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Title: **A Field Survey of Rare Earth Element Concentrations in Process Streams Produced by Coal Preparation Plants in the Eastern United States**

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ABSTRACT

A field study was undertaken to experimentally measure the concentrations of rare earth elements (REEs) contained in the process streams generated by a group of 20 coal preparation plants located in the eastern United States. For each site, representative samples of clean coal product, coarse refuse and fine refuse were collected. Each sample was then partitioned into preselected size and density classes by wet screening/sieving and float-sink testing. The resultant products were dried and subjected to laboratory analyses to determine ash contents and rare earth element concentrations including Yttrium and Scandium. A detailed analysis of the database generated by this exercise showed that coal-based products from these preparation plants contained significant quantities of rare earth elements. In particular, the coarse refuse streams currently discarded by the 20 plants examined were found to contain a sufficient tonnage of REEs to satisfy the current domestic demand for these important elements. The data also showed a strong positive correlation between ash content and REE concentration, which suggested that the primary association of REEs in eastern U.S. bituminous coal sources is likely within fractions containing inorganic impurities. This association was well described using a simple power equation relating ash content and REE concentration. The ratio of heavy-to-light rare earth elements was discovered to be significantly higher in organically-rich fractions of clean coal, suggesting that mineral impurities intimately associated with carbonaceous matter have elevated concentrations of heavy rare earth elements. A similar trend was observed for a grouping of rare earth elements (Y, Nd, Eu, Tb, and Dy) that are likely subject to near-term supply shortages. Finally, the database showed that a linear correlation existed between La for many REEs of interest, although there were several notable exceptions for some high-value REEs (Lu, Pr and Tb).

KEYWORDS

Rare Earth Elements, Critical Elements, Coal Preparation, Coal Characterization

INTRODUCTION

Rare earths elements (REEs) are a series of 17 elements with similar chemical properties that appear together in the periodic table. As shown in Table 1, REEs commonly include the 15 elements in the lanthanide series along with Scandium and Yttrium. Although REEs are relatively abundant in the earth's crust, they are rarely found in mineable concentrations that are sufficiently high to justify commercial extraction, processing and refining (Beauford, 2014). This is unfortunate since REEs are critical to the manufacturing of advanced technologies used in the modern world due to their unique electrochemical, luminescent and magnetic properties (Van Gosen, 2019). REEs are required to manufacture a variety of consumer products such as portable computers, rechargeable batteries, mobile phones, fluorescent lights, digital cameras, global positioning systems, and sports equipment. REEs are also critical for national security since they are needed to manufacture high-tech instruments used in the defense industry such as night-vision goggles, range-finders, precision guidance systems, stealth devices and communication equipment. The *Critical Materials Strategy Report* published by the U.S. Department of Energy identified eight REEs that are essential for the development of clean energy technologies such as wind mills and hybrid electric cars (Fromer, 2011). Five of these REEs (Dysprosium, Neodymium, Terbium, Yttrium and Europium) have been identified as highly critical and vulnerable to shortages (Bauer et al., 2012). As such, the continued development of the nation's advanced manufacturing, defense and information industries will require a reliable and ever-increasing supply of the important class of chemical elements (DOE, 2017).

In terms of global production, approximately 120,000 metric tonnes of rare earth oxides (REOs) were produced in 2016, with China responsible for 85% or more of all production (Zhou et al., 2017). Of the more than 250 known minerals containing rare earth elements, less than 10 accounted for all of the REEs commercially produced to date (Castor and Hederick, 2006). Two of the largest rare earth mines, i.e., the Bayan Obo and Maoniuping mines in China, produced bastnaesite ($LnCO_3F$) concentrates, with small quantities of monazite ($(Ln,Th)PO_4$) and xenotime (YPO_4). China has also developed large extraction operations in the South China block to recover ion-adsorption REEs from clay deposits, which are predominately linked to the weathering of granite (Xie et al., 2016). Despite the low REE grades (0.1-0.2%), the ion-adsorption clays account for about 35% of the China's total REE output and more than 80% of the world's production of heavy rare earth elements (Moldoveanu and Papangelakis, 2016).

