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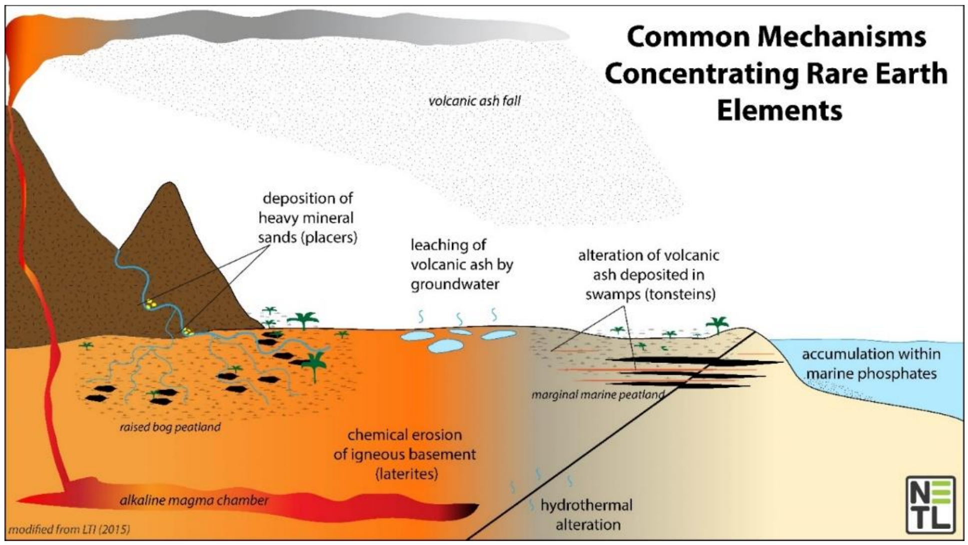

Towards A Geo-Data Science Method for Assessing Rare Earth Element and Critical Mineral Occurrences in Coal and Other Sedimentary Systems

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Cover Illustration: Schematic drawing of the common mechanisms that lead to enrichment of critical minerals and rare earth elements (REE) in sedimentary geologic systems.

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Towards A Geo-Data Science Method for Assessing Rare Earth Element and Critical Mineral Occurrences in Coal and Other Sedimentary Systems

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Acronyms, Abbreviations, and Symbols

Term	Description
Ce	Cerium
CM	Critical minerals
DA	Data Availability
DOE	U.S. Department of Energy's
EDX	Energy Data eXchange
Er	Erbium
HMS	Heavy-mineral sands
Ho	Holmium
HREE	Heavy rare earth elements (Ho, Er, Tm, Yb, and Lu)
La	Lanthanum
LREE	Light rare earth elements (La, Ce, Pr, Nd, and Sm)
Lu	Lutetium
Nd	Neodymium
NETL	National Energy Technology Laboratory
PE	Potential enrichment
Pr	Praseodymium
REE	Rare earth elements
REE-SED	Rare earth element sedimentary assessment method
Sc	Scandium
Sm	Samarium
STA	Subsurface Trend Analysis
Tm	Terbium
URC	Unconventional rare earth and critical mineral
USGS	U.S. Geological Survey
WCED	Weathered crust elution deposits
Y	Yttrium
Yb	Ytterbium

Acknowledgments

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EXECUTIVE SUMMARY

The United States is heavily reliant on imports of certain mineral commodities that are vital to the Nation's security and economic prosperity. This dependency creates a strategic vulnerability for industry and the military to supply chain disruptions, but also limits U.S. competitiveness in a number of growing markets, particularly clean energy technologies. The importance of these key minerals has been underscored by several Executive Orders (The White House Briefing Room, 2021; Executive Order 13817, 2017; DOE, 2017), including the President's recent declaration of a national emergency to deal with the supply chain threat to critical minerals (CM) and rare earth elements (REE).

Conventional sources of REE and CM resources in the U.S. are relatively limited, with some, such as gallium and graphite, being 100% imported from foreign sources (DOE, 2017). However, the U.S. has significant deposits of sedimentary rocks, including coal and carbon ore deposits, that have been documented in case studies to host REEs (NETL, 2020; Bauer et al., 2020). In addition, published studies indicate that CM resources occur in mine waste products, such as acid mine drainage, sludge, tailings and other materials currently viewed as byproducts from coal, iron, and other mineral mines (Sutterlin, 2019; Qin et al., 2015; Wang et al., 2015; Petlovanyi and Medianyk, 2018).

At present, there is no systematic method or approach to predict and identify REE/CM resource potential and occurrence from sedimentary systems, carbon-ores, or mine waste streams. This lack of a systematic assessment method inhibits our ability to predict where these occurrences are likely to be found and quantify the in-place or economically accessible volumes of unconventional REE/CMs in domestic sedimentary, carbon-ore, and mine byproducts. Thus, to quantify and establish a domestic supply of unconventional REE/CMs requires a systematic, data-driven, science-based assessment method and approach. This type of resource assessment method underpins oil, gas, coal, copper, uranium, gold and all other mineral exploration efforts. Development of these assessment methods usually take decades to a hundred years (e.g., oil/gas, gold) to produce. In addition, assessing unconventional REE/CMs offers an opportunity to beneficiate mine waste and byproducts which currently afford no value and may in some instances be the cause of environmental impacts. This data and knowledge are key to both assessing and predicting the occurrence and volumes of REE/CMs, but also to support future separations and extraction technologies to ensure their commercial viability as a domestic resource. Systematic resource assessment methods are proven to work. For example, from the 1800s to the 1970s oil and gas drilling success rate was 50% or less. After development of the petroleum system method in the 1980s, exploration success rate increased to above 90% at present (Wang et al., 2015).

In 2018, the U.S. Department of Energy's (DOE) National Energy Technology Laboratory (NETL) initiated development of an assessment model designed to systematically predict and assess domestic deposits of REEs and CMs from carbon ore and other sedimentary systems (i.e., NETL's REE sedimentary assessment method, REE-SED; NETL, 2020). This systematic approach leverages existing resource assessment methodologies matured through decades of R&D. This report presents the latest version of the NETL **U**nconventional **R**are earth and **C**ritical minerals (NETL-URC) model. The model provides a data-driven path to identify and commercialize domestic deposits containing critical minerals in coal basins. Moreover, the

NETL-URC model surpasses the capability of traditional probabilistic mineral resource assessment models used for reserve calculations by assessing the prospectivity of a deposit based on a comprehensive set of spatial geologic attributes. These additional considerations allow for a more robust assessment and calculation of resource reserves that are not solely based on the occurrence of the reserve, but on the accessibility and feasibility of recovering the resource in a sustainable and economical manner.

This effort aims to accelerate the development of technologies that lead to the commercialization of domestic REE deposits by:

- Guiding determinations of in-place resource estimates.
- Assessing whether REE are likely to occur in sufficient concentrations and recoverable volumes to support commercial extraction from domestic sedimentary basins.
- Revealing key knowledge and data gaps that hinder resource predictions and addresses select gaps in priority areas.

Results from development and initial testing have helped identify key trends and highlight potential opportunities to maximize the utility of coal resources, as well as identify current waste material with potential for REE monetization.

