RARE EARTH ELEMENT CONCENTRATIONS IN COAL AND ASSOCIATED SEDIMENTS IN CENTRAL AND EASTERN MONTANA: YEAR 1 COAL ASSAY RESULTS AND INTERPRETATION

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Front photo: Several coalbeds in the Cedar Creek Anticline, Eastern Montana.

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ABSTRACT

The state of Montana has significant coal fields and a long history of coal production dating back to the late 19th century. Until recently, the rare earth element potential of these coal fields had never been fully studied. The data presented in this paper are from the first-year findings of a 5-year study on rare earth element concentrations in Montana coal and associated sediments. During the first year, 262 samples were collected from the Fort Union Formation (Paleocene) and Hell Creek (Cretaceous) formations in central and eastern Montana. These samples were analyzed by a lab (on a whole-coal basis) for rare earth elements and other critical minerals. Rare earth concentrations in the Fort Union ranged from 5 to 501 parts per million (ppm), and averaged 80 ppm. Rare earth concentrations in the Hell Creek ranged from 23 to 251 ppm, and averaged 77 ppm. The rare earth concentrations in all associated non-coal sediments ranged from 18 to 394 ppm and averaged 101 ppm. The highest concentrations of rare earths tended to occur at the base of coalbeds; however, a more robust sample number is needed for any definitive patterns.

INTRODUCTION

The occurrence of rare earth elements (REE) in coal deposits has drawn the attention of researchers around the world, as technology continues to escalate the demand for these unique metals. Coal underlies over 35% of the state of Montana and generally increases in maturity east to west across the State (lignite–subbituminous–bituminous; fig 1). Major deposits occur in the late Jurassic (163–145 Ma), Cretaceous (145–66 Ma), and Tertiary (66–2.6 Ma). Historically, coal in Montana has been mined for energy purposes, focusing on the Tertiary deposits (Fort Union Formation) because these coalbeds are thick and accessible for strip mining. Coals from all three time periods occur in outcrops that generally decrease in number east to west and south to north. Locating outcrops of coal in northern Montana is especially difficult due to the glacial sedimentary cover left by the Laurentide Ice Sheet advances during the Pleistocene Epoch (2,600,000 to ~12,000 years ago).

Coal is mostly carbon, but also contains hydrogen, sulfur, oxygen, nitrogen, and numerous additional elements, including REE. The concentration of REE within coal is often highly variable, but has nonetheless generated interest in the possibility of

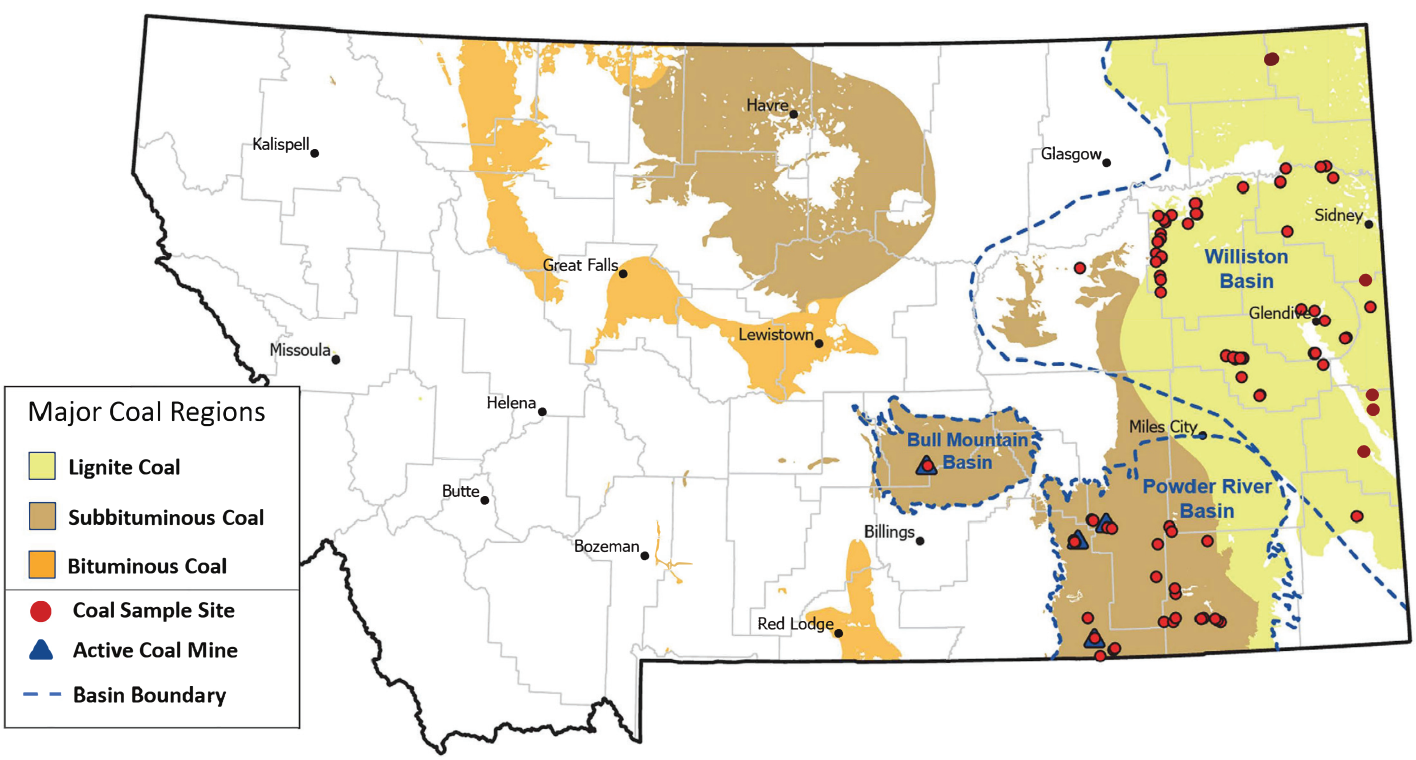

Figure 1. Map of Montana coal regions (modified from Cole and others, 1982).

mining coal for these elements. This paper explores the potential of REE in coal and coal-related materials across Montana.

