

# Development of Rare-Earth Elements Database for the East Kootenay Coalfield of Southeastern British Columbia (NTS 082G/10, 15) Using Field Collected Samples: Preliminary Results

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## Introduction

Rare-earth elements (REEs) are the group of 17 elements in the periodic table that include 15 lanthanides and two chemically similar transition metals—scandium and yttrium. Using their atomic number, REEs are classified as heavy REEs (HREEs) or light REEs (LREEs), with elements from Tb to Lu and Y belonging to the former group and La to Gd and Sc belonging to the latter group (Moldoveanu and Papangelakis, 2013; Zhang et al., 2015). With the emergence of new clean energy and defence-related technologies, consumption of REEs has increased rapidly (Tse, 2011). For example, it is projected that the demand for dysprosium, one of the REEs, is expected to increase by as much as 2600% by 2025 (Standing Committee on Natural Resources, 2014). In addition, traditional rare-earth ore deposits are fast depleting and supply is only expected to meet demand for next 15–20 years (Seredin and Dai, 2012). Based on the projected dwindling supply and increasing demand, REEs are classified as critical elements by the United States and the European Union due to their importance in clean energy and defence applications (U.S. Department of Energy, 2010; Deloitte Sustainability et al., 2017). The National Energy Technology Laboratory (NETL) in the United States has conducted a prospective analysis of coal deposits as a source of REEs using the U.S. Geological Survey's (USGS) coal database, which contains the concentrations of REEs in coalfields across the United States (Bryan et al., 2015). The NETL has launched a research and development program to demonstrate the techno-economic feasibility of developing domestic technologies to separate REEs from coal and/or its byproducts. The study uses samples that contain a minimum of 300 ppm total REEs and concentrating the REEs to a level greater than or equal to 2% (by weight) in processed streams (U.S.

Department of Energy, 2016). The program will focus on areas of research such as resource sampling and characterization, separation technology development, REE sensor development, process and systems modelling, and techno-economic analyses (U.S. Department of Energy, 2016).

There is indication of the presence of REEs in some Canadian coal deposits, especially in British Columbia (BC) coalfields (Goodarzi, 1988; Birk and White, 1991; Goodarzi et al., 2009), however, there is no proper quantification, characterization and extraction analysis currently available for coal deposits in BC or for other coal deposits across Canada. The first objective of this study is to develop a database of REE occurrences in the East Kootenay coalfield (Figure 1), from samples collected in the field, and to identify the best potential coal sources of REEs in the study area. Using the collected data, phase two of the study will explore the possibility of extracting these critical elements from these sources. Some of the initial results of this study were reported previously (Kumar et al., 2018; Kuppusamy and Holuszko, 2019).

