Assessment of Rare Earth Elemental Contents in Select United States Coal Basins

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Tetra Tech – January 2015
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TABLE OF CONTENTS

Executive Summary .................................................................................................. ES-1

Section 1 – Introduction ....................................................................................... 1-1
  1.1 – Background .................................................................................................. 1-3
  1.2 – Methodology ............................................................................................... 1-4

Section 2 – Demonstrated Reserve Base ............................................................... 2-1

Section 3 – REE Observations in CoalQual Coal Samples .................................. 3-1
  3.1 – Global Review – All States ......................................................................... 3-1
  3.2 – Regional Review – Select Eastern and Western States .................................. 3-7
  3.3 – REE Spatial Resource Estimations ............................................................... 3-12

Section 4 – Summary and Recommendations .................................................... 4-1
  4.1 – Summary .................................................................................................... 4-1
  4.2 – Recommendations ...................................................................................... 4-2

Bibliography ........................................................................................................... B-1

LIST OF TABLES

  Table 1.1 General Composition of Kaolinite
  Table 1.2 Relationship Between REE and Aluminum for Select Samples
  Table 2.1 Estimated Recoverable and Demonstrated Coal Reserves – 2012 Data
  Table 2.2 Total REE Grade Cut-Off Table – Western States
  Table 2.3 Total REE Grade Cut-Off Table – Eastern States

Tetra Tech – January 2015
LIST OF FIGURES

Figure 1.1  Sample Distribution of Analyzed Coals in USGS CoalQual Database
Figure 1.2  Possible Source Mechanisms for Introducing REE into Coal Basins
Figure 1.3  Generalized Proportions of Coal and Non-Coal Content of a Coal Seam
Figure 1.4  Relationship Between STDASH and Calculated Kaolinite
Figure 2.1  REE Grade Tonnage Curve - Western State Coals
Figure 2.2  REE Grade - Tonnage Curve for Eastern State Coals
Figure 3.1  Relationship Between Btu Content and STDASH for All CoalQual Coals
Figure 3.2  Filtered Btu versus STDASH for Select States
Figure 3.3  Relationship Between STDASH and Aluminum – All CoalQual Coal Samples
Figure 3.4  Relationship Between STDASH and Silica Content – All CoalQual Coal Samples
Figure 3.5  Relationship Between Total REE and % STDASH – All CoalQual Coal Samples
Figure 3.6  Relationship of Total REE and % STDASH Excluding >300 ppm REE
Figure 3.7  Relationship of Total REE and Aluminum Excluding >30 ppm REE
Figure 3.8  Relationship Between Btu Content and STDASH for Eastern State Coals
Figure 3.9  Relationship Between Btu Content and STDASH for Western State Coals
Figure 3.10 Relationship Between Total REE and Aluminum – Eastern State Coals
Figure 3.11 Relationship Between Total REE and Aluminum – Western State Coals
Figure 3.12 Relationship Between Heavy Metals in Eastern State Coals
Figure 3.13 Variogram for Total REE in Eastern State Coals
Figure 3.14 Kriged Total REE – Eastern State Coals
Figure 3.15 Calculated REE Grade in Equivalent Kaolinite – Eastern State Coals
Figure 3.16 Calculated Tonnage of Total REE – Eastern State Coals
Figure 3.17 Kriged Total REE – Western State Coals
Figure 3.18 Calculated REE Grade in Equivalent Kaolinite – Western State Coals
Figure 3.19 Calculated Tonnage of Total REE – Western State Coals
Figure 4.1  Relationship Between Aluminum and Potassium with Total REE – USGS Coal Samples
Figure 4.2  Oil and Gas Well Locations – Kentucky
Figure 4.3  Oil and Gas Wells with LAS Data – Kentucky
Figure 4.4  Geophysical Well Log of the ARCO Well #1
Figure 4.5  Distribution of the Amburgy Coal Seam in Eastern Kentucky
Figure 4.6  Distribution of the Fire Clay Coal Seam in Eastern Kentucky
Figure 4.7  Distribution of Kriged Total REE in Relation to the ARCO Well #1 in Eastern Kentucky

ATTACHMENT

Attachment 1 – Supporting Data Sets
EXECUTIVE SUMMARY

Rare earth elements (REEs) are increasingly important strategic materials utilized in many high-technology arenas including consumer electronics, defense, energy, and scientific endeavors. Approximately 85 percent of the world’s reserves are controlled by foreign concerns, mainly China. Limited commercial-grade REE deposits in the United States exist at Mountain Pass, California, and Bear Lodge, Wyoming. Numerous other sub economic occurrences of rare earth materials have been identified associated with igneous dikes and intrusive bodies (formed when molten material penetrates into existing rock strata and solidifies) located in the igneous/metamorphic complexes of the eastern Blue Ridge Mountains and associated with igneous dikes and intrusive bodies throughout the western States.

Studies conducted in recent years by the United States Geological Survey (USGS) and private concerns noted that REEs are present in coals throughout the basins of the United States. These REEs appear to be concentrated in the waste products of coals such as finely mixed clays, coal-partings, under-clay, or waste rock and fly or bottom ash (unburned waste remaining after combustion).

In this report, Tetra Tech developed methods for:

1. Predicting where high concentrations of REEs exist in coal basins using published geochemical, geophysical, and geological information.
2. Defining controls and occurrences of REE averages over total coal seams based on the chemistries and locations of samples.
3. Utilizing geostatistics to produce quantitative estimates of REE resources within coal basins.
4. Performing in-field detection and measurement of REEs in surface coal samples using a field portable X-ray fluorescence (XRF) spectrometer.

After implementing these methods and evaluating the results, the following key observations were identified in this study:

- The formation of coal generally occurs in a basin that may also contain REE-enriched sediments from the deposition and/or erosion of volcanic, intrusive, and detrital sources.
- The REEs associated with coal generally favor partitioning to clay materials. Only small amounts have been described in the literature as likely being fixed by ion exchange processes into the organic phase. The reported values therefore are attributed to the inorganic phase only.
- These aluminum-rich minerals are assumed to be the non-organic mineral portion of the coal such as clay, with kaolinite being the most common.
- Kaolinite has been shown to be able to entrap REEs by ionic bonding.
- REEs often are contained within resistive detrital sands such as monazite, rutile, zircon, and xenotime. Specific metals associated with these phases appear to correlate well with each other in the CoalQual samples and could represent a separate but smaller source of REEs in the samples.
REEs appear to be more prevalent in aluminum-rich portions of the coal deposits, and therefore using equivalent kaolinite as the proxy for REE accumulators allows definition of areas in the select coal basins where REE contents are 5 to 10 times greater than the REE content of the coals as a whole.