Rare earth elements are a group of 15 lanthanides plus two transitionary metal elements, Y and Sc. When Y but not Sc is included in a rare earth study, it is referred to as REY (REE + Y). Geochemically, the behavior of Y mirrors heavy lanthanides, whereas Sc is a much smaller cation and exhibits a different geochemical behavior than other rare earths (McLennan, 1989). Therefore, Sc is not included in the main discussion of this paper and only briefly mentioned due to its high economic value. The REY in this study are further split into two groups based on atomic weight: light rare earth elements (LREY: La, Ce, Pr, Nd, Sm, and Eu) and heavy rare earth elements (HREY: Gd, Tb, Dy, Y, Ho, Er, Tm, Yb, and Lu).

GEOLOGY

The Fort Union Formation is a Tertiary (66–58 Ma) formation that covers approximately 35% of the State and is the primary source of commercial coal (fig. 2). It is divided into three members (from youngest to oldest): the Tongue River, Lebo (or Lebo Shale), and Tullock. The Tongue River is characterized by yellowish orange sandstone, siltstone, and carbonaceous shale, and contains Montana's primary economic coal deposits. The Lebo is characterized by dark gray carbonaceous shale, claystone, sandstone, and coal. Coal deposits in the Lebo are much less abundant than coalbeds in either the Tullock or Tongue River. The Tullock is characterized by yellow sandstone interbedded with grayish brown and black shale and thin coalbeds. The Hell Creek Formation is a Late Cretaceous (68–66 Ma) formation that covers a large portion of central and eastern Montana. The coals in this formation are generally thin and dirty, and not considered economically viable for mining. No operating mines exist in the Hell Creek Formation, or in the Lebo and Tullock Members of the Fort Union Formation.

Coalbeds sampled in year 1 were collected from three major basins in central and eastern Montana: the Powder River, Williston, and Bull Mountain Basins (see fig. 1). Historically, most coal mining has taken place in the Powder River Basin. The basin consists of thousands of feet of clastic sediment, including coalbeds formed in long-lived swamps that make the Powder River Basin the primary coal producer in the State. Today, there are three surface mines still producing subbituminous coal in the basin: the Absaloka, Rosebud, and Spring Creek. Coalbeds in the Williston

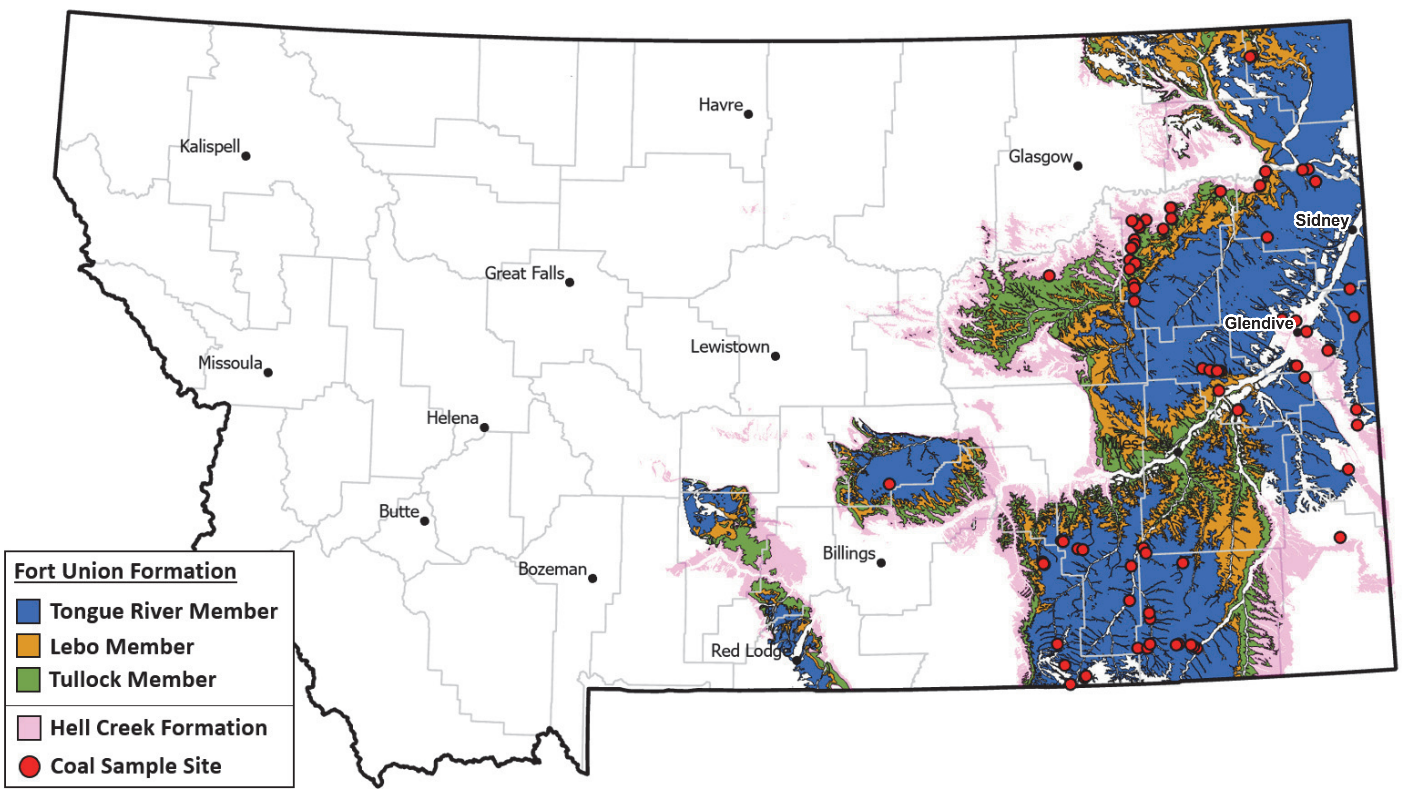

Figure 2. Map of geologic formations sampled.

Basin are not as thick and numerous as they are in the Powder River. They are laterally discontinuous and become sparse to the north, although some can be quite thick. The 15- to 25-ft Pust bed was formerly mined at the Savage Mine, located near Savage, Montana. It was the only commercial mine in the Montana portion of the Williston Basin and the only lignite mine in the State. Approximately 45 min north of Billings, Montana, Signal Peak Energy is mining bituminous coal underground in the Bull Mountain Basin. Signal Peak is the only mine in that area.