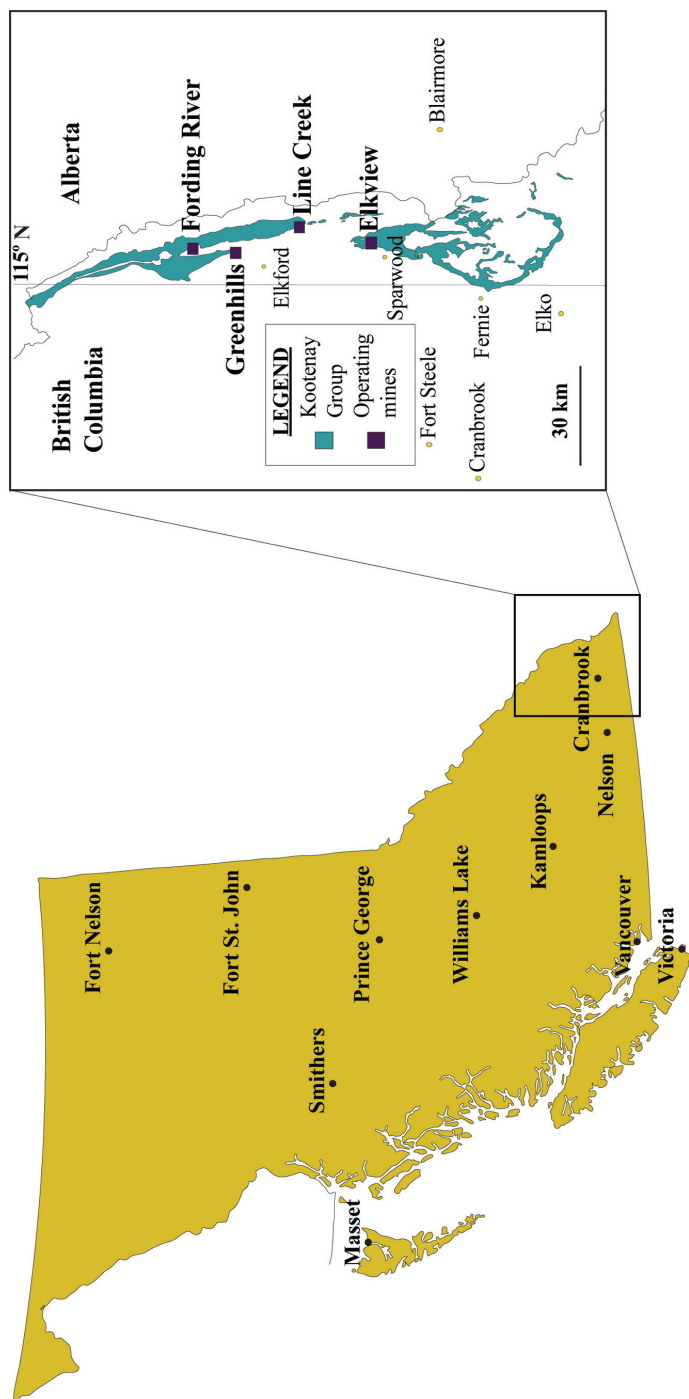
## Background

### Abundance of REEs in Coal

Table 1 shows the average concentration of REEs in coal from different countries, such as the United States, China, the Democratic People's Republic of Korea and Turkey, in comparison to average values for the upper continental crust, black shale and world coal on a whole coal basis. The average REE concentration in world coal on a whole coal basis is 72 ppm (Seredin and Finkelman, 2008; Ketris and Yudovich, 2009), which is 2.5 times lower than the upper continental crust (179 ppm; Taylor and McLennan, 1985) and the average black shale value (182 ppm; Ketris and Yudovich, 2009). The average concentration of REEs in Chinese coal is 101–138 ppm (Dai et al., 2008; Zhang et al., 2015) and is 1.5–2 times the world's average. Similar results were observed for Turkish coal (Karayigit et al., 2000;

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**Figure 1.** Location of East Kootenay coalfield and operating coal mines in southeastern British Columbia (adapted from BC Geological Survey, 2019).

**Table 1.** Concentration of rare-earth elements (on a whole coal basis; ppm) in coal from different countries in comparison to concentrations in the upper continental crust and black shale. Abbreviations: HREE, heavy rare-earth elements; LREE, light rare-earth elements; ND, no data; REE, rare-earth elements.

Coal	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu	Sc	Total REEs	LREEs	HREEs
Upper continental crust <sup>1</sup>	30	64	7.1	26	4.5	0.88	3.8	0.64	3.5	22	0.8	2.3	0.33	2.2	0.32	11	179	147	32
Black shale <sup>2</sup>	28	58	4.2	33	5.4	1.2	4.7	0.75	3	26	0.52	1.9	0.4	2.8	0.4	12	182	147	36
World coal <sup>2,8</sup>	11	23	3.5	12	2	0.47	2.7	0.32	2.1	8.4	0.54	0.93	0.31	1	0.2	3.9	72	59	14
American coal <sup>3</sup>	12	21	2.4	9.5	1.7	0.4	1.8	0.3	1.9	8.5	0.35	1	0.15	0.95	0.14	ND	62	49	13
Chinese coal <sup>4</sup>	26	49	5.5	22	4.3	0.9	3.7	0.7	3.1	18	0.7	1.9	0.27	2.1	0.3	ND	138	111	27
Chinese coal <sup>5</sup>	18	35	3.8	15	3	0.5	3.4	0.52	3.1	9	0.73	2.1	0.34	2	0.32	4	101	83	18
Turkish coal <sup>5,6</sup>	21.12	39.24	4.71	16.85	3.18	0.76	3	0.45	2.42	12.76	0.47	1.37	0.21	1.35	0.21	7.92	116	97	19
North Korean coal <sup>7</sup>	14.5	27.2	2.9	11.1	2.3	0.5	1.4	0.3	2	7.2	0.4	1.1	0.3	1	ND	4.9	77	65	12