Geostatistical methods such as variography and kriging are effective in estimating REE resources spatially.

Estimates of inferred REE resources contained in the aluminum-rich fraction of the coal deposits using a cutoff of 500 ppm REE from the five western States are 6 million metric tonnes and 4.9 million metric tonnes for the four eastern States (Pennsylvania, West Virginia, Virginia, and Kentucky).

The field-portable XRF spectrometer is a valuable tool in locating high-concentration REE areas within coal and aides in selecting samples for more detailed and costly chemical analyses.

Based on the results of this study, it is recommended that future studies include the following:

- Expand the coal database to include areas with limited representation in the current data set, including lignite deposits in the southern Gulf States and conventional coal deposits in select areas of the western United States.
- Calculate the amount of REE tonnages for all of the United States, particularly the remaining top coal-producing States of Illinois, Ohio, and Texas.
- Refine the resource calculations by including data that consider the vertical variability of REEs in coals.
- Continue and expand reconnaissance coal sampling using the field-portable XRF spectrometer.
- Use the field-portable XRF unit to select surface coal samples to be analyzed by inductively coupled plasma-mass spectroscopy (ICP-MS) to define both the light and heavy REE contents of samples.
- After zones of elevated REE content are identified, determine likely metallurgic extraction scenarios and define minimal grades for viable economic development, which could alter what is currently considered an economic coal seam because other constituents could also be extracted during coal mining operations.

The results of this study support the contention that REE occurrences in coal and associated waste materials have the potential to be important and viable source of rare earths, especially if mining costs can be minimized by coupling the extraction process with conventional coal mining practices.
SECTION 1: INTRODUCTION

Rare earth elements (REEs) are of strategic importance because they are used for their unique magnetic, phosphorescent, and catalytic properties in numerous products and processes including high-strength magnets, car batteries, and catalysts and in defense and consumer electronics. There are 16 REEs, 15 in the chemical group lanthanides, atomic numbers 57 through 71, plus the chemically similar element yttrium (atomic number 39). The REEs are divided into light and heavy groupings based on atomic weight, with elements with atomic numbers 57 through 63 considered light REEs, and elements with atomic numbers 64 through 71 considered heavy REEs. Yttrium is classified as a heavy REE based on its physical and chemical similarities to other heavy REEs. Despite their name, REEs are moderately abundant in the earth’s crust, but they are rarely found in quantities that are economically exploitable. These elements are commonly concentrated in late-stage alkalic magmatic rocks (containing large amounts of alkalis, especially sodium and potassium) and their weathering products. Most of the world’s commercial reserves are under the control of China, making it important that the United States identify and develop sources domestically.

REEs can accumulate via chemical and physical processes in clays and near coal/clay interfaces, and some coals have been shown to contain elevated concentrations of REEs. If it can be demonstrated that economically viable sources of REE may be present in coal and/or its waste products, the reliance on foreign sources of REEs can be minimized, and in addition, an additional revenue stream may be available for current and future coal mining operations. Further, in most mining operations, the extraction and amassing of the commodity represents the bulk of the mining expense. If REEs are concentrated in coal waste rock and/or coal fly-ash, the additional expense of mining the REEs would likely be negligible because the coal is already being mined whether or not the REEs are used. Based on this scenario, the grade of REE required for profitable exploitation would most likely be lower than that of a traditional REE mine not associated with coal. For example, in the Bear Lodge Prospect, a carbonatite dike REE occurrence in northeastern Wyoming, grades approach 4.7 percent of total rare earth oxides (RER, 2014). Conceivably, REEs at sub-percent grades could be viable resources if mining and processing startup costs in coal-related REE occurrences could be minimized.

The current study involved the investigation of the association of coal and REEs by assessing the geophysical makeup of coal basins (i.e., using gravity/magnetics to assist in mapping basement features), determining what geologic relationships might exist between REE occurrences in the various coal basins, cataloging elevated REE occurrences in coal and non-coal entities within the basins, and identifying likely sources of REEs in select coal basins in the eastern and western United States. In addition, geostatistical methods were employed to provide a statistically valid interpretation of REE relationships with geology as a means of predicting which coal basins could be viable targets for REE exploration. Data gaps in the current knowledge base were identified, and surface coal samples from western States were collected and analyzed for REEs using a portable XRF spectrometer. Similar efforts have been conducted by other parties in the eastern States.

This report summarizes the results of the review and compilation of published information concerning the geology, geochemistry, and resource estimates of select coal basins in the United States, with emphasis on REEs. These data were evaluated with respect to criteria that could be useful in defining “sweet spots” of these metals in coals and associated waste rock and/or ash. To accomplish this, five western States (Arizona, Colorado, Montana, New Mexico, and Montana) and four eastern States (Pennsylvania, West Virginia, Virginia, and Kentucky) were chosen for study. The United States
 Geological Survey (USGS) Coal Quality (CoalQual) Database was used extensively in this evaluation. Figure 1.1 shows the locations of samples from the lower 48 States included in the CoalQual Database.

Figure 1.1 Sample Distribution of Analyzed Coals in USGS CoalQual Database

1.1 BACKGROUND

Figure 1.2 depicts the following mechanisms by which REEs could have been introduced into coal basins:

1. Volcanic input (ash and flows).
2. Igneous emplacement via diapirs and dikes.
3. Erosional detrital input of heavy, thorium-rich, REE minerals from weathered igneous and metamorphic sources.

REEs do not readily fit into early-formed minerals because their ionic radii are generally incompatible. Because of this incompatibility, they are progressively enriched into the “leftover” igneous magma. So as the magma continues to cool and minerals are formed, the remaining melt becomes progressively enriched in these elements until they are forced into very late-stage minerals and/or form their own assemblages. Hence, late-formed igneous rock commonly contains minerals with greater REE contents. As a result, REEs end up in very late-stage minerals or in volcanic glass at the end of the magmatic process. If these materials are introduced into the coal basin by volcanic tephra (fragmental material of various sizes produced by a volcanic eruption), considerable amounts of REEs can
accumulate in the coal. Further, when chemical or mechanical weathering of the ash occurs, further REE enrichment will occur. Introduction by erosional and detrital processes of heavy resistant minerals (black sands) also introduces REE into a coal basin. Although coal swamps are generally low-energy environments not prone to heavy detrital input, periodic geologic transgression/regression cycles (cyclical increases/decreases in sea level relative to land) can introduce detrital material into the basin between coal layers. Finally, diapiric emplacement (upward movement of igneous material through the rock underlying the basin) can introduce REE material into an area of the basin where associated hydrothermal fluids can mobilize the REEs until they find a suitable receptor material such as kaolinite. Coals evaluated in this report from both the eastern and western United States show episodic volcanic ash falls, diapiric activity, and metamorphic/igneous complexes either within or adjacent to the basins that contain late-stage REE-rich minerals.