METHODS

A total of 262 coal samples were collected across central and eastern Montana from the Fort Union and Hell Creek Formations (see figs. 1, 2). A majority of the sample locations were on Federal land. Samples were collected from the following counties: Sheridan, Garfield, McCone, Richland, Dawson, Prairie, Wibaux, Fallon, Carter, Powder River, Rosebud, Bighorn, and Musselshell. Sampling was focused on the eastern part of the State, as it contains numerous coal outcrops and has been the major commercial producer of coal. Moving west across the State, coal exposures become more elusive, and identifying and sampling them will be a focus for subsequent years.

Outcrop sample sites were described, measured, and photographed, and their coordinates recorded. A rock hammer was used to dig into the outcrop approximately 12 in (30 cm) to avoid sampling coal that had been subjected to significant weathering. A reading was taken using a Niton XL-5 Plus portable x-ray fluorescence analyzer (PXRF). The PXRF was used throughout the field season to test the feasibility of estimating REY concentrations in the field. A 1-gal Ziploc bag was filled with ~1,000 g of sample material, and a brief description of the sample was made. Afterwards, the area was filled in and restored to original condition as much as possible. In general, coalbeds 2 ft thick or greater were sampled at the top of coal (TOC), middle of coal (MOC), and base of coal (BOC). In cases of beds being less than 2 ft thick, one representative sample was taken for the entire coalbed (EC). If bedding was exposed/accessible above or below the coal, then samples were collected there as well. It was also not uncommon to find interbedding within the seam, which was typically clay, ash, or sandstone. These were also collected whenever possible.

Coal was sampled at outcrops located in a variety of settings, making it difficult to consistently collect from top to bottom of a seam. Samples collected in the Hell Creek badland regions were ideal for describing outcrop and sampling entire coal seams, whereas outcrops in the Powder River Basin were often almost completely covered in grass. In many Powder River locations, only the top or top half of the coal seam was visible, so a sample might not represent the REY potential of the entire coal. In these cases, two or more locations were used to represent the seam. In other instances, the seam was visible, but layers above or below the coal were inaccessible.

Sampling at the Absaloka, Rosebud, and Spring Creek mines in the Powder River Basin was restricted to the active areas of their current operations. Samples from the top, middle, and bottom portions of the coal seams were collected. Overburden layers were the most difficult and dangerous samples to collect. These were bypassed due to safety concerns. In the Bull Mountain Basin, Signal Peak uses a longwall mining machine to mine coal underground. Therefore, sampling overburden, underburden, and specific portions of the coal seam was not possible due to safety concerns. Coal samples representative of the entire seam were collected instead.

All samples were prepared and analyzed by ALS in Reno, Nevada on a dry rock/whole-coal basis. Analyses were performed using inductively coupled plasma mass spectrometry (ICP-MS). Samples were tested for the following elements (REY and Sc in bold): Ag, Al, As, Au, B, Ba, Be, Bi, Ca, Cd, **Ce**, Co, Cr, Cs, Cu, Fe, Ga, Ge, Hf, Hg, In, K, **La**, Li, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Rb, Re, S, Sb, **Sc**, Se, Sn, Sr, Ta, Te, Th, Ti, Tl, U, V, W, **Y**, Zn, Zr, **Dy**, **Er**, **Eu**, **Gd**, **Ho**, **Lu**, **Nd**, **Pr**, **Sm**, **Tb**, **Tm**, **Yb**.

ASSAY RESULTS

The average total REY (TREY) content of coal worldwide is 68.5 ppm (Ketris and Yudovich, 2009). Coals in the U.S. have a slightly lower average TREY content of 62.1 ppm (Finkelman, 1993). The Department of Energy (DOE) uses the standard of 300 ppm (whole-coal basis) as a minimum economic threshold for the extraction of REY from coal and coal byproducts (DOE, 2017). Assay results from this study are summarized in table 1. All sample results have been released as an analytical dataset (Davison, 2024).

Table 1. Summary of assay results.

	No. of Samples	TREY (ppm) Min	Max	Mean	Standard Deviation
Tongue River Member	143	5	501	86	76
Lebo Member	39	15	214	60	41
Tullock Member	27	28	133	74	29
Hell Creek Formation	53	23	251	84	42

Fort Union Formation

Tongue River Member

Because of its significance for coal, a majority of the samples are from the Tongue River Member. A total of 143 samples were collected from 46 different sites in the Tongue River. The average TREY concentration is 86 ppm, the minimum is 5 ppm, and the maximum is 501 ppm. Coalbeds ranged from 10 in to 13+ ft in thickness. As previously mentioned, many beds in outcrop were partially exposed, making it difficult to determine a total thickness.

Sample 052C_23 (fig. 3) was taken from the base of an unnamed 3.5 ft coal seam in the Williston Basin along the Cedar Creek Anticline, south of the town of Glendive. The coal seam has no name, but appears in older coal publications and is mapped for over 35 mi. The outcrop runs parallel to the Peuse–Dominy coal outcrop, suggesting that it is likely part of the Tongue River Member. Samples were taken from the top and base of the coal and from a clay layer above the top of coal. The top of the coal is brownish and poor in quality, with a low TREY content of 20 ppm. The clay directly above TOC has a TREY content of 36 ppm. The BOC significantly increases in quality to a TREY concentration of 501 ppm, over 10 times the amount at the top. This area warrants more research to determine the extent of the seam and the mechanism of enrichment.

Sample 087A_23 (fig. 4) is an EC sample from a thin portion of the Pawnee Coal in the Tongue River Member located in the Powder River Basin. This subbituminous coal is black, fine, and powdery, with a TREY concentration of 320 ppm. The outcrop is difficult to sample due to overlying soil and vegetation, but is worth investigating further.

Samples 088D_23 (BOC), 088C_23 (TOC), and 088B_23 (clay above TOC) have increasing TREY concentrations of 144, 340, and 394 ppm, respectively (fig. 5). This 4 ft subbituminous Canyon Coal is from the Tongue River Member of the Fort Union in the Powder River Basin. The assay results suggest that the clay above TOC (088B_23) is a source of REY for the coal. This area of the Powder River Basin is also heavily vegetated, which will make further investigation difficult.