1. INTRODUCTION

Rare earth elements (REE) are a group of 17 chemically similar elements (atomic numbers 57-71, and Sc and Y) that are required to manufacture advanced materials and develop new technologies supporting the United States' infrastructure, defense, and energy needs. REE are typically found in bedrock- and regolith-hosted geologic ore deposits. Domestic coal deposits have also been identified as promising sources of REE (Seredin and Dai, 2012; Hower et al., 2016a; Hower et al., 2020) and other critical minerals (CM) (DOE, 2017). However, there remains a limited understanding of the spatial and volumetric occurrence of these deposits and resources. Specifically, the concentration and type of REE (e.g., excessive vs. critical) that are present and recoverable from domestic coal and other sedimentary deposits is largely unknown. Both concentration and type of REE present determine the economic viability of the deposit and the potential to provide a stable and secure domestic source of critical elements and minerals. Coal and coal-bearing sedimentary rock contains REE and many other trace and critical metal elements that are associated with organic matter (Lin et al., 2017) and inorganic minerals (Seredin and Dai, 2012). Occurrences of REE in domestic coal beds, coal utilization byproducts, and associated sedimentary strata have been investigated in a number of studies (Yang et al., 2020; Hower et al., 2020; Montross et al., 2018; Rozelle et al., 2016; Franus et al., 2015; Seredin and Dai, 2012; Hower et al., 1999), and concentrations typically exceed estimates of average REE concentrations in the upper continental crust (Rudnick and Gao, 2003). In some cases, REE concentrations in coal (on ash basis) are higher than those found in partings (Dai et al., 2018) and can be enriched in the especially important "heavy" REE (HREE: Ho, Er, Tm, Yb, and Lu) over "light" REE (LREE: La, Ce, Pr, Nd, and Sm). During diagenesis and coalification, non-mineral elements (e.g., REEs) and mineral phases are introduced into the coal and adjacent strata. Non-mineral elements can be bound to both the organic macerals and inorganic phases in the coal as described by Dai et al. (2020) and Lin et al. (2017). Both describe the partitioning of LREE and HREE in coal and the different mechanisms that lead to enrichment of HREE over LREE in some coals. Lin et al. (2017) demonstrated a correlation between ash yield, REE concentration, and HREE/LREE ratio in studies of coal samples collected from a preparation plant in Kentucky. Their results indicate that the organic fraction in the coal is enriched in HREE. Other studies have also shown similar trends related to the affinity/complexation of HREE over LREE to organic material in the coal (Lin et al., 2017; Hower et al., 2016a).

While preliminary analyses of data from open-source resources (Ekmann, 2012) show promising concentrations of REE in individual coal samples from a number of sites and basins in the U.S., other sparse data for REE in domestic coal-related strata suggest that many occurrences are low, "subeconomic" concentrations. At present, there is no method for systematically assessing potential sedimentary occurrences of REE. However, the geologic processes responsible for REE occurrences in coal-related strata are systematic; the unpredictability of REE resources in coal-related strata is due to poorly quantified spatial resource trends and the lack of an exploration method tailored to these resources. Thus, there exists a need for a systematic assessment approach that incorporates knowledge of geological variation in the mechanisms of REE enrichment within coal basins to help minimize geologic uncertainty and reduce commercial exploration risk.

2. BACKGROUND

2.1 REE OCCURRENCES

Despite their name, REEs can and do occur in significant concentrations in nature. At present the main sources of REE ore are carbonatite and peralkaline intrusive complexes and their weathered byproducts—termed weathered crust elution deposits (WCED). These types of intrusion-related deposits are found throughout the world. However, in the historical context, few sites have supported economically viable mining. Carbonatites comprise bastnaesite and monazite minerals and generally have high REE+Y (1-10%). Large carbonatite deposits that are predominantly LREE exist within the U.S. at Mountain Pass, CA, originally described in Olson et al. (1954), but these have not provided a stable and secure supply of REE to domestic market that is competitive with foreign sources. Recently, these deposits have received renewed attention with the development of more cost-effective separations (enrichment and recovery) technologies (Anenburg et al., 2020; Weng et al., 2015) and management of waste. Weathered crust elution ore deposits with ion exchangeable REE are sources for all REE (LREE, HREE). The ores are characterized by LREE (0.03–0.25%) and exist within small isolated reserves (Bao and Zhao, 2008; Chi and Tian, 2008). WCED are mined in China successfully due to simple techniques for REE extraction and the economic benefit of recovering critical HREE. Beneficiation from carbonatite and WCED is further challenged by environmental concerns as many deposits contain significant concentrations of uranium and thorium, complicating waste stream management.

2.1.1 REE in Sedimentary Systems

As demand for REE has increased with technological advances over the past decades, other types of geologic systems have been considered as potential sources of these critical minerals. In comparison to traditional bedrock ore deposits, sedimentary REE deposits offer significant potential for domestic feedstocks given the abundance of sedimentary basins within the U.S. (Coleman and Cahan, 2012).

REE occurrences in sedimentary systems are generally classified into two groups: primary- or secondary-type accumulations. Primary-type sedimentary REE accumulations involve contemporaneous deposition of a REE-rich source material and host sediment, such as deposition of volcanic ash within peat, or mineral detritus within channel sand. In contrast, secondary-type accumulations require the following series of events to occur after deposition of a host sediment: mobilization of REE from a source material, transport from the source, and, given suitable geochemical conditions, enrichment of the sediment horizon or zone.

Within the U.S., heavy-mineral sands (HMS) are the main source of titanium feedstock (ilmenite), and also a major source of zircon and monazite, with the latter serving as a valuable REE-Th deposit (Van Gosen et al., 2014). There are two types of HMS deposits, based on their depositional environment: coastal plain and alluvial (stream or river) deposits. Coastal deposits are substantially more voluminous than their counterparts; the largest globally are coastal (Van Gosen et al., 2014). Three of the largest monazite heavy mineral sands deposits in the U.S. are the North Carolina and South Carolina stream (alluvial) deposits, Florida-Georgia beach deposits, and southeastern Idaho stream deposits. Local geology and estimated resources in these districts are well-documented in the literature (Long et al., 2010 and references therein).

2.1.2 REE Occurrences and Concentrations in Coal Systems

Coal and coal utilization by-products are a source of REE and other critical metals and minerals (Montross et al., 2018; Lin et al., 2017; Hower et al., 2016a,b; Seredin and Dai, 2012). Hard coal (e.g., bituminous and anthracite) tends to have the highest REE content of all geologic material (Rudnick and Gao, 2003; Ketris and Yudovich, 2009). For example, global averages for cerium (Ce, used as a metric for LREE concentrations in global reserves) for soft coal (brown lignite) and hard (including bituminous to anthracite) are 22 and 23 ppm, respectively. Typical continental crust values for Ce are 64 ppm (Rudnick and Gao, 2003). Brown and hard coal fly ash, produced from the combustion of coal for power generation, contain nearly 6x this amount of Ce (Ketris and Yudovich, 2009). Heshan coals in southern China are found in late Permian marine carbonate successions and represent another lithofacies in which REE are enriched in high concentrations. The coals are low volatile bituminous coals containing 5–12% sulfur, of which 90% is organic sulfur (Shao et al., 2003; Dai et al., 2013). The coal strata are intercalated with marine carbonates and detrital terrigenous minerals such as quartz and albite. The work by Shao et al. (2003) determined that a variety of factors control the geochemical and mineralogical composition of coal preserved within marine carbonate sequences. The most prevalent factor in this case is multi-stage hydrothermal activity coupled with inputs of organic sulfur and elevated B, Mg, K, Sr, and Rb from seawater.