![Figure 1.2 Possible Source Mechanisms for Introducing REEs into Coal Basins](image)

### 1.2 METHODOLOGY

As part of this study, analytical data provided in the USGS CoalQual Database were evaluated to ascertain relationships between coal characteristics and chemistry. These relationships, in turn, were mapped using Environmental Systems Research Institute’s (ESRI) ArcMap10™ software to determine spatial relationships among the data and to generate REE grade maps of the various coal basins. Based on these analyses and as discussed in further detail below, it was shown that the non-organic contents of the coal samples appear to correlate with REE contents.
Figure 1.3 shows the organic (coal) and inorganic (mineral) portions of a coal seam that are commonly interspersed within the coal or that separate multiple coal seams within a basin. Data from coal samples included in the USGS CoalQual Database, including thickness, aluminum content, and total REE content, were subjected to kriging (a geostatistical method of interpolation via gridding that allows prediction of unknown values using weighted averages of values at neighboring known locations) using geographic information system (GIS) software to produce maps showing their areal distributions. The grids used to conduct the kriging were based on standard 2-dimensional block sizes of 1 kilometer (km) by 1 km with varying thicknesses. By converting the reported aluminum content of the coal samples to the equivalent amount of kaolinite, a bulk density can be assigned to each of the kriged blocks. The bulk density of coal is generally 1.35 grams per cubic centimeter (g/cc) and can be as low as 0.833 g/cc. The bulk density of kaolinite can range from 2.16 to 2.68 g/cc.

As discussed below, estimates can be made of the potential grades of REE material associated with the inorganic portion a coal seam. By calculating the proportion of coal to clay, the resulting REE grade of the clay material can be estimated. This assumes that all but a negligible amount of REE is associated with the inorganic component of the coal. In Figure 1-3, 75 percent of the sample is coal, 10 percent is the inorganic material interspersed within the coal seam, and 15 percent is the mineral partings and underclay. Depending on whether the partings were excluded from the USGS sample, the organic to inorganic proportions could range from 75 percent organic and 25 percent inorganic to 89 percent organic to 11 percent inorganic. In this study, an average density of 1.25 g/cc was used for the organic proportion of the coal, and an average density of 2.42 g/cc was used for the equivalent kaolinite fraction derived from the aluminum content. Hence, if a standard 1-km by 1-km kriged block was 3 meters thick, excluding the parting/underclay material, the density of the block used to define REE grade would be as follows:

\[
\text{Block Density} = \text{clay proportion} \times \text{kaolinite density} + (1 - \text{clay proportion}) \times \text{coal density}
\]

Therefore:
\[
\text{Block Density} = 0.11 \times 2.42 \text{ g/cc} + (1 - 0.11) \times 1.25 \text{ g/cc},
\]

or

\[
\text{Block Density} = 1.38 \text{ g/cc}
\]

Review of the CoalQual Database indicates that the concentration of total REE in coal varies proportionally to the concentration of aluminum. As shown in Table 1.1, kaolinite contains up to 20.9 percent aluminum and therefore converting aluminum content in parts per million (ppm) to an equivalent kaolinite content involves dividing the aluminum content by 209,000 (which equals 20.9 percent converted to ppm).
Figure 1.3 Generalized Proportions of Coal and Non-Coal Content of a Coal Seam

Sigularia and Fern-Rich Coal Swamp
Estimates of the REE content of several coal samples by select States appears in Table 1.2. The USGS CoalQual Database measured REE concentrations over the complete coal seam. The total rare earth concentration (REE) is the sum of the concentrations of the individual light and heavy rare earth concentrations in ppm, as follows:

\[
REE = (\text{Ce} + \text{La} + \text{Nd} + \text{Pr} + \text{Sm}) + (\text{Dy} + \text{Er} + \text{Eu} + \text{Gd} + \text{Ho} + \text{Lu} + \text{Tb} + \text{Tm} + \text{Y} + \text{Yb})
\]

See Table 1.2 for chemical symbol key.

### Table 1.1 General Composition of Kaolinite

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Element</th>
<th>Atomic Weight</th>
<th>Atoms</th>
<th>Mass Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>Aluminum</td>
<td>26.98154</td>
<td>2</td>
<td>20.90%</td>
</tr>
<tr>
<td>Si</td>
<td>Silicon</td>
<td>28.0855</td>
<td>2</td>
<td>21.76%</td>
</tr>
<tr>
<td>O</td>
<td>Oxygen</td>
<td>15.9994</td>
<td>9</td>
<td>55.78%</td>
</tr>
<tr>
<td>H</td>
<td>Hydrogen</td>
<td>1.00794</td>
<td>4</td>
<td>1.56%</td>
</tr>
</tbody>
</table>

http://www.webqc.org/mmcalc.php
By using the published USGS coal reserves data in the various States included in this study coupled with the thickness/grade maps generated in ArcMap 10™ from the USGS CoalQual Database, an estimate of total REE resource potential can be calculated, as discussed below.

The rationale for normalizing samples to their equivalent kaolinite is based on the observed relationship in the USGS CoalQual Database between standard ash content (expressed as a percentage) (STDASH) and reported aluminum content. As shown on Figure 1.4, there is a fair correlation (R² = 0.6999) between reported STDASH values and aluminum-derived calculated kaolinite contents of coal samples in the CoalQual Database. Further, total REE values show similar relationships with STDASH and aluminum content.
SECTION 2: DEMONSTRATED RESERVE BASE

USGS has estimated that the United States demonstrated coal reserve base in 2012 was 481,385 million short tons (436,705 million metric tons or tonnes), which includes coal resources that have been identified to specified levels of accuracy and support economic mining with technologies available at the time of the estimation. These data include publically available information on coal reserves that have been mapped and verified to be technologically minable. The estimated recoverable reserves in the United States, a subset of the demonstrated reserves, totaled 257,648 million short tons (191,976 million tonnes). Estimated recoverable reserves include coal in the demonstrated reserve base considered recoverable after excluding coal estimated to be unavailable due to land use restrictions and after applying assumed mining recovery rates.