Figure 3. Samples 052A_23, 052B_23, and 052C_23.

Figure 4. Sample 087A_23.

Figure 5. Samples 088B_23, 088C_23, and 088D_23.

Figure 6. Spring Creek Mine. Anderson and Dietz coalbeds separated by thin ashes seen in the middle.

ICP-MS results indicate that none of the active mines have TREY concentrations near the DOE economic threshold of 300 ppm. The coal samples collected at the mines averaged 71 ppm TREY, with a minimum of 17 ppm, and a maximum of 162 ppm. The Spring Creek Mine targets the ~80-ft-thick Anderson and Dietz coal seams, which are joined together and separated by thin ashes (fig. 6). The Absaloka mine targets the Rosebud and McKay coal seams (also joined) that together average 32 ft in thickness. The Rosebud mine targets the Rosebud coal seam that averages 23 ft thick, and the separate, lower McKay at an average of 8 ft thick. The Signal Peak Mine (Bull Mountain Basin) targets the Mammoth coal seam, which ranges from 8 to 15 ft thick. All of these mines target thick, high-quality Tongue River Member coal for energy purposes, but none of the coals contain high concentrations of REY.

Lebo Member

A total of 39 samples at 16 locations were collected in the Lebo Member of the Fort Union Formation. Coalbeds ranged from 1 to 11 ft in thickness. The average TREY is 60 ppm, the minimum 15 ppm, and the maximum 214 ppm. Samples 064A_23 and 064B_23 are taken from a thin lignite coal named the D-Coal in the northeast corner of Montana, near Plentywood (fig. 7). Sample 064A_23 (TOC) came in low with a TREY concentration of 58 ppm, while 064B_23 (BOC) was a much higher 214 ppm. Unfortunately, this coal is in an area of Montana that is very difficult to sample due to overlaying debris and grass. Attempts will be made in subsequent years to locate more outcrops. Despite some thick coal seams (fig. 8), the average TREY content of the Lebo Member coals is the lowest of both the Fort Union and Hell Creek Formations, and lower than the U.S. average of 62.1 ppm. On the other hand, the sample size for this member is somewhat small and there is potential for higher concentrations in additional sampling.

Tullock Member

A total of 27 coal samples were taken at 10 locations from coals in the Tullock. The average TREY is 74 ppm, the minimum is 28 ppm, and the maximum is 133 ppm. Coalbeds ranged from 1 to 10 ft in thickness. The Tullock coal at these locations was primarily low-quality lignite with significant amounts of shale. The thickest coal sampled was 6 ft, but the quality was

Figure 7. D-Coal samples 064A_23 and 064B_23 from the Lebo member.

Figure 8. Thick T-Coal of the Lebo Member (samples 018A_22, 018B_22, 018C_22).

Figure 9. Sample 071B_23, Contact Coal of the Tullock Member.

very poor. Sample 071B_23 had the highest TREY concentration of the Tullock at 133 ppm (fig. 9). This sample is from a local coal seam and is very thin with a high shale content. The TREY concentration numbers for the Tullock are not as low as for the Lebo and fairly average for the Fort Union Formation as a whole.

Hell Creek Formation

Coalbeds sampled in the Hell Creek Formation ranged from 7 in to 6 ft in thickness. Coals tended to be low-quality with a significant amount of sediment, which explains why they are not typically named in literature, as they are not ideal for energy production. A total of 53 samples were collected from 21 sites identified as Hell Creek. None of the Hell Creek samples reached the DOE economic threshold of 300 ppm. Of the 53 samples taken, only two samples were over 200 ppm, and 14 samples over 100 ppm. The average TREY is 84 ppm, the minimum is 23 ppm, and the maximum is 251 ppm. Samples 053B_23 (clay above TOC) and 053C_23 (EC) were from a very thin coalbed (fig. 10). Sample 053C_23 had a TREY concentration of 225 ppm, and the clay above 053B_23 had a concentration of 251 ppm. Thin coals like these will likely never become economic sources of REY, but due to the highly exposed geologic layers in the badlands, they can potentially reveal clues about how they became enriched.

Concentration Numbers and Bed Position

Average coal TREY concentrations for all TOC samples is 65 ppm, MOC is 58 ppm, and BOC is 87 ppm. BOC samples have the highest coal TREY concentration numbers overall; however, the highest concentrations were in the associated non-coal layers (sandstone, shale, ash, etc.) Non-coal samples above the TOC seams averaged 99 ppm, below BOC averaged 110 ppm, and interburden averaged 99 ppm. Overall, non-coal sediments ranged from 18 to 394 ppm and averaged 101 ppm. The higher ppm averages for these non-coal samples vs. coal samples cannot be ignored. Again, it suggests that the source of REY for coals may come from bounding layers.

DISCUSSION

Of the 262 samples collected, 11 had TREY concentrations greater than 200 ppm, and 4 were greater than the DOE-suggested threshold of 300 ppm. The average TREY for all coal samples is 80 ppm. This

Figure 10. Thin Hell Creek coal, samples 053B_23 and 053C_23.

number is higher than the U.S. coal average of 62.1 ppm, but far below the suggested 300 ppm threshold. However, roughly half of all samples, coal and non-coal, were above the U.S. average and roughly 25% were above 100 ppm.

Considering the differing geologic environments the coals were collected from, the two different coal types and the overall differences in the coal seams themselves, the TREY averages suggest that there are some very low REY concentrations in coal seams and some very high ones. These preliminary results further suggest that most coalbeds have a low concentration of REY; however, there is also the possibility of a small percentage of beds with highly elevated levels.

Chondrite Normalization

The ICP-MS elemental results for all coals were chondrite normalized using values from Taylor and McLennan (1985). This was done to show the relationship between LREY and HREY, and how the REY concentrations compare to other coal deposits in the U.S and around the world. The results show a shallow slope from LREY on the left to HREY on the right, indicating that LREY are not severely disproportionate to HREY (fig. 11). This pattern, showing the normalized average REY concentrations, is typical of coals and an advantage over REY ores that have a higher proportion of LREY to HREY. Depending on the market, HREY are generally worth more than LREY and can be the driving force in making a mine economic. All Montana coals tested have very similar average REY distributions and compare nicely to the U.S. and world coal averages (fig. 12). World averages are slightly elevated compared to the U.S. and Montana averages, but overall the proportions of LREY to HREY are similar.