<sup>1</sup>Taylor and McLennan, 1985; <sup>2</sup>Ketris and Yudovich, 2009; <sup>3</sup>Finkelman, 1993; <sup>4</sup>Dai et al., 2008; <sup>5</sup>Zhang et al., 2015; <sup>6</sup>Karayigit et al., 2000; <sup>7</sup>Hu et al., 2006; <sup>8</sup>Seredin and Finkelman, 2008

Zhang et al., 2015). But the average concentration in coals from the United States and Democratic People's Republic of Korea is close to the world's average (Finkelman, 1993; Hu et al., 2006). The average REEs concentration in world coal on an ash basis is 404 ppm (Seredin and Dai, 2012). However, coal with enriched concentrations of REEs on an ash basis have been found in different coalfields such as in the Sydney Coal Basin, Nova Scotia, Canada (72–483 ppm; Birk and White, 1991), the Far East, Russia (300–1000 ppm; Seredin, 1996) and the Fire Clay coal bed, central Appalachian basin, United States (500–4000 ppm; Hower et al., 1999). Approximately 80% of the total concentration of REEs in coal is LREEs (Zhang et al., 2015). The anomalies of REE enrichment in coal and REE distribution patterns are discussed in detail elsewhere (Dai et al., 2016).

Some of the enriched coal (0.68 to 2.03% of rare earth oxide) has REE concentrations comparable to traditional commercial rare-earth deposits and coal ashes from coal mining operations can be viewed as a potential source for these elements (Seredin and Dai, 2012).

### U.S. Geological Survey's COALQUAL Database

During the 1970s energy crisis, the USGS developed the National Coal Resources Data System (NCRDS), which is a comprehensive database on coal resources in the United States (Finkelman et al., 1994). The USCHEM, a coal quality geochemical database for the United States, is an interactive digital version of the NCRDS, which contains information on more than 13 000 samples from major coal basins in the United States (Palmer et al., 2015). The COALQUAL database, which contains a subset of 7430 samples from USCHEM database, was published in 1994 (Bragg et al., 1994). For each sample, 136 parameters were collected including coal type, proximate analysis data, ultimate analysis data, and major-, minor- and trace- elements analysis data (Finkelman et al., 1994). Figure 2 shows an example of a COALQUAL sample detail page. In 2015, the updated version (3.0) of the COALQUAL database was published with 7657 samples (Palmer et al., 2015). Using the original database as a guideline, further investigations were carried out to characterize and physically enrich the REE from coal-related feedstocks (e.g., produced coals, coal waste, coal ash from power plants) in the United States (Akdogan and Ghosh, 2014; Honaker et al., 2015; Miskovic, 2015; Soundarrajan et al., 2015).

### Objectives

The purpose of this investigation is to assess BC coal deposits as possible sources for extraction of REEs. The assessment will be performed by creating a database of REE distribution in the East Kootenay coalfield; it will be a simplified version of the USGS COALQUAL database. The

first phase of the project will be the development of a database, which starts with the collection of roof, floor, partings and coal samples in the study area. These samples will be characterized and analyzed by inductively coupled plasma–mass spectrometry (ICP-MS), sequential extraction, X-ray diffraction (XRD) and scanning electron microscope (SEM). Phase two of the project will focus on the advanced characterization of the REEs and development of the physical methods used to enrich the REE from the coal-related feedstocks.

## Materials and Methods

### Sampling

For the purpose of developing the REE database, coal seam (referred to as coal) samples along with their corresponding roof, floor and partings samples were collected from the East Kootenay coalfield and shipped in barrels to the Coal and Mineral Processing Laboratory at The University of British Columbia (Vancouver, BC). All the samples used in this study were collected by geologists from their respective mines to enhance the quality of the collected samples. Representative samples were obtained for further testing using the standard procedure for coal sample preparation—ASTM D2013/D2013M-12 (2012). Proximate analysis was conducted in duplicate on the representative samples using the standard methods—ASTM D3172-13 (2013), D3173/D3173M-17a (2017), D3174-12 (2012) and D3175-17 (2017).