Table 2.1 shows the U.S. Energy Information Administration (EIA) demonstrated reserve base (DRB) and estimated recoverable reserves (EstRR) of coal in millions of short tons segregated by States. EIA is a principal agency of the United States Federal Statistical System responsible for collecting, analyzing, and disseminating energy information. In this report, estimated quantities of coal are based on the DRB of the study areas evaluated. The western study area includes Montana, Wyoming, Colorado, New Mexico, and Utah, with a DRB of 211,617 million short tons (191,976 million tonnes); and the eastern study area includes Kentucky, Virginia, West Virginia, and Pennsylvania, with a DRB of 88,079 million short tons (79,904 million tonnes). The combined western and eastern study areas include 56 percent of the total DRB of the United States.

The geologic resource quantity of coal was generated using the USGS CoalQual Database and ArcMap10™. In turn, the geologic resources were adjusted to reflect the selected area’s DRB. From these adjusted values, the total REE contents in the DRB areas were estimated. For this report, all tonnages of coal, equivalent-kaolinite, and REE have been converted to their metric form, signified as tonnes, as follows:

\[
\text{Metric Tonnes} = 0.907187 \times \text{Short Tons}
\]

These steps predict what the grade of REE values will be within kaolinite clay as a proxy for the inorganic portion (“ash”) starting with an average grade for the total coal seam. Figure 1.3 shows a common set of values found in the CoalQual Database. The example presented on this figure shows the average REE concentration of 100 ppm over a 40-meter coal seam will be enhanced to 1,000 ppm when the overall percentage of clay is 10 percent. This enhancement in REE concentration compares favorably with an average REE concentration of 54.9 ppm (range 0.2–1,031 ppm) in coal enhanced to 1,723 ppm (range 721 to 8,426 ppm) in assayed coals and ash around the world (2013, D. Mayfield). This check helps support the premise that REEs are mainly associated with the inorganic portion of coal, which remains within the “ash” upon processing and/or combustion.

Figure 2.1 shows a graph of the grade-tonnage curves for the western study area. The graph is read in the following manner:

- a) REE represents total REE in the graph.
- b) The graph’s X-axis charts increasing cutoff grades of REE, from 0 to 1,650 ppm.
c) The graph’s red line charts the tonnes (left Y-axis) of REE greater than any selected REE cut-off grade.

d) The graph’s blue line charts the average grade (right Y-axis) of REE greater than the same REE cut-off grade. For example, if an REE cut-off grade of 1,000 ppm is selected, the tonnage of REE shown by the blue line indicates that there are 1 million tonnes of REE.

e) At that same 1,000 cut-off grade, those tonnes have an average REE grade of 1,200 ppm. This grade-tonnage relationship is shown in tabular form in Table 2.2 for selected cut-off REE grades from 250 to 1,250 ppm by 250-ppm steps.

Figure 2.2 and Table 2.2 show the grade-tonnage relationships for the eastern States selected in this study. As shown, appreciably less total REE is available in the eastern States than in the western States, but this curve only considers the four eastern States whose areal extent is approximately one-third that of the western States utilized in the study. For example the combined area of the eastern States is 150,500 square miles. Wyoming alone is 97,818 square miles, and the area of the western States reviewed in this study is 460,766 square miles. The lower end of the eastern States curves are relatively flat as a result of the Kriging algorithm, so while no appreciable difference occurs below a 750-ppm cutoff, 500 ppm was chosen for consistency with the western data. In general, a more conservative approach would be to discount any value under 750 ppm.

### Table 2.1 Estimated Recoverable and Demonstrated Coal Reserves - 2012 Data

<table>
<thead>
<tr>
<th>Ranking</th>
<th>Coal Resource by State * (2012 Data)</th>
<th>EstRR (Million Short Tons)</th>
<th>DRB (Million Short Tons)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Montana</td>
<td>74,644</td>
<td>118,851</td>
</tr>
<tr>
<td>2</td>
<td>Illinois</td>
<td>37,835</td>
<td>103,995</td>
</tr>
<tr>
<td>3</td>
<td>Wyoming</td>
<td>37,413</td>
<td>59,951</td>
</tr>
<tr>
<td>4</td>
<td>West Virginia</td>
<td>17,013</td>
<td>31,281</td>
</tr>
<tr>
<td>5</td>
<td>Kentucky</td>
<td>14,213</td>
<td>28,713</td>
</tr>
<tr>
<td>6</td>
<td>Ohio</td>
<td>11,331</td>
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<tr>
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<td>Pennsylvania</td>
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<td>New Mexico</td>
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<td>Oregon</td>
<td>9</td>
<td>17</td>
</tr>
<tr>
<td>29</td>
<td>North Carolina</td>
<td>5</td>
<td>11</td>
</tr>
<tr>
<td>30</td>
<td>Georgia</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>31</td>
<td>Idaho</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td>All 31 States</td>
<td></td>
<td>257,648</td>
<td>481,385</td>
</tr>
<tr>
<td>Western Study Area</td>
<td></td>
<td>131,026</td>
<td>211,617</td>
</tr>
<tr>
<td>Eastern Study Area</td>
<td></td>
<td>28,335</td>
<td>82,351</td>
</tr>
</tbody>
</table>

* Source: Recoverable Coal Reserves at Producing Mines, Estimated Recoverable Reserves, and Demonstrated Reserve by Mining Method, 2012 (http://www.eia.gov/coal/reserves/)

Short Tons (tons) = 0.907187 Metric Tons (tonnes)

---

**Figure 2.1 REE Grade Tonnage Curve - Western State Coals**
Table 2.2 Total REE Grade Cutoff Table – Western States

<table>
<thead>
<tr>
<th>Western Total REE Grade-Tonnage</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>REE Cutoff (ppm)</strong></td>
<td><strong>REE Average Grade (ppm)</strong></td>
</tr>
<tr>
<td>250</td>
<td>700</td>
</tr>
<tr>
<td>500</td>
<td>800</td>
</tr>
<tr>
<td>750</td>
<td>980</td>
</tr>
<tr>
<td>1,000</td>
<td>1,200</td>
</tr>
<tr>
<td>1,250</td>
<td>1,400</td>
</tr>
</tbody>
</table>

Implied minimum grade cutoff

Figure 2.2 REE Grade - Tonnage Curve for Eastern State Coals

Table 2.3 Total REE Grade Cutoff Table – Eastern States

<table>
<thead>
<tr>
<th>Eastern Total REE Grade-Tonnage</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>REE Cutoff (ppm)</strong></td>
<td><strong>REE Average Grade (ppm)</strong></td>
</tr>
<tr>
<td>250</td>
<td>1,000</td>
</tr>
<tr>
<td>500</td>
<td>1,000</td>
</tr>
<tr>
<td>750</td>
<td>1,000</td>
</tr>
<tr>
<td>1,000</td>
<td>1,200</td>
</tr>
<tr>
<td>1,250</td>
<td>1,350</td>
</tr>
<tr>
<td>1,500</td>
<td>1,575</td>
</tr>
</tbody>
</table>

Implied minimum grade cutoff
In summary, the REE resource estimates were developed for a selected set of eastern and western States and must be understood in context. The analysis sought to establish fundamentally sound resource estimates of the total amounts of REEs that could be found within and around coal deposits. It assumes several things:

- The REEs associated with coal generally favor partitioning to clay materials. Only small amounts have been described in the literature as likely being fixed by ion exchange into the organic phase. The reported values therefore are attributed to the inorganic phase only.