The Occurrence of Scandium (Sc)

Scandium is briefly mentioned due to its high value and association with REY. Scandium occurred in all year 1 coal samples. The average Sc content of sampled Montana coals is 3.75 ppm and the maximum is 17.2 ppm. The average is slightly lower than the worldwide average of 3.9 ppm (Ketris and Yudovich, 2009) and the U.S. average of 4.2 ppm (Finkelman, 1993). No trend was found between Sc and the occurrence of other REY. Likewise, there is no trend between Sc and LREY, HREY, or TREY (LREY + HREY). While Sc occurs with REY in coal deposits

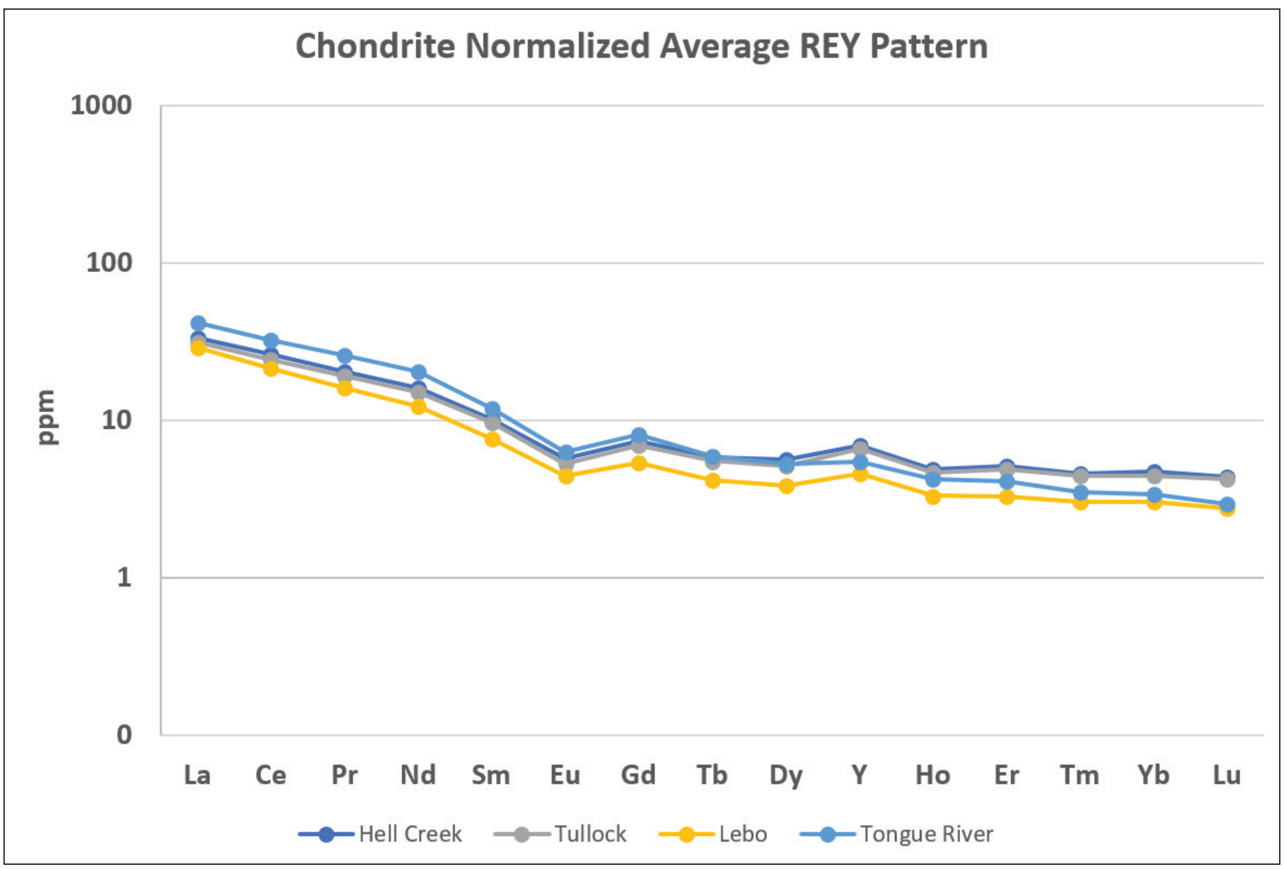

Figure 11. Chondrite normalized average REY values from sampled Montana coals. Chondrite values are from Taylor and McLennan (1985).