### Chemical Analysis of REEs and Other Minor Elements


For total REE quantification, feed and test product samples (0.2 g) were added to lithium metaborate and lithium tetraborate flux and mixed thoroughly. The samples were then fused in a furnace at 1025°C. Finally, resulting melts were cooled and digested in an acid mixture containing nitric, hydrochloric and hydrofluoric acids. The digested solutions were then analyzed by ICP-MS. The analytical results were corrected for interelemental spectral interference. In this study, REEs in coal are expressed as follows: on a whole coal basis (REE concentration in the coal sample) and on an ash basis (REE concentration in the ash of the coal sample).

Base metal and other minor elements were analyzed by four-acid digestion followed by inductively coupled plasma–emission spectroscopy (ICP-ES). For a few highly reactive samples, aqua-regia digestion was used for the analysis. All the chemical analysis was conducted by ALS-Geochemistry (North Vancouver, BC).

## Results and Discussions

Since more samples are currently being collected for the study, this paper only contains preliminary results based on



 <b>COALQUAL Database</b>							
Sample Detail for W218790							
View <a href="#">Definitions of Qualifiers and Parameters</a>							
Sample Description		Proximate & Ultimate		Oxide		Trace Element	
Sample ID	W218790	Sample ID	W218790	Sample ID	W218790	Sample ID	W218790
State	Kentucky	Moisture	3.53	Remnant Moisture	1.23	GS Ash Dry	4.15
County	CLAY	Moisture Q		Remnant Moisture Q	<a href="#">r</a>	GS Ash Dry Q	
Latitude	37.2203	Volatile Matter	39.99	GS Ash	4.1	Si	9550
Longitude	-83.8561	Volatile Matter Q		GS Ash Q		Si Q	
Province	EASTERN	Fixed Carbon	52.34	SiO <sub>2</sub>	49.2	Al	7180
Region	CENTRAL APPALACHIAN	Fixed Carbon Q		SiO <sub>2</sub> Q	<a href="#">Q</a>	Al Q	
Field		Standard Ash	4.14	Al <sub>2</sub> O <sub>3</sub>	32.7	Ca	490
District	SOUTHWESTERN	Standard Ash Q		Al <sub>2</sub> O <sub>3</sub> Q	<a href="#">Q</a>	Ca Q	
Formation	BREATHITT	Proximate Validation	Acceptable	CaO	1.65	Mg	191
Group		Hydrogen	5.38	CaO Q	<a href="#">Q</a>	Mg Q	
Bed	MANCHESTER	Hydrogen Q		MgO	0.764	Na	286
Member		Carbon	77.2	MgO Q	<a href="#">Q</a>	Na Q	
Coal Zone		Carbon Q		MnO	0.0057	K	436
Depth (in)	0	Nitrogen	1.8	MnO Q		K Q	
Thickness (in)	9.4	Nitrogen Q		Na <sub>2</sub> O	0.932	Fe	951
System	Pennsylvanian	Oxygen	7.39	Na <sub>2</sub> O Q	<a href="#">Q</a>	Ti	370
Series/Epoch		Oxygen Q		K <sub>2</sub> O	1.26	Ti Q	
Literature		Sulfur	0.56	K <sub>2</sub> O Q	<a href="#">Q</a>	TS	
Comments		Sulfur Q		Fe <sub>2</sub> O <sub>3</sub>	3.27	TS Q	<a href="#">B</a>
Map	MANCHESTER (7.5")	Ultimate Validation	Excellent	Fe <sub>2</sub> O <sub>3</sub> Q	<a href="#">Q</a>	Ag	0.0145
Collector	KYGS-CURRENS J C	Btu	13915	TiO <sub>2</sub>	1.49	Ag Q	
Mine/Power Plant	SURFACE MINE	Btu Q		TiO <sub>2</sub> Q	<a href="#">Q</a>	As	0.506
Drill Core No		Sulfate Sulfur	0.01	P <sub>2</sub> O <sub>5</sub>		As Q	
Point Id	KGS 698	Sulfate Sulfur Q		P <sub>2</sub> O <sub>5</sub> Q	<a href="#">B</a>	Au	0.29
Submit Date	12/16/1982	Pyritic Sulfur	0.03	SO <sub>3</sub>		Au Q	<a href="#">L</a>
Sample Description	BITUMINOUS COAL	Pyritic Sulfur Q		SO <sub>3</sub> Q	<a href="#">B</a>	B	17.4
Estimated Rank	BITUMINOUS	Organic Sulfur	0.52	LOI		B Q	
Apparent Rank	High volatile A bituminous	Organic Sulfur Q				Ba	15.4
Analytical Labs	GT and USGS	Ash Deformation	2800			Ba Q	
Sample Type	Channel	Ash Deformation Q	<a href="#">G</a>			Be	2.78
Analysis Type	As Received	Ash Softening	2800			Be Q	
Values Represent	Single sample	Ash Softening Q	<a href="#">G</a>			Bi	0.42
Township		Ash Fluid	2800			Bi Q	<a href="#">L</a>
Range		Ash Fluid Q	<a href="#">G</a>				