- The analysis was a type of reconnaissance—using statistical methods to suggest bulk concentration values in the large volumes of coal and mineral matter that can be defined by the data points compiled in the USGS CoalQual Database. These data include a measure of the total thickness of the core that was taken to represent any individual sample point. In this data set, large partings (greater than 4 inches in thickness) were removed before the chemical analysis of the elemental composition (including the rare earths) was determined.

- The 500-ppm cut-off grade emerged from the analysis as a figure of merit to define a quantity (tons of REEs in situ) that might be locatable and recoverable should appropriate technology be available. Any practical attempt to exploit a coal seam to recover REEs—even one currently in production—would require that additional samples be taken to characterize the amount and distribution of REEs in the seam. This requires the creation of a mining plan customized to produce the coal, REEs, and any other valuable by-products present (cobalt and platinum group metals, for example).

- Not every “block” of material within the set of blocks used to define the study areas (actually volumes) would contain material of the cut-off concentration nor should one expect that material of this concentration would thread contiguously throughout a volume underlying a large portion of the study areas. Some portions of the deposits would exhibit average concentrations less than the cut-off concentration, and others would exhibit concentrations greater than that value. (These can be seen from the kriged values for REEs in coal deposits shown in some of the other figures provided by Tetra Tech).

- The cut-off value and the average value apply to the in situ coal (interspersed with bits of clay) and to clay partings and portions of the roof and floor close to the coal bed) not to the as-mined coal. Coal mining extracts coal with an eye toward the efficiency of coal extraction. Current mining practices are focused solely on extracting coal, taking as little extraneous material as possible. Some of the variance seen between in situ estimates and the concentration of REEs in this study arises from this difference.
SECTION 3: REE OBSERVATIONS IN COALQUAL COAL SAMPLES

This section describes the REE-coal distribution in the United States along with the estimated grades and tonnages of REE resources in the various coal basins based on available data and inferred relationships between REEs and chemical character of the coals. Review of the USGS CoalQual Database was performed to evaluate the relationship of REEs in coals with geologic processes, structural controls, and spatial disposition of coal basins in relation to igneous and metamorphic terrains.

In the initial review, coal parameters such as British thermal unit (Btu) content, which is a measurement of the energy quality of the coal, and STDASH were used to identify trends within the data set. This occurred on a global (complete USGS CoalQual Database) as well as on a regional (select eastern and western States) basis.

3.1 GLOBAL REVIEW – ALL STATES

As might be expected, as the ash content (STDASH) increases, there is a tendency for Btu values to decrease. This is because as the relative proportion of inorganics increases, there is less carbon to consume in the coal and hence less energy content. Figure 3.1 shows the relationship between BTUs and STDASH of all samples in the USGS CoalQual Database. There exists a distinct boundary within the data above which the Btu content is limited by the ash content. At any given STDASH, the Btu value can extend from a minimum of just below 4,000 Btu but cannot extend beyond the value defined by the maximum defined by the slope of the upper bounding line.

![Figure 3.1 Relationship Between Btu Content and STDASH for All CoalQual Coals](image-url)
If one excludes samples with Btu values greater than 11,000, Figure 3.2 reveals that a likely singular relationship exists for all studied States for Btu versus percent STDASH. Further, the assumption is that STDASH is mainly derived from aluminum, silica, and other major lithologic cations. To test this assumption, concentrations of both aluminum and silica were plotted against STDASH (Figures 3.3 and 3.4, respectively). As shown, as the ash content (STDASH) increases, the aluminum and silica contents of the coal samples also increase. Next the relationship between total REE and STDASH was determined. Because the use of coal in power plants results in varying amounts of ash, it was assumed that this class of elements might favor coals that had greater potential for ash and therefore would necessarily contain greater amounts of impurities such as clay, alunite, etc. If this were the case, one would expect a linear relationship between STDASH and total REE content. Further, if the REEs were tied mainly in aluminum-bearing minerals, one should see a similar trend between total REE and aluminum contents. Total REE plotted against STDASH appears on Figures 3.5, and a subset of total REE truncated at 300 ppm plotted against STDASH content appears on Figure 3.6. On Figure 3.5, there are some high REE samples that appear somewhat randomly above the main cloud of samples that show a maximum content of about 275 ppm total REE. Upon eliminating any sample greater than 300 ppm REE, two clear populations or subgroups emerge, suggesting that the REE-to-aluminum control may be due to two different mineralogical or geochemical controls. This subdivision will need to be further investigated in any future work because it is not within the scope of this report. To evaluate this issue, it is recommended that detailed microprobe analysis be conducted to identify the mineral contents and elemental analyses of the organic and inorganic
portions of the coal at least on select samples that contain appreciable REE content.

Going further, Figure 3.7 shows total REE plotted against aluminum. Excluding the “anomalous” samples with total REE contents greater than 300 ppm results in a plot very similar to Figure 3.6.

**Figure 3.3 Relationship Between STDASH and Aluminum – All CoalQual Coal Samples**
Figure 3.4 Relationship Between STDASH and Silica Content – All CoalQual Coal Samples

\[ y = 2458.7x - 3030.3 \]

\[ R^2 = 0.6961 \]

Figure 3.5 Relationship Between Total REE and % STDASH – All CoalQual Coal Samples
Figure 3.6 Relationship of Total REE and % STDASH Excluding >300 ppm REE

Figure 3.7 Relationship of Total REE and Aluminum Excluding >300 ppm REE
These relationships are the basis for the approach taken in this report that the aluminum content of the coal can be used to calculate kaolinite as a surrogate for where the REE accumulates and is in fact directly correlated to the ash content (STDASH) of the coal. Kaolinite was chosen because many REE deposits throughout the world are associated with this particular clay. This is not to negate the possibility that other minerals could also act as accumulators for REE. Detailed microprobe work would be required to better define the relationship. This should be tested further, but in essence, both the eastern coals utilizing samples collected from the four eastern States and the western coals utilizing samples from the five western States show similar trends with aluminum, with the exception that the western coals show the bipopulation trend shown above, whereas the eastern samples appear to represent a single REE population.