The relative distribution of REE associated with organic and inorganic material in coal is dependent on the depositional environment and presence of molecules or mineral phases that can capture REE (Given and Miller, 1987). Lin et al. (2017) showed that the REE in coal are not only associated with inorganic component (e.g., detrital minerals), but also the organic matter, and the organic matter is relatively enriched in HREE compared with the inorganic matter. Many coals are slightly enriched in HREE relative to LREE, compared to chondrite and shale (Lin et al., 2017; Eskenazy, 1999). Eskenazy (1999) hypothesized that fractionation of HREE/LREE may be linked to the complexation of HREE with organic compounds that are more stable than those formed by LREE. The authors demonstrated the ability for organic matter to adsorb up to 0.2 mEq/0.5g of xylain, using organic molecules xylain and humic acids; and demonstrated the cation exchange mechanism in which Na, K, Ca, and Mg bound to –COOH and –OH groups were replaced by REE cations. However, there was no difference in HREE/LREE in ion exchange and therefore no evidence that fractionation of HREE and LREE are controlled by organic material in the sample. The experimental work by Eskenazy (1999) and Sonke and Slaters (2006) confirmed that strong complexation of REE occurred when humic substances were present. The coupled results indicate that humic substances play an important role in the fractionation of REE in geologic systems.

Seredin and Dai (2002) proposed four distinct mechanisms for the accumulation of REE in coal:

1. Terrigenous: input by surface water
2. Tuffaceous: connected with falling and leaching of acid and alkaline volcanic ash
3. Infiltrated: meteoric (ground) water driven
4. Hydrothermal: connected with ascending flows of thermal mineral water and deep reduced fluids

Inter-seam tonstein layers have been found in some coal seams around the world (Dai et al., 2014; Spears, 2012; Rozelle et al., 2016; Zhou et al., 2000). Dai et al. (2014) investigated the mineralogy and geochemistry of dacite magma derived tonstein layers in Chinese coal. The tonstein layers were dominated by kaolinite, mixed layer illite/smectite, and minor quartz. Hydrothermal leaching of the buried volcanic ash layer led to the removal of certain ions resulting in Nb/Ta, Zr/Hf, and U/Th ratios much lower in the tonsteins than in the adjacent coal beds or floor/roof rock. Pennsylvania-aged fire clay coal is a source of REE (Hower et al., 2020; Zhang et al., 2018) and is distributed across the central and southern Appalachian basin, predominately concentrated in the Central and Southern coal regions of WV, KY, and AL. Coals within fire clay seams have distinct REE and Sc concentrations that are related to the position of the lithotype within the coal seam (Hower et al., 2020). Geochemical relationships such as the ratio of light to heavy REE are also related to the lithotype position within the coal seam. Hydrothermal alteration may also play a role in the enrichment of REE in certain layers that are in proximity to the tonstein (Hower et al., 2020; Hower et al., 2016b). Seredin and Dai (2012) proposed four modes of REE enrichment in coal to go along with an organic association. Kentucky fire clay coal is enriched in REE and other critical minerals via emplacement by volcanic ash fall into the coal mire. Researchers have demonstrated these tonstein layers can be pervasive horizontally throughout the coal seam and partings (Hower et al., 2018) and are the source of REE in the rock partings and within the coal itself (Seredin and Dai, 2012; Hower et al., 2018). The predominant mechanism of REE enrichment in the fire clay is from tuffaceous/volcanic, infiltrational/leaching, and hydrothermal processes that began in the coal depositional environment during coal formation and continued through subsequent diagenetic (e.g., dissolution and weathering in the subsurface) events (Hower et al., 2016a).

2.2 GEOLOGIC RESOURCE ASSESSMENT METHODOLOGIES

Geologic systems-based analytical methods are proven tools for predicting subsurface properties, having served over several decades as key tools for discovering and assessing *in situ* geologic resources. For example, from the 1800s to the 1970s oil and gas drilling success rate was 50% or lower. After development of the petroleum system concept in the 1980s and other technological breakthroughs, exploration success rate increased from only 60% to above 90% at present (Cochener, 2010). The endeavor to systematically predict, find, quantify, and characterize REE accumulations in sedimentary carbon ore deposits can be accelerated based on lessons learned from decades to centuries of geologic and geochemical data, and science knowledge. In this section, existing resource assessment methods and complementary analytical approaches are reviewed and summarized to provide the basis for formulating the U.S. Department of Energy's (DOE) National Energy Technology Laboratory (NETL) **U**nconventional **R**are earth and **C**ritical minerals (NETL-URC) model method.

Geologic resource assessment methodologies are used for exploration and quantification of critical resources, such as precious metals, coal, or petroleum. Any assessment methodology begins with an understanding of the geologic processes and systems that result in the deposition of the target resource. This knowledge base provides a conceptual foundation or framework to identify the appropriate datasets and analytics to develop the methodology. Data and knowledge frameworks on these systems are then applied using analytical techniques to either determine the prospectivity of the commodity within an area and/or obtain volumetric ore estimates.

Building a knowledge framework for a given mineral or hydrocarbon resource involves a spatial and temporal understanding of the geologic processes or components that contribute to or are required for accumulation. This typically requires an understanding of the source, transport mechanism, and containment for a given resource. For example, the petroleum system is a conceptual process model that considers the origin and evolution of the geologic system that produced—or at least was capable of producing—an accumulation of hydrocarbons (Magoon, 1988). Application of this concept relies on the existence of a genetic relation between timing of deposition, source rock, reservoir rock, seal rock, trap formation, and thermal maturation (i.e., the critical moment). Another type of knowledge framework relies less on the interdependencies between these processes or components, but instead focuses on the spatial and temporal coincidence of the critical independent contextual factors for ore genesis (Hronsky and Kreuzer, 2019). Factors for this approach include favorable lithospheric architecture, fertility (favorable geochemical conditions, transient favorable geodynamics; i.e., tectonic/stress conditions over time), and ore preservation (of primary depositional zone) (McCuaig and Hronsky, 2014). The development of a knowledge framework may depend on the availability of knowledge or data for a specific mineral resource.

Information beneficial for a regional evaluation of mineral resources includes: 1) well-defined key descriptive terms; 2) geologic map data, to identify genetically related geologic map units and determine the geologic history; 3) mineral occurrence data, to establish attributes and locations of known mineral deposits, and relate deposits to host rocks and tectonic origins; 4) geochemical and geophysical data, to locate chemical anomalies of pathfinders on the surface and interpret the subsurface structure, respectively; and 5) exploration history and mineral deposit models of known deposits in the region, to understand outcome of previous assessments and infer the potential of nearby undiscovered deposits (Nokleberg et al., 2007; Lipin and Bawiec, 2000). Geological estimates of the deposit, such as grade, tonnage, and spatial extent, can (and often do) introduce significant amounts of error and uncertainty in assessing mineral resources and ore reserves (Dominy, 2002). Ultimately, these types and quantities of data can inform analytical techniques to map and quantify ore estimates.