**Figure 2.** Example of a 'Sample Detail' page from the U.S. Geological Survey's (USGS) COALQUAL database (Palmer et al., 2015).

the early sample data from 49 samples. Results of the proximate analysis of the coal samples are shown in Table 2. According to ASTM D388-17 (2017), all the coal samples are classified as medium volatile bituminous coal, which is largely a metallurgical quality grade. For the database, more than 60 parameters were collected for each sample including type, proximate analysis results and major-, minor- and trace-elements data. The complete dataset will be released in the future. Seam ID and the specific locations of the individual samples are not supplied to maintain a confidentiality agreement.

Among the samples analyzed, the REE concentration on ash basis varies from 137 to 686 ppm. Table 3 shows that REE concentrations on ash basis are similar for roof, floor and partings material, whereas coal showed enhanced concentrations of REEs. Among the 17 REEs, the five elements of Ce, La, Nd, Y and Sc accounted for more than 80% of the total REE present in these samples. Figure 3 shows that on a whole coal basis, however, the concentration of REE increases with the ash content of the material. This

indicates the association of REE with mineral matter, which is comparable with previously published results (Kuppusamy and Holuszko, 2019). During the coal beneficiation process, the REEs associated with mineral matter are generally concentrated into waste tailings streams.

### Preliminary Economic Evaluation for BC Coal Samples

Seredin and Dai (2012) estimated that a coal seam with a thickness greater than 5 m may be considered as a potential source of REEs if its rare-earth oxide content is above 800–900 ppm on an ash basis. Zhang et al. (2015) estimated the minimum average REE content to be 720 ppm on an ash basis, assuming a valence state of +3 for all of the REEs and a relative atomic mass of 132.5. In the United States, coal seams with an REE content of more than 300 ppm on ash basis are considered a potential source of REEs (U.S. Department of Energy, 2016).

It can be inferred that the REE concentration in the feed samples (see Figure 4) did not meet the cutoff grade as pro-

**Table 2.** Proximate analysis results for coal samples (as-determined basis) from the East Kootenay coalfield, southeastern British Columbia.

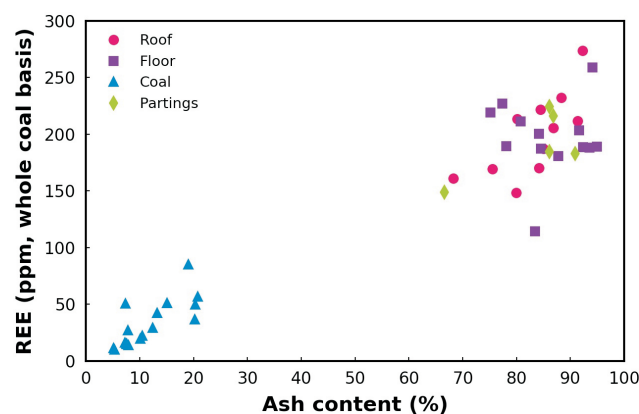
Sample ID	Type	Moisture content (%)	Ash content (%)	Volatile matter (%)	Fixed carbon (%)
10	Coal	2.41	7.24	22.85	67.5
11	Coal	2.81	10.12	21.92	65.14
12	Coal	2.39	7.66	23	66.95
13	Coal	2.41	7.52	22.92	67.14
14	Coal	2.43	7.93	23.02	66.62
15	Coal	2.5	10.46	22.3	64.74
16	Coal	2.62	5.1	24.08	68.2
17	Coal	2.1	5.29	23.69	68.93
18	Coal	2.09	20.24	27.29	50.38
19	Coal	2.75	7.37	25.68	64.2
20	Coal	1.36	12.39	24.67	61.59
22	Coal	0.85	19.05	19.39	60.71
26	Coal	1.25	7.84	22.73	68.19
27	Coal	1.24	13.19	19.32	66.26
41	Coal	1.31	20.74	23.11	54.84
48	Coal	0.66	20.34	20.15	58.85
49	Coal	0.87	15.06	22.16	61.91

**Table 3.** Maximum, minimum and average rare-earth element (REE) concentrations (on ash basis; ppm) in the roof, floor, coal and partings samples from the East Kootenay coalfield, southeastern British Columbia.