Looking again at total REE plotted against aluminum for both the eastern and western States utilized in this report, we see that the Pennsylvania and West Virginia samples show a single population, whereas the western States are clearly defined as two populations. Note that these differences are not understood at this time, and more investigation including collection of additional samples may be required because the original USGS database involved numerous samplers, analytical methods, and vintages of sampling events. Any one of these could effect the overall quality and validity of the final analytical result. Care in ongoing work should be placed on standardizing the protocol used to collect, analyse, and process data.

Figure 3.8 shows the relationship between BTU and STDASH for the eastern State coals, and Figure 3.9 shows this relationship for the western State coals reviewed in this study.

3.2 REGIONAL REVIEW – SELECT EASTERN AND WESTERN STATES

Basement structural maps were compiled utilizing published fault map data from the USGS and State geological surveys (www.wsgs.wvnet.edu). Where data were sparse (such as in the Rome Trough Region of West Virginia), gravity and aeromagnetic data were used to infer basement fault features. These data were used to generate ESRI shape files and used in ArcMap10™ to identify distances for total REE and thorium from the faults. Thorium was considered because numerous thorium-rich breccia pipes often show an association with REE and because thorium-rich minerals such as monazite are potential sources of REEs. The premise was that at least some of the REEs present in the coal basins might be derived from deep-sourced fluids. Any sample greater than 10 kilometers from a mapped fault was discarded. This investigation could not find any sound statistical relationship between basement fault distances and total REE or thorium occurrences.

The relationship of STDASH to Btu content for the two main areas shows that there is less variability in Btu content in the eastern State data.

Figure 3.8 depicts the relationships between Btu content and STDASH for samples from Kentucky, Virginia, West Virginia, and Pennsylvania. As shown in this figure, the relationship between the two components is fairly tight, with the main population showing a negative linear Pearson correlation coefficient of -0.6674.
The western States, on the other hand, while exhibiting a similar inverse relationship between Btu content and STDASH, exhibited a wider range of Btu content for any given percent standard ash content (STDASH). Again, however, these data are capped by the maximum line discussed for the global USGS CoalQual data set. Figure 3.9 shows this relationship.
In regard to total REE plotted against aluminum for both the eastern and the western States utilized in this report, we see that the samples from Pennsylvania, West Virginia, Kentucky, and Virginia show a single population, whereas the western States are clearly defined as two populations.

Figure 3.10 shows the relationship between total REE and aluminum in eastern State coals, and Figure 3.11 shows this relationship for western State coals.
Figure 3.10 Relationship Between Total REE and Aluminum – Eastern State Coals

Figure 3.11 Relationship Between Total REE and Aluminum – Western State Coals
Utilizing the apparent relationship of total REE content in coal to aluminum content, all aluminum was converted to equivalent kaolinite using the relationship that 100 percent kaolinite contains 209,000 ppm aluminum. While other aluminum-bearing minerals are most likely involved, until detailed microprobe work is conducted on the samples, normalizing REE content to kaolinite allows one to estimate grades of REE in the clays, determine volume proportions of kaolinite to coal in the gridded sample blocks, and calculate grade-tonnage estimates of REEs in the various coal basins.

In addition to looking at the relationship between aluminum and total REE, it is known that heavy resistive detrital sands often are associated with rare earth occurrences. In that these detrital sands can be considered as potentially economic assets (such as the black beach sands of the Carolinas and similar “black” sands in India), the relationship between titanium, zirconium, and thorium for the Pennsylvania and West Virginia coal samples was investigated. The premise here was that at least some of the REE occurrences in the east could be derived from detrital sands finding their way into the coal basins from the weathering of crystalline igneous and metamorphic rocks from adjacent highlands during the periods that the coal swamps existed.

Figure 3.12 show this relationship among common metals found in heavy sands. Clearly these elements show a common variability.
3.3 REE Spatial Resource Estimations

Determinations of the distribution of REEs in coals for the eastern and western subsections were derived by generating concentration grids within relevant sections of the coal basins through the use of ordinary kriging (a Gaussian gridding/interpolation process) in ArcMap10™ with appropriate polygon clipping of the resulting grid using Geospatial Modelling Environment (GME), Version 0.7.2.0 software.

Figure 3.13 shows the variograms and statistical estimated error for total REE in two of the eastern States coal samples (Pennsylvania and West Virginia). A more refined kriged map is presented as Figure 3.14 for Kentucky, West Virginia, Virginia, and Pennsylvania.

On the figure, yellow shades represent lower concentrations and red shades indicate higher concentrations of REEs. The northern sections of the map near the margins of the basin in Pennsylvania show the greatest REE contents. In Kentucky, the southern edge of the basin and a zone crosscutting the basin toward the northwest have the greatest REE contents. In West Virginia, the higher values appear to be on the basin margins. In that many of the basin margin samples are closer to metamorphic and igneous complexes that were present during the period the coal was forming, these elevated samples could at least partially be resultant of detrital input of heavy sands. Detailed microprobe analysis might be appropriate to see if the mineral content is substantially different in the coals of this region compared to coals in the rest of the basin. It should also be noted that a somewhat widespread volcanic ash fall has been reported throughout the basin during the Carboniferous Period. This layer has been defined as a tonstein deposit by Lyons et al., 1992. A tonstein layer generally is a hard, compact, sedimentary bed composed mainly of kaolinite, and its presence could explain the elevated REE content in the coals. Because it was widespread, the differences in REE contents in different portions of the basin could indicate that multiple sources were at play as well. Further, diapirs and other igneous activity, although rare and difficult to identify in such a thick sedimentary column, are known to exist in the east. These also could be secondary sources of REEs in the basin. Locations of these features appear as black stars on Figures 3.14 through 3.16.
Figure 3.13 Variogram for Total REE in Eastern State Coals
It is the opinion of the authors that REEs are mainly associated with the mineral content of the coals. Prior plots shown in this report indicate a more than casual relationship between the ash content (STDASH) of the coal and aluminum content. Further there is a fairly strong correlation between aluminum and silica content with reported REE content. Because of this apparent relationship, aluminum was normalized to kaolinite and the kriged blocks were adjusted for a bulk density of the proportion of coal to inferred kaolinite to calculate a REE grade for the clay content. Block sizes were set at 1,000 m by 1,000 m. If the REE content is assigned only to the non-organic portion of the block, the grade increases substantially. This enrichment is often 10 times greater than the grade of the combined coal-mineral matrix block. Figure 3.15 shows the result of this determination.