For many decades mineral resource assessments from the U.S. Geological Survey (USGS) have followed, almost exclusively, a three-part quantitative assessment methodology. This approach, now seen as the “traditional approach” for quantitative assessments of nonfuel mineral deposits, uses known mineral site locations and deposit resources in combination with associated geologic features to predict regions that potentially host undiscovered mineral deposits (Singer, 2010). The first step in this approach is to delineate geographic regions that appear favorable for the occurrence of mineral deposits (e.g., Pb-Zn) based on known deposits, mineral deposit models, and geologic regimes. Favorable regions identified in this step are called permissive tracts; it is necessary to use a geologic map to delineate these regions. Next, the number of undiscovered deposits which may exist in the permissive tract is estimated using probabilistic estimates. Finally, the probable amount of the undiscovered mineral resource within the permissive tract is calculated using Monte Carlo simulations based on grade-tonnage models of known deposits and the number of undiscovered deposits estimated in the previous step (Singer, 1993, 2010). In recent years, resource assessment methodologies have expanded beyond the “traditional approach” to include remote sensing techniques (Zarasvandi et al., 2008) and more advanced mathematical methods (Bardossy et al., 2003). Zarasvandi et al. (2008) used multispectral remote sensing techniques to complete reconnaissance mapping for karst bauxite deposits across the Zagros Mountain Belt, which is remote and treacherous to map on-foot. The authors also compiled a database of existing geological and geomorphological data for the assessment. The data from both techniques were input into GIS to prepare maps, from which the authors determined potential mineral prospects. Hammarstrom and Dicken (2019) present an assessment method that characterized different areas of the U.S. by “focus” area in order to collect data on the distribution of critical minerals and REE. The assessment model is based on the distribution of non-fuel mineral systems and REE-bearing deposits that are identified within three focus zones located in the contiguous U.S. and Alaska. The deposits are determined by characteristics of data collected on mineral assemblage and other meta-attributes of samples and GIS mapping of deposits within each zone.

These analytical techniques can be categorized into data- and knowledge-driven methods to integrate known mineral occurrences with spatial evidence layers (i.e., proxy maps). When there is a relatively large number of known mineral occurrences and their relationships to explanatory datasets are well known within a study area, data-driven modeling techniques such as neural networks (Zaremotlagh and Hezarkhani, 2017; Wang et al., 2011), logistic regression (Xiong and Zuo, 2018), Naïve Bayes (Zaremotlagh and Hezarkhani, 2016), weights of evidence (Nykanen et al., 2008), and Monte Carlo simulations (Lysytsyn , 2015) may be appropriate. Data-driven techniques also allow for the quantification of uncertainty due to statistical randomness. Knowledge-driven techniques rely on expert knowledge due to few or no known mineral occurrences within a study area, thus incorporating uncertainty due to ambiguity of data or lack of knowledge (Lysytsyn, 2015). Common knowledge-driven methods successfully applied specifically to delineate potential mineral targets are variations of fuzzy logic (Lysytsyn, 2015; Abedi et al., 2017; Levaniemi et al., 2017; Dag and Mert, 2008; Nykanen et al., 2008; Yousefi and Carranza, 2015) and various ranking and weighting techniques (Karl et al., 2016). For example, Bardossy et al. (2003) assessed karst bauxite deposits in Hungary using the fuzzy set theory. A key advantage of the fuzzy set theory is the ability to express uncertainty at different levels of possibility and the ability to evaluate both quantitative and semi-quantitative inputs. Hybrid data- and knowledge-driven methods have also been proposed probabilistic fuzzy logic for example (Lysytsyn, 2015).

As data science capabilities to process and analyze big data evolve, so do methodologies that can further improve predictions for poorly constrained resources through the integration of data science. The Subsurface Trend Analysis (STA) method developed by (Rose et al., 2020) is a systematic, science- and data-driven method that utilizes geologic systems knowledge to inform statistical analyses of subsurface properties and resources. The workflow integrates geologic data used in traditional assessment approaches (e.g., geologic maps, in situ core) with appropriate statistical methods (e.g., geostatistics, dimensional analysis) to characterize the subsurface property of interest. Moreover, machine learning and data mining techniques allow these assessments to efficiently collect and integrate large, disparate datasets for better informed analyses.

The development of a systematic approach for predicting and quantifying the occurrence of REE in coal and related strata requires: (1) knowledge and data associated with REE concentration mechanisms, and (2) integration of key elements from previous methods.

3. DISCUSSION: REALIZATION OF THE REE-SED METHOD FOR COAL SYSTEMS

3.1 KNOWLEDGE-DATA FRAMEWORK

The NETL-URC model foundational knowledge-data framework includes an understanding of the systematic nature of mineral accumulation in sediments and leverages data related to the processes of critical mineral and REE accumulation and *in situ* rock properties. In this section, a knowledge-data framework is presented for applications of the NETL-URC model in coal sedimentary systems. Although the model and supporting database as presented herein align to coal-REE assessments, the method is highly adaptable and applicable for predicting and assessing REE occurrences in other sedimentary systems as well.

For purposes of convenience, the coal-REE knowledge-data framework reclassifies the genetic types of coal-REE accumulation defined by Seredin and Dai (2012) into six distinct occurrence types. Occurrences differ by the timing of REE enrichment relative to sediment deposition, REE transport mode, and resulting REE phase (Table 1). Conceptual deposit models developed for the various coal-REE occurrences type represent the knowledge of how and why REE accumulate in sedimentary systems. Similar to the conceptualization of other mineral deposit models (Cox and Singer, 1986) or the petroleum system, the coal-REE deposit models involve unique combinations of REE sources and accumulation processes (Figure 1). Additional geological factors such as depositional history, tectonic and diagenetic influences, and geochemical alteration are also represented in the models as individual geologic components. Since the coal-REE deposit models comprise the core of knowledge-data framework, it is critical that the models accurately depict the various accumulation mechanisms. Accordingly, the components and relationships defined in the deposit models are the culmination of an extensive literature review on the current state of knowledge of the coal-REE system (NETL, 2021a). As the scientific understanding of REE occurrences in coal sedimentary systems continues to evolve and improve, these deposit models will be refined, and additional deposit models may be developed.

Having developed the coal-REE deposit models, the key geologic components needed for REE accumulation can then be assessed per the model using a data science approach, such that associated data can be queried to test for evidence that a process or component occurred in the geologic past. Formally defining the relationships between the datasets and process of sedimentary REE accumulation is vital to a successful assessment at the basin, mine, or stratigraphic layer scales.