Type	Maximum	Minimum	Average
Roof	296.8	185.7	238.8
Floor	293.3	136.9	230.2
Coal	685.9	172.8	273.5
Partings	260.4	201.2	229.6

**Table 4.** Maximum, minimum and average heavy rare-earth elements (HREEs) to light rare-earth elements (LREEs) ratio and outlook coefficient in the different coal sample types from East Kootenay coalfield, southeastern British Columbia.

Type	HREEs/LREEs			Outlook coefficient		
	Maximum	Minimum	Average	Maximum	Minimum	Average
Roof	0.36	0.20	0.27	1.28	0.83	1.03
Floor	0.33	0.18	0.26	1.22	0.85	1.02
Coal	0.54	0.27	0.36	1.54	0.92	1.21
Partings	0.34	0.21	0.28	1.27	0.82	1.10



**Figure 3.** Rare-earth element (REE) concentration (on whole coal basis; ppm) versus ash content (%) for the roof, floor, coal and partings samples from the East Kootenay coalfield, southeastern British Columbia.

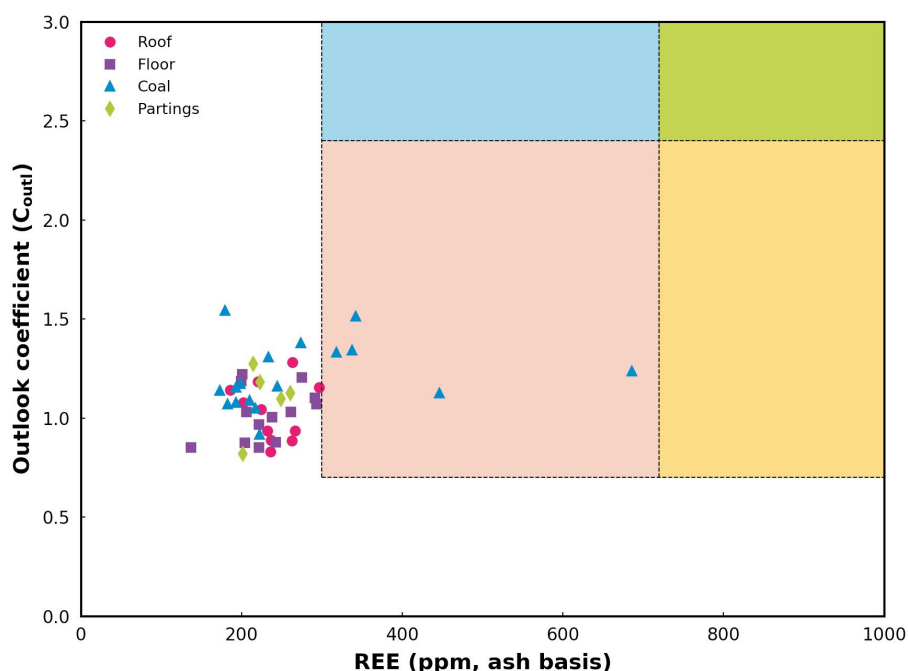
posed by Seredin and Dai (2012), but some of the coal showed significant potential. Using the U.S. Department of Energy's (2016) resource cutoff of 300 ppm of REE on an ash basis, it can be concluded that some of the samples met the cutoff grade requirements.

Outlook coefficient ( $C_{outl}$ ) is another factor that can be used to assess the quality of the REEs present in the coal seam. It is defined as the ratio between the relative amounts of critical REEs in the sample to the relative amounts of excessive REEs in the sample (Seredin and Dai, 2012). It can be calculated as

$$C_{outl} = \frac{\frac{\text{sum concentrations of Nd, Eu, Tb, Dy, Er, Y}}{\text{total REE concentration}}}{\frac{\text{sum concentrations of Ce, Ho, Tm, Yb, Lu}}{\text{total REE concentration}}}$$