This figure in essence is a map showing the REE grade of the non-coal matrix and could be used to highlight areas that have strategic REE potential from those that do not.

If one were to adjust the REE content to the total tonnes of clay (non-coal) material in the basin, a predictive map of tonnes of REE per 1-km² block could be estimated. These data appear on Figure 3.16. Such a map could be used to delineate more likely exploration targets over raw total REE content in coals by emphasizing areas where REE content is concentrated in the clay and where clay is likely to occur.
Figure 3.15 Calculated REE Grade in Equivalent Kaolinite – Eastern State Coals

Figure 3.16 Calculated Tonnes of Total REE – Eastern State Coals
A similar approach was made for the western subset samples. In this area, coal basins exist over much of the area but are interspersed between numerous igneous and metamorphic terrains. Also, episodic volcanic ash falls occurred in the region. Just as a tonstein deposit is present in the Appalachian basin, similar kaolinitic volcanic ash deposits have been noted in the west. In Raton Basin, one such deposit was discussed by Bohor and Pillmore (1976). A surface coal sample collected by the authors west of Trinidad showed an elevated Y content in excess of 270 ppm, and numerous USGS samples in the region showed elevated total REE contents. In addition to tonstein deposits, numerous REE deposits and dikes are known to exist throughout the western State study areas. Further, igneous bedrock is exposed more readily than in the east. This presents an opportunity to analyze and define the REE contents of these features much more readily than what is possible in the eastern basins. In other words, defining REE sources in the west is not as enigmatic as it is in the east. Another important difference between western and eastern coal basins is the age of the coal deposits. In the west, much of the coal was formed during the Cretaceous Period or even earlier. In the eastern coal fields, most coals are from the Carboniferous Period (Pennsylvanian and Mississippian epochs). Preservation of the younger western coals should be somewhat better than the older eastern coals.

Figure 3.17 shows the kriged results of REE contents in the coal samples. Again green shades indicate lower REE contents, and red shades indicate greater REE content. On this map, black stars indicate some known REE occurrences.
As was the case when looking at eastern REE and fault associations, the western data showed no statistically relevant correlation between REEs in coals and fault distance. Higher REE contents were noted northeast of the Wind River Range in Wyoming, east of the central Front Range and east of the Spanish Peaks volcanic complex (and Wet Mountains) of Colorado, and in the New Mexico volcanics southeast of the Four Corners area suggesting that some other geologic control (possibly igneous activity) is affecting the REE contents in these areas.

Once the REE content is adjusted for calculated kaolinite content, the grade map, Figure 3.18, indicates that much wider portions of the sampled coal basins have rich REE potentials. Grades for most of the area range from less than 100 ppm REE to over 500 ppm, with several areas exceeding 1,500 ppm total REE.

Adjusting grade to total tonnes of clay in each 1-km² block highlights areas where both grade and tonnage might show the highest recovery potential. This is shown on Figure 3.19.
Figure 3.19 Calculated Tonnes of Total REE – Western State Coals
SECTION 4: SUMMARY AND RECOMMENDATIONS

4.1 SUMMARY

Resource calculations of REEs in known coal reserves in both the eastern and western States utilized in this study suggest that the grade of kaolinite equivalent aluminum content of coal can be used to map areas of higher REE resource probability. By adjusting the REE content of coal to proportional kaolinite volumes, it is estimated that at a cutoff at 500 ppm REE, $6.0 \times 10^6$ metric tonnes of REE are associated with select western State coal basins and that an additional $4.9 \times 10^6$ metric tonnes are available from the eastern coal basins of Kentucky, Virginia, West Virginia, and Pennsylvania. These numbers were arrived at by taking a relatively conservative approach. While other aluminum-bearing minerals are most likely present, kaolinite was chosen as the surrogate since numerous REE deposits in the world are associated with kaolinite, and a widespread ash-derived tonstein deposit felt to be the source of some of the REE in the eastern coal basin generally is enriched in kaolinite. Similar tonstein occurrences have also been noted in western coal basins.

Due to the limited distribution and range of samples utilized in this study, these numbers are believed to be at the lower limit of the possible REE resources that ultimately are present. When looking at the relationship between aluminum and REE content and adjusting the grade of REE to equivalent kaolinite, the order of magnitude of REEs in the clay is comparable to published REE contents in select coal-ash samples. It is our contention that if this mineral fraction can be separated from the coal by preprocessing, during combustion at coal-fired plants, or both, a reliable and viable economic source of REEs exists. Further, it may be possible that the cost benefit of exploiting these metals could favorably impact the current coal industry by providing a secondary income stream and allow for the mining of currently sub economic coals. This would result in a much expanded coal reserve for the United States.

Based on the observed relationship between Btu content, STDASH, and metal content in coal, it is apparent that a refined analysis of the actual coal matrix needs to be conducted. We recommend that future programs include performing microprobe analysis of matrix material and REEs to determine the following:

- Mineral makeup of the inorganic materials in coal
- Chemical makeup of inorganic and organic portions of the coal
- REE contents of these materials
- Which minerals present have an affinity to REEs

Further, it would be useful to expand future study to include non-coal-related materials associated with the seams, such as clay partings, under clay, and roof material, to determine their REE contents and mineralogies.

In addition to studying the actual mineral contents of the coals, expanding sample coverage through the use of a portable XRF unit to assist in selecting samples for more detailed and costly analysis is recommended. Currently, several tens of samples were collected from Colorado, Wyoming, New Mexico, and Montana by the authors, and our client has initiated a similar task in several of the eastern and central United States coal fields. We highly recommend that this phase of the program be
continued in the future with our XRF and the client’s being calibrated to facilitate sharing and interpretation of data.

It is our contention that when REE occurrences are identified and mapped, coal operations could capitalize on the relationship of coal impurities and REEs by allowing for the profitable mining of lower quality coal using the REE content to offset processing cost to scrub or separate clay materials from the coal. This could provide both a coal income stream and an REE metals income stream. If economic analysis shows that there is a net gain in revenue over cost, this could have a positive impact on the coal market and result not only in a reliable REE source but expand the national coal reserves by accommodating what are today sub economic coal resources.

It should be noted that this study involved investigating the relationships and spatial distribution of total REEs. Future work should involve identifying both the light REE and heavy REE contents of coal. Such an approach could highlight areas and/or coal seams of greater strategic and economic importance. The XRF could be used to screen REE occurrences and the samples could then be submitted for more detailed ICP-MS analysis to define heavy REE areas.