The major role that China plays in the supply of REEs and the volatility in export policies have generated interest in developing domestic resources for reliably supplying REEs and other critical materials (Lin et al., 2017a; King, 2019). One pathway that has garnered considerable attention during the past several years is the potential extraction of REEs from coal and coal byproducts. The coal database maintained by the U.S. Geological Survey (USGS, 2014) indicates that coal seams mined in the U.S. contain elevated amounts of REEs relative to crustal concentrations. Recent analyses conducted by Leonardo Technologies (Ekmann, 2012) showed that the domestic coal industry has the potential to produce sufficient REEs to supply the entire U.S. demand if appropriate separation technologies can be developed to extract these valuable elements. In light of this potential, the authors of this article undertook a detailed field study to evaluate whether the nation's fleet of existing coal preparation facilities have the potential to play a key role in facilitating the development of this new resource. Modern coal preparation plants, which utilize a variety of low-cost physical separation processes to efficiently segregate organic and inorganic matter in run-of-mine coals (Noble and Luttrell, 2015), have the potential to generate clean coal and/or waste refuse products that are preferentially enriched in REEs. The data generated from the current study was used to (i) determine the tonnages of REEs present in clean coal and waste refuse streams, (ii) examine associations between common coal quality indicators (e.g., ash content) and REE concentrations and (iii) develop statistical correlations for associations of different REE groupings (light, heavy and critical REEs) contained in the coal-based feedstocks. This article summarizes the field work, experimental data and engineering analyses performed as part of this investigation.

EXPERIMENTAL

Representative samples of products generated by 20 different coal preparation plants were collected for use in this study. The plant locations were distributed throughout the eastern states of Kentucky, Pennsylvania, Virginia and West Virginia. Preparation plants located in southern West Virginia include plants 1, 5-6, 8-11 and 20. Plants located in central/northern West Virginia include plants 2, 4 and 7. Plants located in western Pennsylvania consist of plants 12-15, while plants located in Virginia include plants 16-19. Plant 3 was located in eastern Kentucky. For all sites, individual samples were taken from the plant feed, clean coal, coarse refuse and fine refuse streams. Figure 1 provides a generic flowsheet indicating the locations of sampling points used for each coal preparation facility.

Standard ASTM sampling procedures were employed to obtain, split, transport, and store all samples. After collection, the samples were air-dried and split into four or five particle size classes that best represented the process circuitry at each plant. Table 2 shows the specific size classes utilized for each plant. All of the size classes larger than 0.15 mm (100 mesh) were subjected to float-sink analysis to generate a set of three density fractions at a minimum generically representing clean coal, middlings and refuse. The specific gravities (SG) used during the float-sink tests, which are summarized in Table 3, were chosen to best reflect the separating density used by each plant. Normally, the float-sink work included partitioning at separating gravities of 1.5 SG and 1.8 SG. However, in some cases, additional separating gravities were included for specific plants to fully cover the range of separating gravities utilized by the facility. In one case (Plant 4), an entire range of float-sink gravities from 1.3 to 2.2 SG was utilized to span the full spectrum of organic and inorganic matter contained in this particular coal source. The data from the partitioning work generated a total of 771 individual samples for laboratory analyses.

All samples from the particle size-by-size float-sink analyses were analyzed for ash content via thermogravimetric analysis (ASTM D3174). After ashing, the elemental composition of REEs contained in each sample was determined by digesting the solid sample of ash residue in a mixture of aqua regia and hydrofluoric acid (ASTM D6357). The metal concentration in solution was measured using inductively coupled plasma optical emission spectroscopy (ICP-OES). The total REE content (TREE) was determined by summing the concentrations of the elements listed in Table 1. It should be noted that, for this particular study, the reported TREE concentrations include both Sc and Y. The total content of light REEs (LREE) was obtained by summing the concentrations of La, Ce, Pr, Nd, Sm, and Eu, while the total content of heavy REEs (HREE) was obtained by summing the concentrations of Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc and Y. For comparative purposes, the concentrations of REEs were often reported on both a whole-sample basis prior to ashing (i.e., REE mass per unit mass of total dry sample) as well as on an ash-residue basis after ashing (i.e., REE mass per unit mass of total dry ash residue).