Table 1: Classification of the Various REE-Coal Occurrence Types as Considered by the NETL-URC Model (Adopted and modified from Seredin and Dai (2012))

Timing of REE Enrichment	Transport Mode	REE Phase	Occurrence Type
Syn-depositional	Eolian	Allogenic mineral	Primary – Eolian
Syn-depositional	Fluvial	Allogenic mineral	Primary – Fluvial
Post-depositional	Meteoric fluids	Ion adsorption	Secondary – Meteoric Adsorption
Post-depositional	Meteoric fluids	Authigenic mineral	Secondary – Meteoric Precipitation
Post-depositional	Hydrothermal fluids	Ion adsorption	Secondary – Hydrothermal Adsorption
Post-depositional	Hydrothermal fluids	Authigenic mineral	Secondary – Hydrothermal Precipitation

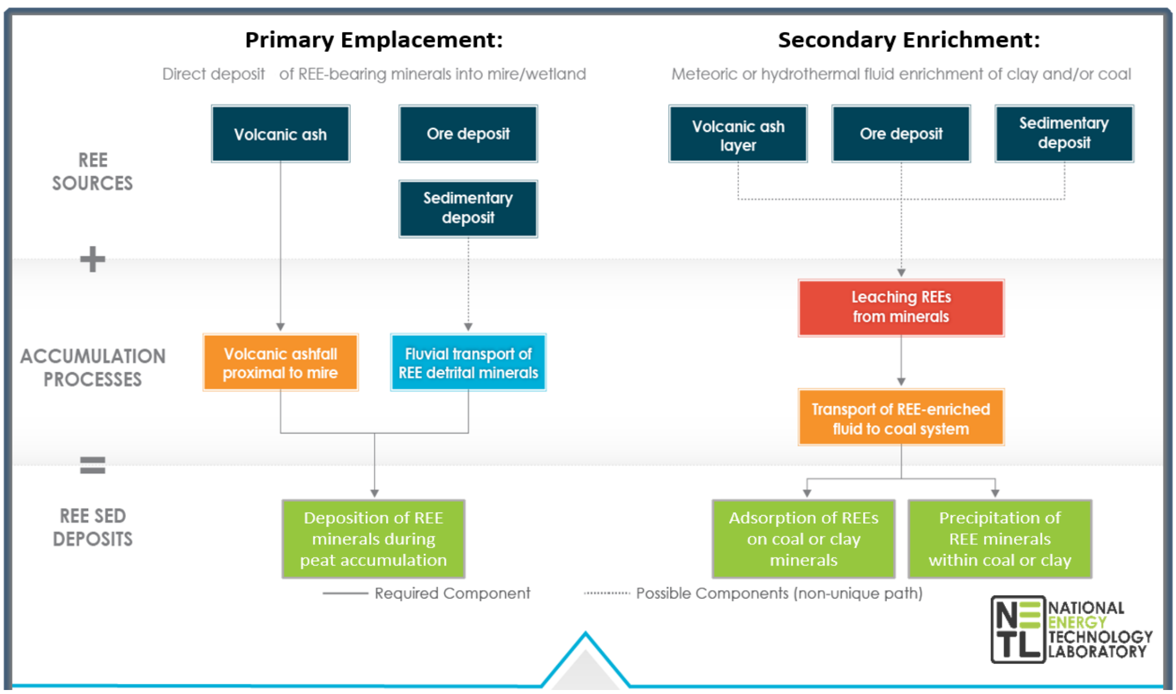

Figure 1: Flowchart of pathways for primary- and secondary-type occurrences of REE in sedimentary geologic material. Each unique pathway (arrow path) involves a source of REE and an accumulation process that can result in an REE sedimentary deposit. Boxes are a simplification of the geologic components assessed by the NETL-URC model, as per the NETL-URC method.

During formulation of the NETL-URC model, a collection of publicly available resources was compiled to support testing and development. This data collection, named the REE Coal Open Geodatabase (NETL, 2018), comprises both spatial and non-spatial geological, geochemical, and geospatial datasets spanning across the U.S. Key data include surface lithology, mineralogy, structural geology, geochemical analytical samples, sediment provenance, and paleogeographic reconstructions, from sources such as the USGS, U.S. DOE, state geologic surveys, and other government agencies. The collection is available for download via U.S. DOE NETL's Energy Data eXchange (EDX) REE Coal Open Geodatabase (NETL, 2018).

The abundance of available data varies significantly across the U.S. (Figure 2). Visualizing where data are available prior to implementing an assessment can help to identify suitable locations. However, given the proprietary nature of most local or mine scale coal data, the REE Coal Open Geodatabase is mostly applicable for basin scale analyses (Figure 2). Note the relative abundance of data in the Powder River Basin is largely due to the latest USGS coal resource assessment (Kinney et al., 2015; Luppens et al., 2015).

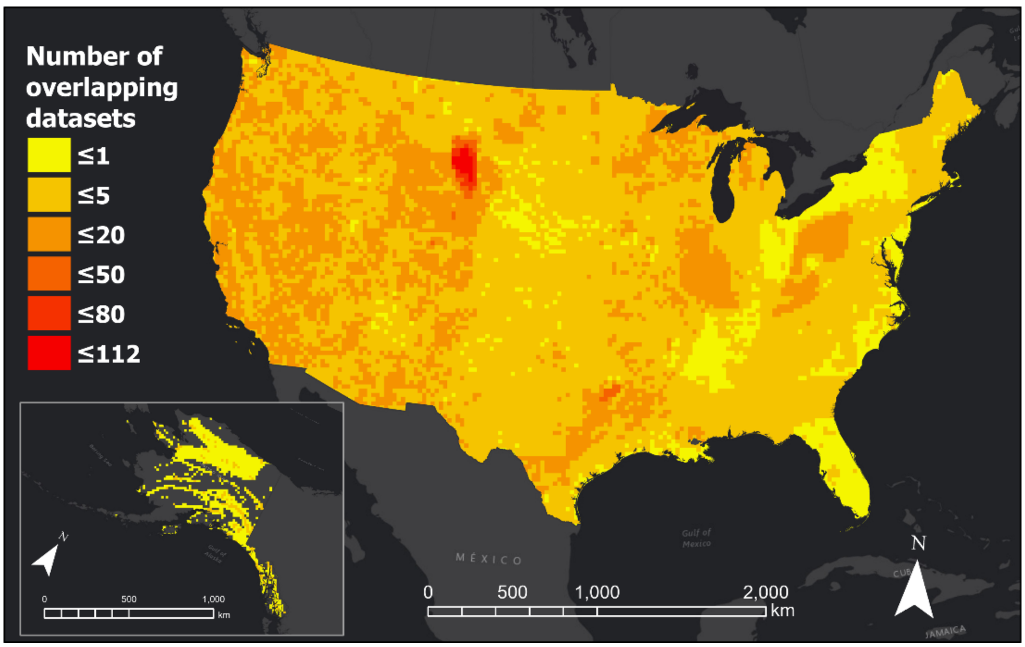

Figure 2: Count of overlapping datasets within the REE Coal Open Geodatabase (NETL, 2018), collected for development and testing of the NETL-URC model. Grid cells are 25 x 25 km. Warmer colors (shades of red) indicate a greater number of available datasets. Analysis performed using NETL's Cumulative Spatial Impact Layers tool (Romeo et al., 2019).