Finally, as the REE contents of these materials are better estimated, work with metallurgists should be initiated to define methods of separation and extraction of these metals from coal and coal-related waste material. In that the coal infrastructure already exists, capitalizing on the REE content is likely to be economically and technically feasible.

When estimating economic cutoffs of REE grade in normal rare earth mining operations, a large portion of the cost involves the mining and concentration processes. In the case of REEs associated with coal and coal waste products, much of this cost would be minimized because it would be a part of the existing coal mining operation. This would allow for lower grades of REE content to be considered in a joint coal-REE operation compared to those involving REE extraction alone. Similarly, it could be argued that by extracting REE from coal and coal waste materials, the quality of coal that might be economically exploited could also be reduced, with REEs providing an offset to the reduced income stream of mining that coal alone. It is recommended that additional coal sampling for REEs be conducted to include areas, particularly in the western States where sampling is less dense than that of the eastern States. Further, waste rock piles, partings, and under clay material and existing coal ash piles at historical and active coal-fired plants should be routinely evaluated for REE content.

4.2 RECOMMENDATIONS

To better define REE distributions, characteristics, and resource estimates the following tasks are recommended:

- Extend the sample database to include coal basins in the Midwest (Illinois, etc.), and also Montana and North Dakota, plus the coal/lignite basins in the southern/southern United States and coastal plains (Texas, etc.) to get a better estimate of REE spatial distributions and characteristics and to calculate the amount of REE tonnages for all of the United States.

- Identify regions in the United States where current REE coal information is absent or sparse and devise a sampling plan to include those areas.
Verify and refine the USGS data by selective resampling. These samples should be analyzed using both in-field (portable XRF) and laboratory (ICP-MS) methods.

Perform detailed microprobe analysis of the organic and inorganic portions of coal deposits to ascertain what relationships exist between REEs and the organic and non-organic (mineral) matrices of the coal.

Expand the current study to include relationships between light REE and heavy REE materials. This should be done from a geologic and spatial context to determine whether predictive relationships can be derived.

As additional data becomes available, continue generating REE-grade maps of areas of interest. Blind studies designed to evaluate the grade calculations should be conducted to assist in refining the model as the program develops.

Determine what metallurgical processes might be advantageous in extracting REE material from coal and coal waste material, and concurrently and economically increasing coal quality.

Based on the USGS database, there appears to be a definite relationship between aluminum, potassium, and total REE content, (Figure 4.1). Therefore, when geophysical logs are available, there should be a relationship between well log gamma responses in clay-rich and/or detrital-rich coal sections with total REE content from the influence of the radioactive Potassium-40 isotope on the gamma response. Other researchers such as Ekman (2012) have noted the relationship between thorium and REEs. Further, gamma ray response will be affected by the presence of thorium and uranium. In some instances REEs can be associated with thorium-rich detrital sands, and uranium is known to exist at Bell Branch in the Red River Gorge of Kentucky associated with small coal stringers (Richers, 1981). Therefore utilizing gamma-ray logs along with additional petrophysical information could identify areas within coal basins that might host REEs.
Figure 4.1 Relationship Between Aluminum and Potassium with Total REE - USGS Coal Samples

Figure 4.2 shows the distribution of oil and gas wells (small dark green dots) on record by the Kentucky Geological Survey for the Commonwealth of Kentucky. As shown, there are in excess of 147,585 wells in their database. Of these, only 1,645 wells are reported to have electronic log and surface data (LAS) data available (Figure 4.3). Geophysical data from ARCO Well#1, shown as a bright green dot in Perry County were used in the following discussion.
Figure 4.2 Oil and Gas Well Locations - Kentucky

Figure 4.3 Oil and Gas Wells with LAS Data - Kentucky
ARCO Well #1 in Perry County had LAS well logs for borings that penetrated the Pennsylvanian section and is located in the portion of the county with several coal beds. Logs included Density Porosity, Caliper, Gamma-Ray, Density, and Neutron Porosity. Figure 4.4 shows these traces for the entire well as well as a more detailed view for the Pennsylvanian and Upper Mississippian sections of the well.

As shown, there is a well-defined interval at a depth 1,420 feet indicative of coal with low density and slightly elevated neutron porosity. This interval also has a slight gamma response that could be related to the presence of some radioactive material such as clays and/or thorium or uranium. In that this is single channel data, it is not possible to identify the radioactive source.

Figure 4.4 Geophysical Well Log of the ARCO Well #1

The location of this well (Figures 4.5 and 4.6) does show that at least two coal seams (Amburgy and Fire Clay Coal, respectively) are penetrated by this well. Further, kriged total REE coal data (Figure 4.7) indicate that this region of Perry County was rated as having elevated REE content in coal material. This relationship implies that investigating the relationship between petrophysical data, coal occurrence, and REE content might be a useful tool in defining areas with REE potential. Not all wells in the database penetrate the Pennsylvanian sections and may not have useful well log information in this section, but those that do could be utilized to define target zones for REE evaluation. Similarly, if paper logs are available for the coal-bearing zones, they also could be utilized to identify areas with elevated gamma response to assist in targeting REE investigations.
Figure 4.5 Distribution of the Amburgy Coal Seam in Eastern Kentucky

* Source: Kentucky Geological Survey
Figure 4.6 Distribution of the Fire Clay Coal Seam in Eastern Kentucky

Figure 4.7 Distribution of Kriged Total REE in Relation to the ARCO Well #1 in Eastern Kentucky
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Assessment of Rare Earth Elemental Contents in Select United States Coal Basins

Document No: 114-910178X-100-REP-R001-00


Simmons, W.M B and Heinrich, E. W.M, Colorado Geological Survey Department of Natural Resources, Denver, Colorado / 1980; Rare-Earth Pegmatites of the LIBRARY South Platte District, Colorado


University of Colorado Boulder, Undergraduate Honors Theses Honors Program, Spring 1-5-2014, Magmatic Evolution and Petrochemistry of Xenoliths contained within an Andesitic Dike of, Western Spanish Peak, Colorado, Ion Contreras.


US Energy Information Administration, (EIA), (http://www.eia.gov/coal/reserves/) Recoverable Coal Reserves at Producing Mines, Estimated Recoverable Reserves, and Demonstrated Reserve by Mining Method (Table 15), 2012.


Data sets utilized in generating this report can be found on the accompanying DVD. These data include information from the USGS CoalQual Database and the ESRI shape files used in preparing the figures.