RESULTS AND DISCUSSION

Data Overview

Table 4 shows the completed material balances for each of the 20 coal processing facilities examined in the current study. The cumulated data show that this fleet of coal preparation plants was fed approximately 101.9 million metric tonnes per year (Mt/yr) of dry feed solids containing over 49.5 Mt/yr of ash residue. These facilities produced approximately 44.9 Mt/yr of clean coal containing nearly 4.2 Mt/yr of ash. The plants also rejected 57.0 Mt/yr of waste as either coarse refuse (47.0 Mt/yr) or fine refuse (10.0 Mt/yr). As should be expected, the overwhelming majority (91.6%) of the ash residue originally contained in the feed stream reported to the coarse refuse (79.3%) and fine refuse (12.3%) streams, with the balance (8.4%) shipped off property to downstream consumers of the clean coal products. Table 4 also shows that the cumulative feed to the 20 plants contained 12,732 metric tonnes per year (t/yr) of TREEs. This tonnage value significantly exceeds the reported U.S. demand of 9,500 t/yr for these elements (USGS, 2019). This finding is encouraging given that the 20 plants evaluated in this study represent less than 8% of the 252 coal processing facilities currently listed as active and operational in the most recent census of coal preparation plants in the United States (Fiscor, 2018).

Figure 2 shows the tonnage distributions of ash and TREEs in the clean coal, coarse refuse and fine refuse streams for each of the 20 preparation plants. Figure 2(a) shows that the total tonnage of clean coal and refuse varies considerably from plant-to-plant. These fluctuations reflect the highly variable nature of the stratigraphic beds contained within the coal seams that comprise the eastern U.S. coalfields. In contrast, the data shown in Figures 2(b) and 2(c) demonstrate that the overwhelming majority of ash residue and REEs contained in these products reported to the coarse refuse streams from each plant. In fact, the combined coarse refuse streams from these 20 coal processing facilities contain sufficient REEs (9,230 t/yr) to nearly satisfy current domestic consumption (9,500 t/yr). Unfortunately, the data plotted in Figure 3 shows that the concentrations of REEs in the various products from the preparation plants are very low. Figure 3(a) shows the part-per-million REE concentrations on a whole-sample basis (ppm_w) for the clean coal, coarse refuse and fine refuse streams from each individual plant. Figure 3(b) presents the same information, but for part-per-million concentrations on an ash-residue basis (ppm_a). The data given in Figure 3(a) indicates that the concentrations of TREEs in the clean coal products ranged from a low of only 23 to a high of 84 ppm_w when reported on a whole-sample basis. For the coarse refuse, the REE concentrations were substantially higher and ranged from a low of 130 to a high of 266 ppm_w. Interestingly, the concentrations of TREEs in the fine refuse where found to be slightly lower, with a low value of 81 and a high value of 224 ppm_w. For comparison, Figure 3(b) shows the concentrations of TREEs present in the ash residue from the clean coal, coarse refuse and fine refuse streams. When reported on this ash-residue basis, the data indicated that the ash-residue in the clean coal products contained considerably higher residual concentrations of TREEs. The increase in mass concentration was expected since TREEs are not volatile and are concentrated during combustion of organic matter within the coal samples. The concentrations of TREEs in the clean coal products ranged from a low of 201 to a high of 767 ppm_a when reported on an ash-residue basis. The coarse and fine refuse products contained similar concentrations of TREEs for each coal site, and typically ranged from 162 to 359 ppm_a for the coarse refuse and 156 to 327 ppm_a for the fine refuse when reported on an ash-residue basis.

Ash Correlation

The aforementioned plant product data suggest a strong correlation between ash content and TREE concentration. To further examine this association, the concentrations of TREEs was plotted versus dry ash content in Figure 4 for the entire set of 771 splits of samples generated by the particle size and density partitioning of the clean coal, coarse refuse and fine refuse samples collected in the current study. The part-per-million TREE concentrations are presented in Figure 4(a) on a whole-sample (ppm_w) basis and in Figure 4(b) on an ash-residue (ppm_a) basis. Although significant data scatter is apparent due to the widely different characteristics of the coal feedstocks supplied to the 20 plants, the data plotted in Figure 4(a) generally show that the total concentration of REEs increased steadily with sample ash content when reported on a whole-sample basis. This trend suggests the REEs present in these streams have a strong association with the inorganic mineral matter contained in the various size and density fractions generated from the plant products. Overall, the lowest whole-sample TREE concentration of 8.9 ppm_w was observed for a float 1.3 SG fraction of 3.1% ash clean coal, while the highest whole-sample TREE concentration of 322 ppm_w was observed for a 1.6x1.8 SG fraction of coarse plant middlings containing 68.1% ash.