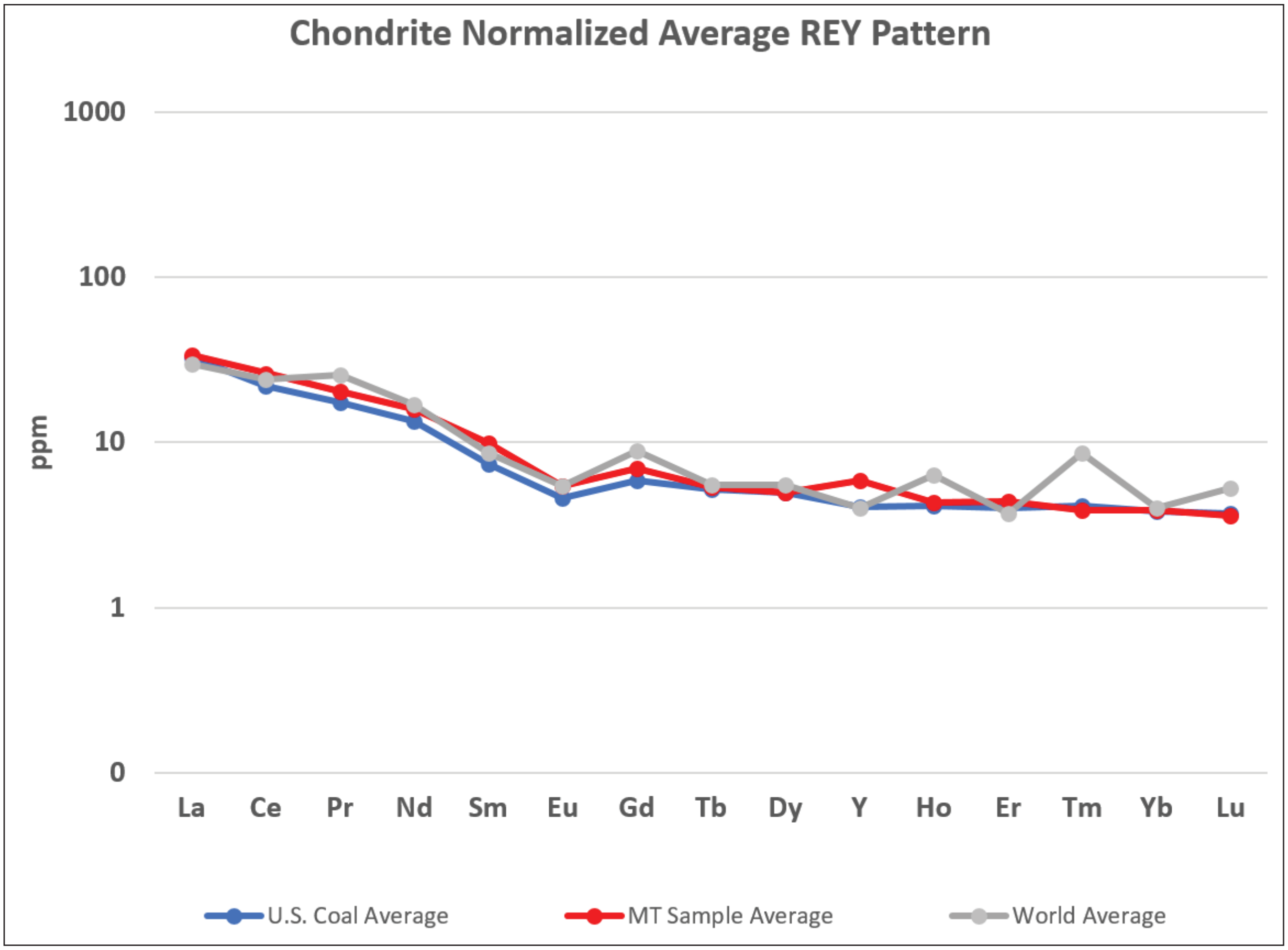

Figure 12. Chondrite normalized average REY values from U.S. coals, world coals, and total sampled Montana coals. Chondrite values are from Taylor and McLennan (1985); U.S. REY in coal averages from Finkelman (1993); world coal averages from Ketris and Yudovich (2009).

around the world, the concentrations are not predictable and generally do not occur in large enough quantities to be economic (Van Gosen and others, 2019). This also appears to be the case in Montana coals. Conversely, Y exhibits a very different behavior than Sc and is discussed in the next section.

X-Ray Fluorescence

A Niton XL-5 Plus portable x-ray fluorescence analyzer was used in the field to estimate the REY content of sampled coals. Since the concentrations of most REY in coal are typically too low to obtain a reading with a PXRF, the indicator element Y is used to estimate total rare earths. In the following example, I use the simple method of estimating total REE from Uhrin (2018). In the Uhrin paper Sc and Y are included, so I have included them as an example. It is not clear what Uhrin considered heavy rare-earth elements (for example, sometimes Gd isn't included), so I will be using Gd, Tb, Dy, Y, Ho, Er, Tm, Yb, and Lu.

Indicator elements are typically high in abundance, associated with rare earths, and detectable with a PXRF (Uhrin, 2018). The lab ICP-MS results for Montana coals show a strong correlation between Y and total HREE values (fig. 13). Additionally, there is a good correlation between ICP-MS total HREE and total REE (fig. 14). In theory, if we can create a relationship between Y-XRF and Y-ICP, we can estimate total rare earths from Y-XRF in the field.

In this study, total HREE is an average of 57% of the Y-value (fig. 13), the average percentage of HREE in total REE is 9% (fig. 14), and the Y-ICP value is 65% of the Y-XRF value (fig. 15). Using these numbers, a Y-XRF reading of 73 ppm is needed to indicate a coal with 300 ppm total rare earths. This is a much higher value than the Y value of 25–30 ppm Uhrin concluded. This is to be expected, as coals are highly variable in terms of quality and content even across small areas. The coal sample set used in the Uhrin paper is likely very different than the one used in this paper. On top of that, many factors in the field can affect

