas (phenanthrene and 2-methylfluorene) [36].

Finally, Gentzis and Goodarzi [39] demonstrated that the ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ in coals is an important indicator of the performance of electrostatic precipitators (ESPs) installed in a modern power station. The above ratio is dependent on the nature of clays in the coal (e.g. kaolinite that has a low ratio or montmorillonite that has a high ratio). Again, this shows the importance of the type of country rocks that contributed to the input of detrital material and the weathering pattern. It is known that major oxides (CaO , Fe_2O_3 and SiO_2) and others affect the propensity of coal ash to form slag deposits. In the Whitewood Mine coals, high concentrations of CaO (average 10 wt.%) was attributed to the recent deposition of the mineral gypsum on the highwall surface of the mine from groundwater discharge.

2.7. Effect of coal utilization on humans

The effect of toxic metals on humans depends not only on its concentration but also on its valence state. Hexavalent Cr (Cr^{+6}) is known to be toxic and causes lung cancer

Table 5

Mean concentrations of selected radionuclides (Bq/kg) in thermal coals from Alberta. Also shown are the ranges for other coals from Canada, Australia, the US, the UK and the world. Data for the Canadian, Australian, US and UK coals compiled from Refs. [21,36]; data for world coals taken from Ref. [40]

	Highvale	Whitewood	Canadian coals	US coals	UK coals	Australian coals	World coals
Radionuclide							
Uranium 238	27	35	13–47	6.0–73	8.0–31	8.5–47	15–20
Thorium 230	BD	BD	20–93	NA	9.0–25	21–68	NA
Thorium 232	20	20	9.0–23	4.0–21	7.0–19	11.0–69	7–110
Lead 210	20	30	12.0–64	12.0–78	NA	20–33	10.0–50
Radium 226	30	40	9.0–36	9.0–59	8.0–22	19–24	1.5–250

BD = Below detection.

NA = Not available.

if inhaled as chromate dust (Cr_2O_4), whereas it is harmless and even essential to mammals when present in the trivalent form (Cr^{+3}) [40]. Deficiency of Cr^{+3} might cause diabetes. Approximately 95% of Cr in the milled coals at the Genesee power station is of the Cr^{+3} type based on XANES spectroscopy [36]. Nickel is found as an alloy in cutlery, which is used on a daily basis. Some Ni compounds are known human carcinogens [e.g., the carbonyl and sulfide compounds or $\text{Ni}(\text{Co})_4$ and NiS , respectively]. None of these two compounds are present in the milled coal or coal ash from the Genesee station. Rather, Ni appears to be predominantly Ni^{2+} in oxygen coordination, based on XANES spectra [36].

Table 5 shows the available data for radionuclides in coals from Australia, Canada, UK and the US. All radionuclides in Alberta coals are below detection and only ^{238}U and ^{232}Th have values above detection limit. Alberta coals have low to moderate concentrations of ^{238}U (27–35 Bq/kg) and ^{232}Th (20 Bq/kg) compared to other world coals. These values are also within the range of Canadian milled coals [37]. The total amount of ^{235}U in the same milled coals is estimated to be 14.8–52.3 kg/yr. The total yearly emission of Th and U radionuclides from Alberta power stations is in the range of 12–70 kg, with ^{235}U representing 0.082–0.227 kg/yr. The greater propensity of Th to cause radioactive stress in the environment is attributed to direct inhalation of ^{232}Th and ^{228}Ra and deposition of ^{232}Th decay chain isotopes.

Radionuclides that affect human organs, such as liver, kidneys, spleen and lungs include ^{226}Ra , ^{228}Ra , ^{210}Pb , ^{210}Po and ^{222}Rn [41,42]. The first two end up in beef, milk and vegetables, while ^{210}Pb , ^{210}Po and ^{222}Rn are inhaled. The study of Goodarzi [36], which used *Sphagnum* moss traps as collection sites of airborne particles in the vicinity of power stations in Alberta demonstrated that deposition of radionuclides was low and no impact by isotopes of Th and U was registered over a period of one year.

3. Summary

The results presented here show that geological processes, whether short-term (igneous intrusions) or long-term (weathering, tectonism, groundwater movement) can

have a pronounced effect on the concentration and distribution of elements in coal seams and coal-bearing strata. Thus, the role of geology is extremely important regarding the utilization of coals for power generation. With respect to the subbituminous coals from Alberta, it can be stated unequivocally that they are “clean” by world standards. Furthermore, the emissions of elements and radionuclides of environmental concern and environmental interest are minimal and far below the allowable rates set by the US EPA and by Environment Canada.

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