A more interesting trend was observed when the TREE concentrations were plotted on an ash-residue basis. As shown in Figure 4(b), the plotted data indicate that the residual ash contained in the low-ash clean coal and medium-ash middling products contained substantially higher concentrations of TREEs. In particular, the TREE concentrations trended sharply upwards as the content of inorganic matter dropped below 10-20% ash. Concentrations of TREEs in this region of the plot were up to three times higher than those in the high-ash region of the plot. In fact, some of the cleanest products in the 3-6% ash region gave spikes in the ash-residue TREE concentrations that approached 1,000 ppm_a. This plot suggests that the concentrations of REEs in the mineral species dispersed within the organic fractions of the coal seams are more highly enriched in REEs than the mineral species contained within the refuse products. One explanation for the lower REE concentrations in the high-ash refuse products may be the increased dilution of these samples by out-of-seam rock. Extraneous crustal material, which is unavoidably recovered by mechanized mining, would be expected to contain relatively low concentrations of TREEs. Another possibility is that some of the REEs contained in these feedstocks may have a small, but still significant, association with organic matter. The small organic affinity would inadvertently create an increase in ash-residue TREE concentrations as the ash content approached zero.

Another interesting observation was made when comparing the ash versus TREE plots for each individual plant. For several of the sites, simple power relationships of the form:

$$\text{TREE } (\text{ppm}_w) = K[A]^m \qquad [1]$$

$$\text{TREE } (\text{ppm}_a) = K[A]^{m-1} \qquad [2]$$

were found to provide excellent predictions of the TREE concentrations (ppm_w or ppm_a) as a function of fractional ash content (A). In these equations, K and m represent best-fit constants from empirical regression. Since the ash contents in the equations are fractional values between 0 and 1, the constant K mathematically equates to the projected TREE concentration at 100% ash for each set of data. Likewise, the constant m represents how rapidly the TREE concentration deteriorates from 100% to 0% ash as the REE-bearing strata become diluted with low REE organic matter. It should be noted that simple linear fitting expressions, which have been proposed in the literature (Lin et al., 2017b; Huang et al., 2018), were unable to provide satisfactory fits between the ash contents and TREE concentrations for this particular study.

For ease of comparison, the fitted data are plotted in Figure 5 (whole-sample basis) and Figure 6 (ash-residue basis) for each of the 20 plant sites. The dashed lines in each plot represent the 90% prediction bands for the empirically fitted power equations. The fitting constants K and m are summarized in Table 5 for each of the 20 plant sites. The constants are listed in order of highest to lowest correlation coefficient (R^2), which indicates the goodness-of-fit of Eqs. [1] for each set of plant data. As shown, the K-values for this particular study ranged from a low of 175 to a high of 307 ppm_w, while the m-values ranged from a low of 0.54 to a high of 0.90. Most of the empirical fits (15 of 20) were "excellent" or "good" with R^2 values in excess of 0.8. In particular, Eqs. [1] and [2] fit the TREE versus fractional ash data for plant sites 3, 11 and 13-16 extremely well with $R^2>0.94$. On the other hand, the empirical expressions did not fit as well for several of the data sets (i.e., plants 1, 6, 7, 10 and 17) due to significant variations in TREE concentrations as the ash contents increased. The high degree of scatter suggests that the stratigraphic layers of REE-bearing rock in the coal seams fed to these particular plants was more highly variable than the coal seams fed to some of the other preparation plants examined in this study. It should also be noted that some of the middlings fractions containing 40-50% ash appear to contain elevated contents of REEs. Similar observations have been reported elsewhere in the literature (Honaker et al., 2017).

Distribution of Heavy and Light REEs

REEs are often broken down into two separate groups, heavy and light. Heavy REEs are generally rarer and of higher economic value than light REEs. The heavy REEs (HREEs) considered in this study included Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y. The elements of La, Ce, Pr, Nd, Sm, Eu, and Gd were considered as light REEs (LREEs). Figures 7 and 8 show the concentrations of heavy and light REEs found in the various size and density fractions from the products of the 20 coal preparation plants evaluated in this study. The data shows that the concentrations of heavy REEs (HREEs) are approximately five times lower than those observed for light REEs (LREEs). Also, when comparing Figures 7(b) and 8(b), the plots show that the HREEs are more pronounced in the low-ash fractions when reported on an ash-residue basis. This phenomenon is best illustrated in Figure 9, which shows the ratio of heavy-to-light REEs as a function of ash content. The data indicates that for ash contents greater than 50%, the HREE/LREE ratio remains relatively constant at about 0.2. However, as the ash level drops to below about 20% ash, the HREE/LREE ratio rises sharply. HREE/LREE ratios in excess of 0.4 were observed for several of the sites that produced single-digit ash values. The spike in the low-ash region suggests that HREEs preferentially associate with species dispersed within the organic matter of the coal samples.