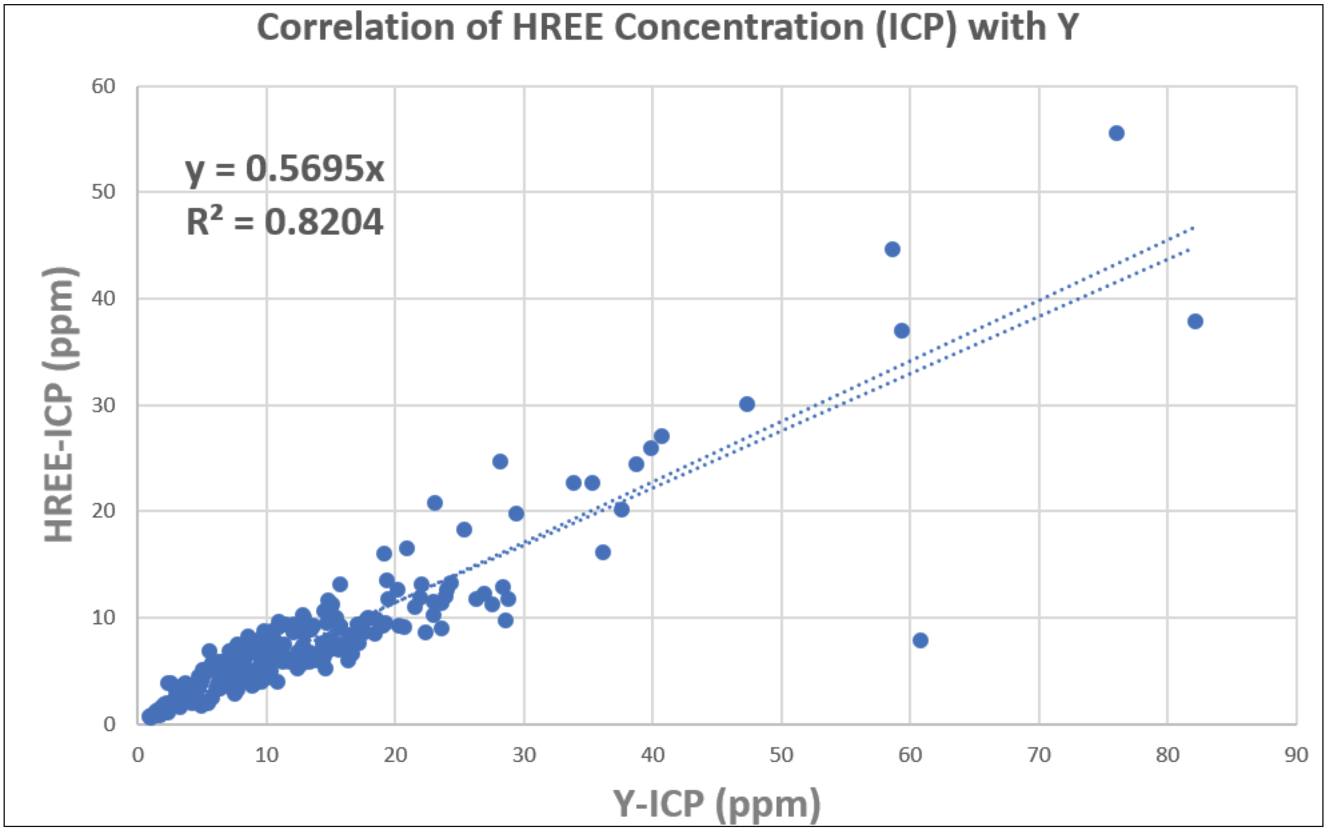

Figure 13. HREE correlation with Y (ICP).

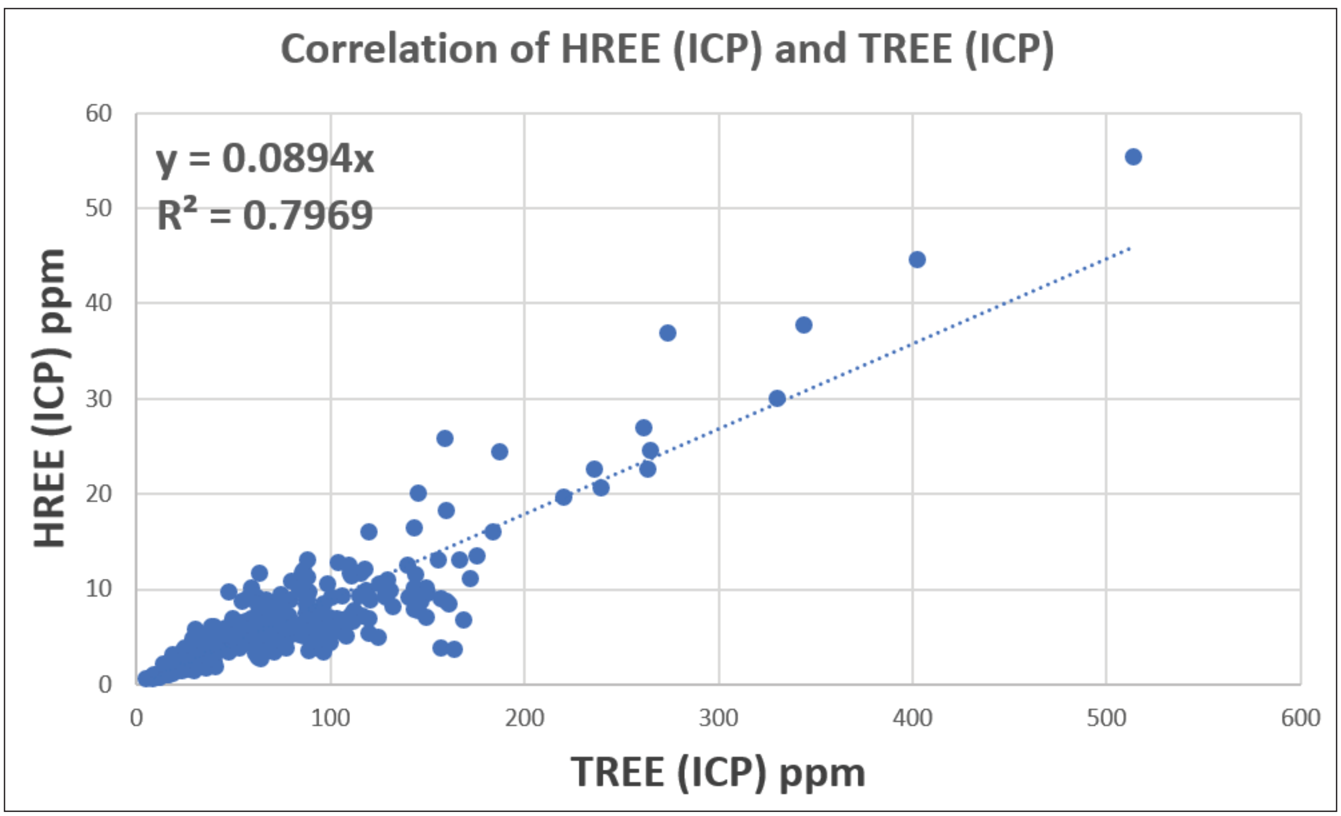

Figure 14. HREE (ICP) correlation with TREY (ICP).