3.2 APPLYING THE KNOWLEDGE-DATA FRAMEWORK TO THE ANALYTICAL METHOD

The NETL-URC model entails two primary aspects: geological characterization and spatial/volumetric assessment. Geological characterization involves reconstructing depositional, structural, and diagenetic history of the region of interest using geological, geochemical, and geophysical data. The characterization aspect of the assessment informs of locations where past conditions were favorable for REE accumulation, presented as a "Potential Enrichment" (PE) Score. In this context, the term enrichment is used inclusively to represent both pre- and post-depositional accumulation of REEs in the sediment. The spatial assessment aspect incorporates results from the geologic characterization with REE concentration data, coalbed geometry and/or volumetric calculations, and other relevant spatial data into a geostatistical interpolation method to estimate the inferred resource quantity. In areas with no REE measurements in close proximity, only the PE Score is applied to predict regions of higher REE accumulation potential.

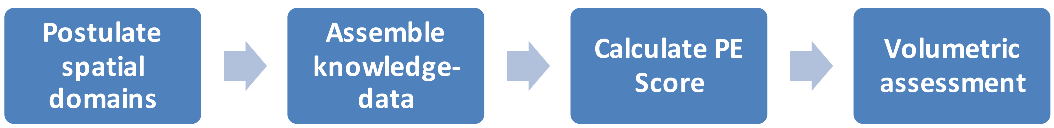

Figure 3: Generalized workflow for implementing the REE-SED assessment method.

In practice, implementation of the assessment method occurs in four stages, with the first three addressing geological characterization of the assessment area and the last stage integrating the available information and applying a spatial (volumetric) calculation to estimate the resource quantity (Figure 3). The NETL-URC model workflow is summarized as follows:

Step 1: Postulate spatial domains on the basis of a common geologic attributes. This involves characterizing the geologic history of the region of interest and identifying areas with common geologic histories, such as basin sediments with shared provenance or regions with similar structural style. Domains are defined using geologic knowledge about three primary factors affecting subsurface properties: lithology, structure, and secondary alteration (Figure 4). Each unique spatial domain identifies areas with a distinctive geologic history (Figure 4).

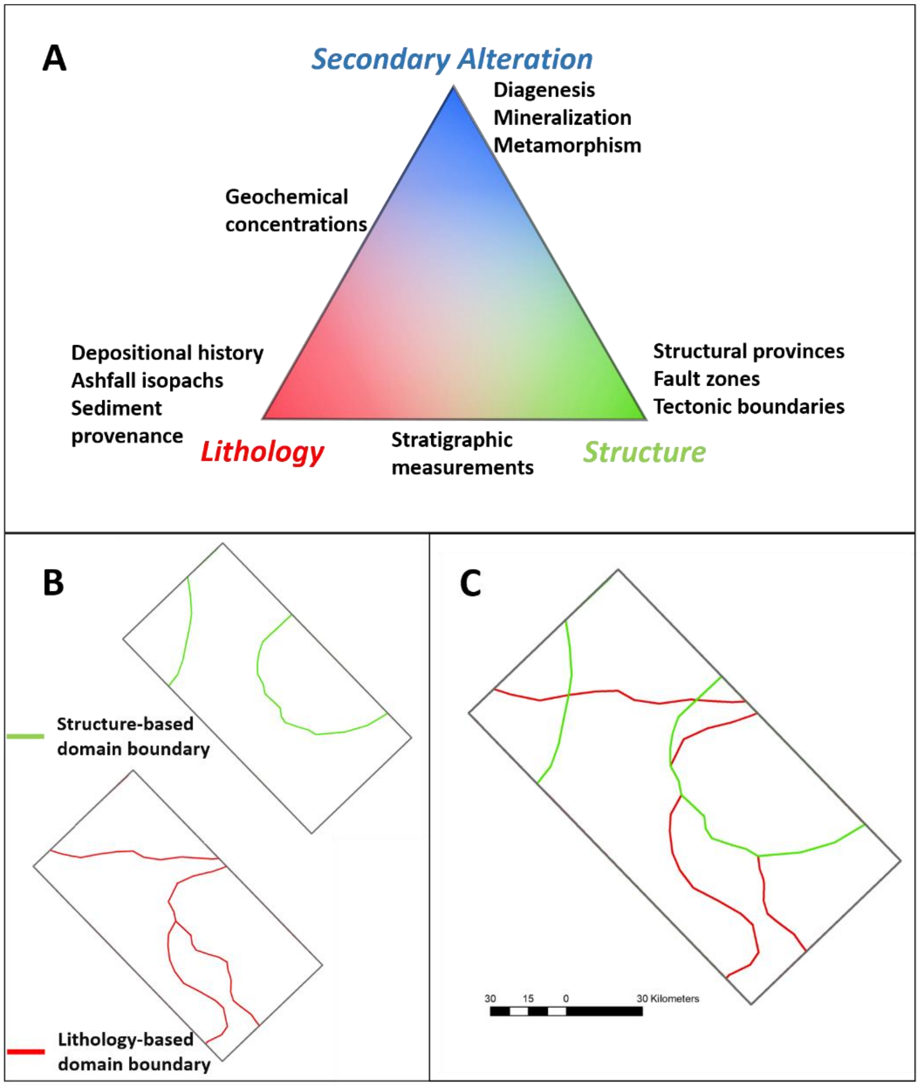

Figure 4: Example application of the Subsurface Trend Analysis method for postulating genetically related spatial domains in Step 1 of an assessment (see detailed discussion in Rose et al., 2020). (A) Ternary diagram with the primary factors affecting subsurface properties as end-members (colored text). Spatial domains are postulated for each geologic end-member. Black text lists example data and information utilized in an assessment, plotted according to association with geologic end-members (i.e., "fault zones" information provides constraints for structure domains). (B) Delineated spatial domains for an anonymous test area. Grey box is the assessment area boundary, colored lines are delineated domain boundaries based on available data and knowledge for the given domain type (structure, green; lithology, red). (C) Aggregation of unique lithologic and structure domains from (B), for use in later steps of the assessment method.

Step 2: Assemble and inventory knowledge-data for the coal-REE deposit models to assess limitations of data availability. Like any other resource assessment or modeling effort, the REE-SED method requires data. A "Data Availability" (DA) metric is calculated to represent the amount of uncertainty in the assessment due to missing or incomplete information. The metric is simply a ratio of the number of geologic components with related data versus the total number of components for the occurrence type (e.g., Primary – Eolian). In this step, individual components in the NETL-URC model are related to datasets to establish the occurrence of required geologic components, as part of the geologic characterization. Each dataset used in the assessment must be related to at least one geologic component. For example, observation of a volcanic ash layer in coal core represents a potential REE source, thus the observation can be related to that component. Some components are not directly testable and thus have no related data. For example, it is challenging, if not impractical or impossible to acquire data that evidences a mire was downwind of contemporaneous volcanic activity. In select cases such as this, evidence for non-testable components can be inferred. In the ash-bearing coal example above, presence of the ash layer implies the (then) mire was downwind of contemporaneous volcanism. This also allows multiple geologic components to be represented by a single observation or dataset. Implicit relationships are also utilized to assign an occurrence of geologic components for an area. Considering again the ash-bearing coal example above, given a sufficiently thick ash and proximity to volcanism and no tectonic influences, one can assume the ash layer extends beyond the immediate vicinity of the core. Thus, in the absence of data suggesting otherwise, occurrence of the ash can be inferred for local area around the cored site. Implicit assignment of a geologic component based on spatially explicit (i.e., coal core) data permits an assessment of an area or volume using discrete measurements. Uncertainty associated with these assumptions is in the subsequent step.