A higher value for this index represents a higher market value for the REEs in the coal seam since the concentration of critical REEs increases with the index. Dai et al. (2017) proposed an evaluation plot using REE concentration and  $C_{outl}$ . In this study, a similar plot was used (Figure 4) but modified to accommodate the resource cutoff suggested by the U.S. Department of Energy (2016). Accordingly, all of the samples were divided into five categories: unpromising source (REE <300 ppm on ash basis or  $C_{outl}$  <0.7); promising resource (300<REE<720 ppm on ash basis and 0.7< $C_{outl}$ <2.4); highly promising resource (300<REE<720 ppm on ash basis and  $C_{outl}$  >2.4); promising source (REE >720 ppm on ash basis and 0.7< $C_{outl}$ <2.4); and highly promising source (REE >720 ppm on ash basis and  $C_{outl}$  >2.4).

The  $C_{outl}$  values for most of the samples are greater than 1, implying that the critical REE concentration is significant and accounts for,



**Figure 4.** Outlook coefficient ( $C_{outl}$ ) versus rare-earth element (REE) concentration for various types of coal samples from the East Kootenay coalfield, southeastern British Columbia. The REE resource categories for coal sources: green, highly promising source; yellow, promising source; blue, highly promising resource; light salmon: promising resource.

on average, 35% of the total REEs. Further, it can be noticed from Table 4 that HREE concentrations are more significantly concentrated in coal, in some cases they total more than 50% of total LREEs, than in roof, partings and floor materials, which contributes to the better  $C_{outl}$  values. The average  $C_{outl}$  for the coal samples is found to be 1.2. Since the reported  $C_{outl}$  for world coal is 0.64 (Zhang et al., 2015, Dai et al., 2017), these results show that BC coals may be a viable source of REEs if extraction practices are refined.

Correlation analysis also shows a strong correlation between coal ash and REE content (except Sc) when calculated on a whole coal basis ( $r = +0.88$  to  $+0.96$ ). This indicates the presence of mineral phases containing REEs in the coal samples. One of the rare-earth carriers in these types of metalliferous coal is zircon, which can originate from volcanic ash or authigenic minerals and it can be identified by enrichment of Hf, Th, U and HREEs (Finkelman, 1981; Seredin, 2004). A compelling correlation between Hf, Th, U, Y and other REEs implies that zircon is one of the source minerals of REEs and indicates an input of volcanic ash containing REEs into the coal. Elemental analysis results using ICP-MS proved the presence of zirconium in the samples. Also, volcanic ash is believed to be the source of tonsteins associated with the coal beds in the Mist Mountain Formation in the East Kootenay coalfield (Grieve, 1993), which further validates the inference made in this study.

No correlation was observed between ash and REEs content when calculated on an ash basis ( $r = -0.03$  to  $0.18$ ). This points toward an insignificant association of REEs with organic matter in the studied samples. To confirm this, the next step was to look for a strong correlation between REEs and W, which is believed to be organically fixed in coal. However, a very weak correlation was shown between REE and W in the studied samples ( $r = +0.21$  to  $0.34$ ) indicating inorganically associated REEs.

In the samples, on the whole coal basis, REEs strongly correlated with uranium ( $r > +0.84$ ) and thorium ( $r > +0.85$ ). This suggests that one of the REE mineral phases could be monazite, but a more detailed mineralogical study is required to confirm this.

Most of these preliminary results are similar to those previously reported for this study (Kumar et al., 2018).

## Conclusions

One of the main objectives of the study is the development of a rare-earth element (REE) database for the East Kootenay coalfield. In this regard, coal samples from the East Kootenay coal deposits were tested for the presence of REEs and it was found that total REE concentrations on ash basis varied from 137 to 686 ppm. It was inferred from the data that REEs in the coal samples are associated with both organic and inorganic portions of the coal constituents, but inorganic association seems to be dominant. The information presented in this paper represents only the preliminary

data and the final database will be reported on in future reports. As part of phase two of the study, a few select samples will be identified and detailed characterization of the REEs will be undertaken. In addition, REE enrichment of the coal using physical separation will be tested.

With the development of extraction techniques, REEs could be extracted from coal as byproducts of coal mining, which can strengthen brownfield operations by increasing profitability and possibly generating green credits, as these REEs are used in clean energy technologies. In the case of greenfield operations, REE extraction will increase the economic viability of the deposit and the potential for its future development. Overall, it will enhance British Columbia's coal resources.

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