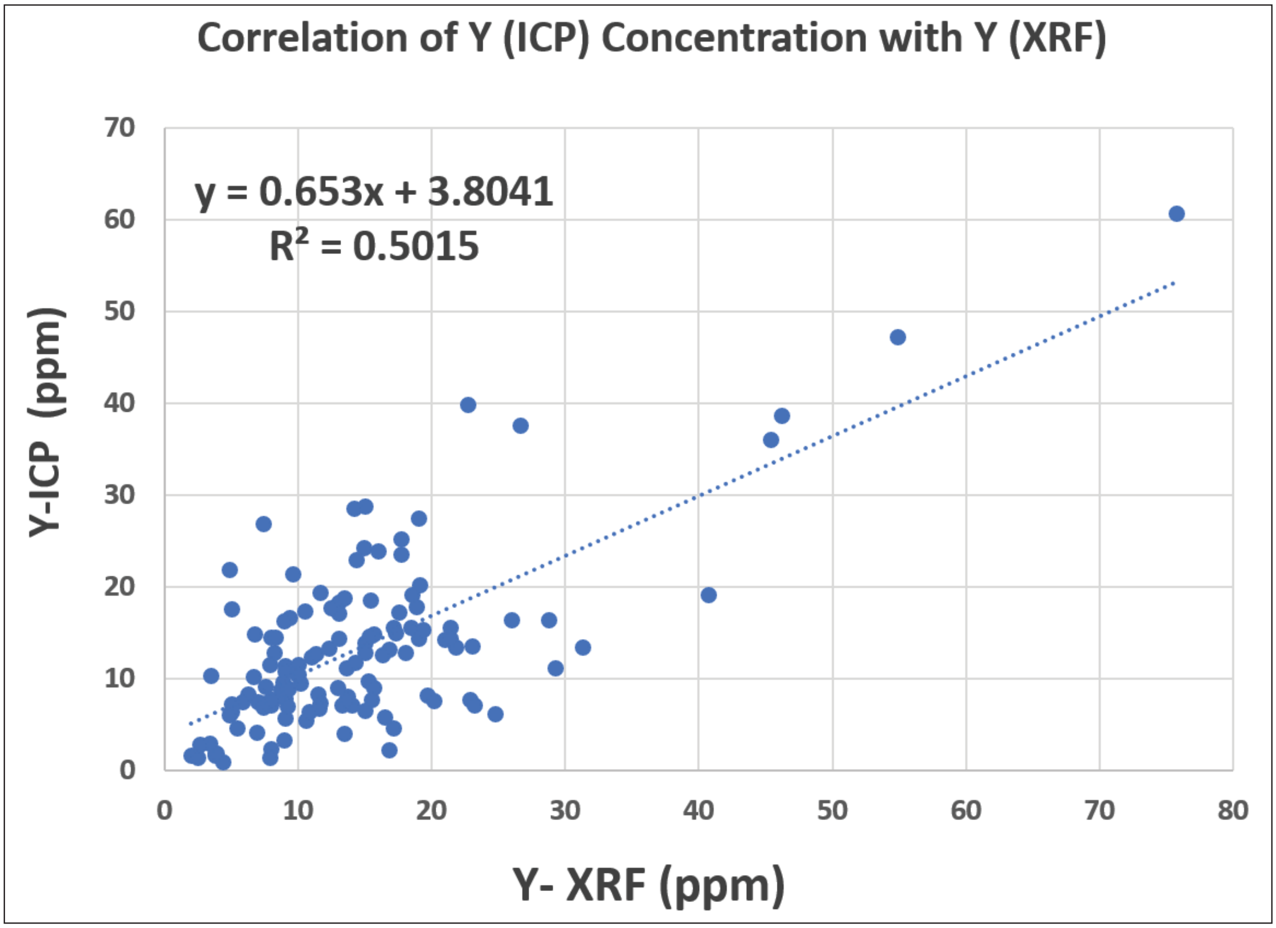

Figure 15. Y-ICP correlation with Y-XRF.

XRF analysis, including moisture, proximity of the device to the sample area, and sample consistency.

In general, a Y PXRF reading of 50 ppm or higher in the field usually indicated a REE-enriched coal. However, it was never accurate enough to estimate exact total rare earth ppm values. In the future, different sampling methods will be used in an attempt to gain more accurate PXRF readings and increase the correlation between Y-ICP and Y-XRF.

Weathering

Towards the end of year 1, five locations were chosen in the Powder River Basin to test the effects of weathering on REY concentrations in coal outcrops. Samples were taken at two different depths into the outcrop. For all previous samples, a minimum 12 in (30 cm) excavation was made into the outcrop, and in some cases a deeper depth depending on the quality of the coal. For the five locations, a 14.5 x 2 in (36.8 x 5 cm) core barrel was used to drill into the outcrop further to a total depth of ~26.5 in (67.3 cm). These samples were then compared to the originals taken at 12 in (table 2). The dataset is small, but indicates that weathering is potentially a significant factor in leaching REY from coal outcrops. Four out of the five cored samples show an increase in TREY, and three out of five show a considerable significant increase. Samples 088A_23 and 088C_23 are the most profound, with a percentage difference of 118.5%. The coals in table 2 were all sampled at roadcuts, revealing something about the relatively short timeframe required to leach REY from the outcrop after road construction. Additional outcrop testing will shed more light on potential key differences in REY leaching between the coal maturities and roadcut vs. natural outcrop.

CONCLUSIONS

No coal seams with potentially minable resources of REY were discovered in the first year of this study. However, the presence of coalbeds with TREY concentrations greater than 300 ppm suggests that Montana coal has the potential to hold economic amounts of REY. Clues about how Montana coals become enriched with REY can lead to predictive modeling and thus economic seams. There appear to be some early indications about possible enrichment methods

Table 2. Cored outcrop REY concentration results.

Sample	Sample Location	HREY ppm	LREY ppm	TREY ppm	% Difference
075A_23	TOC	3.13	7.74	10.87	
075D_23	CORE of TOC	2.85	3.98	6.83	
				-4.04	**-45.65%**
076C_23	TOC	25.35	67.43	92.78	
076F_23	CORE of TOC	37.36	95.67	133.03	
				40.25	**35.65%**
081C_23	BOC	63.15	117.42	180.57	
081D_23	CORE OF BOC	96.29	170.54	266.83	
				86.26	**38.56%**
088A_23	TOC	38.34	49.18	87.52	
088C_23	CORE of TOC	119.98	220.2	340.18	
				252.66	**118.15%**
091A_23	TOC	32.47	42.35	74.82	
091C_23	CORE of TOC	35.95	44.94	80.89	
				6.07	**7.80%**

based on the proximity of high-REY coals to high-REY bounding, non-coal layers. Further sampling, technique refinement, and a focus on promising areas will assist in shedding light on the nature of REY in Montana coal.

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