Step 3: Calculate PE score using prospectivity analytics to assess potential REE occurrences. Component datasets are queried for evidence of coal REE occurrences based on geological interpretation of the data. For example, core descriptions could be queried for observations of ash horizons, which provides evidence of a possible REE source. For each unit area, a PE Score is calculated that indicates the number of components with supporting data versus the total number of components for each occurrence type. Each REE-coal occurrence type is assigned a PE Score value ranging from zero to one. Zero indicates no evidence for favorable conditions, one indicates supporting evidence for all required components, and values in between indicate supporting evidence for a portion of the required components. For a given area (cell) or volume (voxel), the PE Score is equal to or less than the value for DA. Results from this step indicate locations with potentially undiscovered REE occurrences and also supplement the spatial/volumetric assessment (Step 4).

Uncertainty is accounted in this step using a spatialized component additive method with fuzzy logic. Each component of a coal-REE deposit model may be represented by one or more datasets that are either explicit or implicit (Figure 5). Explicit data include data with exact locations and representations of geology or geochemistry that support evidence of REE enrichment. Implicit data represent combinations of structural, lithologic, and secondary alteration domains (or general areas) that are more likely to support REE enrichment. To better represent the influence of these data, fuzzy logic is employed to weight their importance (as evidence of REE enrichment) across the study area. Fuzzy representations of explicit and implicit data are combined to represent the component itself and the associated uncertainty (Figure 5). Fuzzy component representation is then created by adding the implicit and explicit fuzzy weighted data. The additive approach allows the explicit data to carry more importance than the implicit data and better prioritizes known evidence for each component within the deposit model. Conversely, the uncertainty of each component subtracts the explicit fuzzy representation from the implicit fuzzy representation. Use of a subtractive approach allows the location of the explicit data to have the lowest uncertainty, and if the implicit data does not spatially coincide with explicit data, those locations will have the highest uncertainty (Figure 5). For each component these two outputs are then applied to the deposit model to obtain the final PE Scores and associated uncertainty (Figure 5).

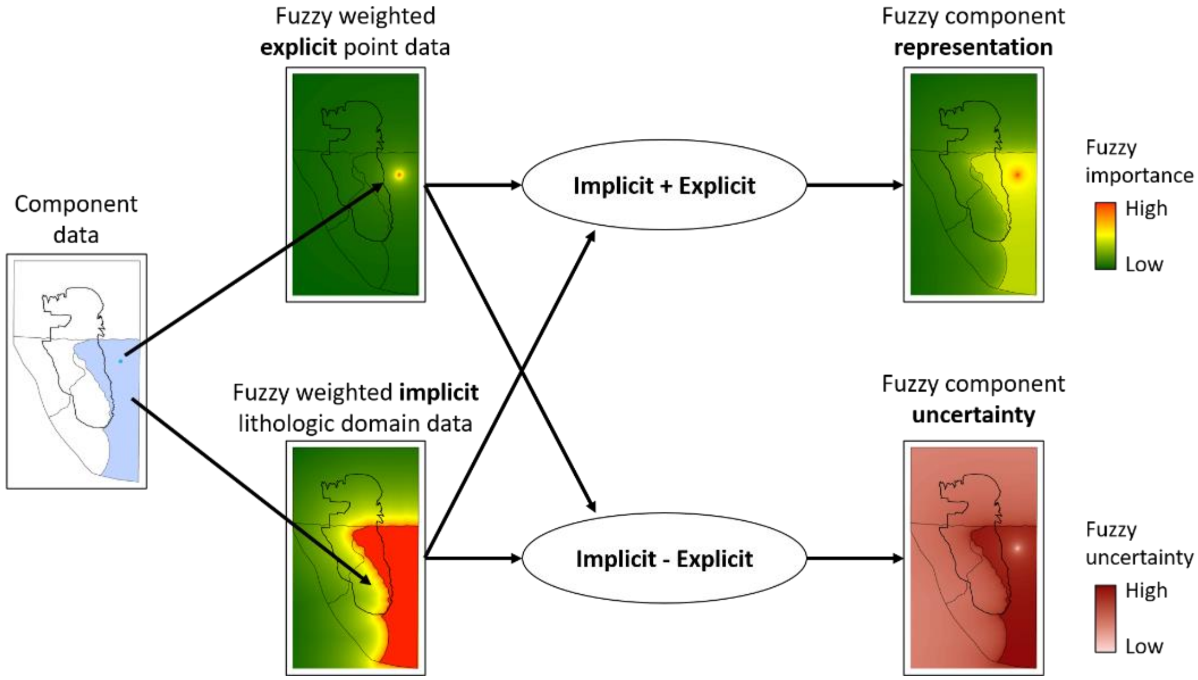

Figure 5: Example fuzzy logic application for representing implicitly assigned component data and associated uncertainty in Step 3 of the NETL-URC model. See Step 3 above for detailed discussion.

Step 4: Spatial/volumetric assessment to estimate inferred resource quantities. First, PE Scores and REE concentration data are evaluated for spatial autocorrelation to determine whether values are spatially clustered, dispersed, or randomly distributed. Evaluations are performed for the area of assessment and within individual spatial domains. In domains without REE concentration measurements, the PE Score is integrated with coal volume estimates to produce maps of potential resources with varying degrees of certainty. Where REE measurements are sufficiently available, REE concentrations in areas with highest PE Score and a subset of validation data (geochemical measurements from rock samples) are interpolated to assign enrichment potential. Interpolations are only applied in areas with moderate to high confidence in the PE score. If no data exists to interpolate REE concentrations in areas of high PE Score, rankings of high vs low PE Score and confidence indicate a qualitative metric for REE enrichment potential. Implementation of the NETL-URC model involves a multistep workflow (Figure 3).

A simpler, straightforward approach to characterizing *in situ* REE resources would be applying a geostatistical algorithm such as inverse distance weighting to interpolate between measured REE concentrations in coal core samples over an area or zone of interest. However, there are major drawbacks to this approach. First, few samples have the information needed and those samples are not distributed evenly across U.S. coal basins (Figure 6). For example, the USGS CoalQual database (https://ncrdspublic.er.usgs.gov/coalqual/) (Palmer et al., 2015) is considered a preeminent dataset for REE concentrations in domestic coal basins, yet these data were collected for a different original purpose, and the homogenized nature of the analyses significantly limits the utility of that data for these types of assessments. Second, and perhaps more importantly, the simple interpolation approach neglects valuable information that can be gleaned from important contextual data. For example, interpolating across geologic domain boundaries (e.g., across a fault zone or across areas with vastly different lithologies) ignores changes in rock properties that may affect REE accumulation. Conversely, as demonstrated by the success of the petroleum systems concept in petroleum exploration, incorporating geologic contextual data into an assessment can significantly improve results. With knowledge of the geologic mechanisms of REE accumulation in coal, one could predict where REE likely occur in significant concentrations for a given area that experienced favorable conditions for accumulation, without having directly measured the rare earth concentrations. This consideration enables an assessment of potential enrichment for areas with sparse or even no REE measurements. Thus, the NETL-URC model involves analysis of both measured geochemical concentrations (i.e., REE and other critical elements) and contextual data and information.

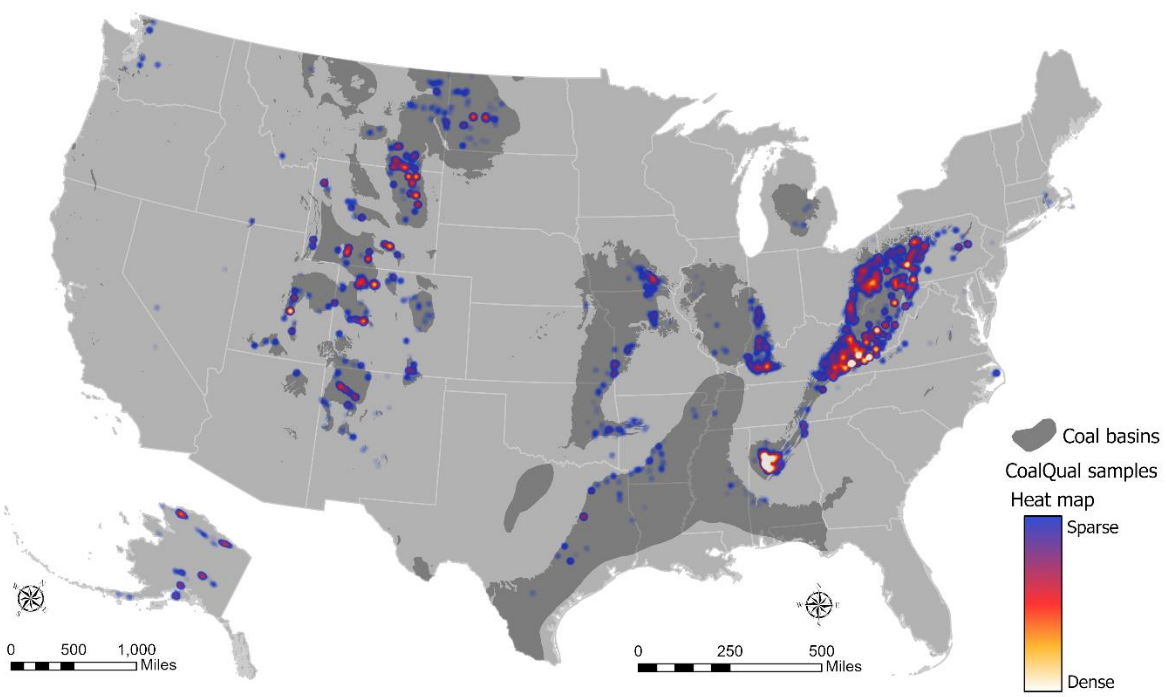

Figure 6: Heat map displaying the density of CoalQual samples (Palmer et al., 2015) in U.S, coal basins (dark gray). Warmer colors indicate a higher relative density of coal samples.

3.3 CURRENT STATUS

The beta version of the NETL-URC model is in the final development stage and three major components have been advanced and integrated. The components are:

1. Integration of raw data collection into structured machine-readable assessment "ready" databases (DA and DS)
2. Calculation of PE Score for all REE enrichment mechanisms
3. Application of fuzzy logic to better represent spatial and geologic uncertainty of the input datasets

The tool was enhanced by modifying application scripts using open-source python libraries in order to optimize data processing and reduce overall run time. Other enhancements include the development of python script to process NETL-URC model data for sensitivity analysis for input into the NETL-URC model beta-tool. The enhancements made to scripting and data processing have increased utilization of existing data by 23% by applying new geologic constraints and refining select NETL-URC model algorithms, which leads to improved QA/QC protocols to increase processing efficiency and ensure data are correctly represented.

3.4 NEXT STEPS: DEMONSTRATION AND VALIDATION

NETL's partnership activities with extramural stakeholders will continue to focus on identifying prospective unconventional REE/CMs from domestic mine byproducts that are relevant to domestic supply. While the resource assessment efforts target identifying in-place, technically and potentially economically recoverable scales of evaluation at the mine, seam and basin scale, they rely upon finer-scale data and studies as well. Core, outcrop, and mine scale data and information are key to constraining and understanding how and why unconventional REE/CMs form in these types of systems, and the processes and conditions that result in the vertical and lateral heterogeneity of these deposits. In addition, contextual geologic data and information at these finer scales is important for constraining the lithologic, structural, and secondary alteration history of these systems to improve predictions of the occurrence and concentrations of unconventional REE deposits at all scales. At present, much of these data and knowledge either does not exist in forms appropriate for supporting unconventional REE/CM assessments or is incomplete. For example, the U.S. Geological Survey CoalQual database (Palmer et al., 2015) is often cited as relevant to meeting these needs, but was collected for a different original purpose, and the homogenized nature of the analyses significantly limits the utility of that data for these types of assessments.

For coal and related strata, coal-byproducts, and industry waste streams, it is also key to understand the human engineering and history of these systems as well. With sufficient data and knowledge from dedicated studies of these systems, the predictability and accuracy of the unconventional rare earth and critical minerals model will be greatly improved. To mitigate current data gaps, a template for sample data is provided to guide future data collection efforts (NETL, 2021a,b). The template is a spreadsheet for the data and information needed for geological samples used in the NETL-URC model. Relevant metadata categories include sample identification (unique ID numbers), sample description (sample, type, thickness, age), geolocation (latitude, longitude, depth), site characteristics (owner, geologic features, associated lithologies), collection information (lab name, data analyzed, etc.), other (various contact and source information), and chemical analyses (elemental, proximate, ultimate, etc.). The metadata also includes a "level of importance" designation for each field (or attribute) contained within the template. Of most importance is if a field is labeled as "critical", these fields are the minimum requirements for each record (or sample). Sample data can be entered in the "Sample_Metadata" and "Chemical_Analysis" tabs.

4. CONCLUSION

The NETL-URC model is a big-data, machine learning enabled geoscience approach to improve prediction and identification of high concentration deposits of unconventional CM and REE in sedimentary, carbon-ore based systems. Underpinning the NETL-URC model is an understanding that occurrences of sedimentary natural resources are not random, but rather a product of systematic geologic processes and unique syn- and post-depositional histories. The holistic approach utilizes a knowledge-data framework to apply knowledge of how REE accumulate in sedimentary systems using various geologic and geospatial data-driven methods. Currently, no other fully developed methods or tools exist that are designed to model or simulate the complex behavior of REE and mineral phases within coal and related lithofacies.

Resource assessments using approaches such as a the REE-SED method are key to the successful identification of promising geologic deposits that host the resources. Key insights and data about unconventional REE and CM resource estimates, host materials, and ore minerology are valuable to the development of commercially viable extraction and separation technologies. The assessment model data can also aid in the development of technological and systems-economic assessments that are critical to aspects of DOE’s research portfolio and for policy and commercial decision making.

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