Distribution of Critical REEs

The U.S. Department of Energy (DOE) has identified five REEs that are particularly critical and likely to be subject to shortages between now and 2035 (Bauer et al., 2012; DOE, 2017). The elements include Nd, Dy, Eu, Y and Tb. In light of this concern, the data from the current study was replotted in Figure 10 to show only the total concentrations of these five critical elements (CREE) as a function of fractional ash content. The data set tends to mirror that of the HREEs in that CREEs increase with ash on a whole-sample basis (Figure 10(a)) and peak in the lower ash fractions of the clean coal products when reported on an ash-residue basis (Figure 10(b)). The increased relative quantities of critical elements in the clean coal fractions are illustrated in Figure 11, which shows the concentrations of the five CREEs relative to the total concentration (TREE) of REEs present in the various size and float-sink fractions. As shown, the CREE/TREE ratio falls between 0.25 and 0.30 for the vast majority of the data points with ash contents higher than 15-20%. Below this critical ash level, the CREE/TREE ratio climbs sharply up to approximately 0.40. As with the spike in HREEs, the data in the low-ash region suggests that CREEs preferentially associate with species in the organic matter of the coal samples. Therefore, while the data obtained in this study indicate that the highest tonnages of REEs are present in the high-ash refuse products, the heavy and critical REEs tend to be disproportionally concentrated in the clean coal fractions generated by the preparation plants. The later may be best recovered from the downstream fly ash generated by consumers, while the former may be most suitable for separation at the preparation facility prior to combustion.

Correlation with Lanthanum

Linear regression plots were constructed for all of the REE concentration data sets generated for each of the 20 coal preparation facilities. The plots were used to determine the linear correlation coefficients (R^2 values) that indicate whether meaningful statistical associations exist between the various REEs. R^2 values approaching 1 suggest a strong positive correlation between elements, while R^2 values near 0 indicate little or no correlation. The R^2 values are summarized in Table 6. Literature reports suggest that strong associations typically exist between La and other REEs (Ekmann, 2012). This suggests that La may be used as a trace indicator of REE enrichment for exploratory REE characterization work. As shown in Table 4, the data collected in the current study does indicate a strong correlation between La and Ce (R^2=0.95) and La and Nd (R^2=0.88). These elements are all light REEs. On the other hand, weak, but statistically significant ($0.49<R^2<0.61$), associations were noted between La and Y (R^2=0.56), Sm (R^2=0.59), Eu (R^2=0.53), Gd (R^2=0.60), Dy (R^2=0.56), Ho (R^2=0.61), Er (R^2=0.49) and Yb (R^2=0.50). Unfortunately, La did not correlate well with Sc (R^2=0.35) and Tm (R^2=0.32) and essentially no correlation existed between La and Pr (R^2=0.07), Tb (R^2=0.12) and Lu (R^2=0). These poorer correlations may be due to the lack of a geological/mineralogical association or problems associated with the assaying procedures used in the current study (e.g., instrument detection limits). One interesting finding from this study was the strong correlation between Y and elements such as Eu (R^2=0.86), Gd (R^2=0.86), Dy (R^2=0.95) and Yb (R^2=0.94). As such, this data set suggests that some of the heavy REEs (e.g., Dy and Yb) correlate better with Y than with La.

SUMMARY

The concentration of REEs were measured in the clean coal, middling and refuse streams from 20 different eastern U.S. coal preparation plants operating in Kentucky, Pennsylvania, Virginia and West Virginia. The products were separated into size and density classes in such a way as to closely represent the process circuity of each plant site. The products were then subjected to ash and REE analysis. The resultant database showed that the waste products from these 20 preparation plants contain sufficient REEs to satisfy the current annual domestic demand for this important class of strategic elements. A strong positive correlation was also noted between ash content and REE concentration. This relationship suggests that the primary association of REEs in eastern U.S. bituminous coals is dominated by the inorganic waste rock that is rejected by these preparation facilities. For many of the plants evaluated in this study, a simple power equation provided a good fit between ash and total REE concentration. The data also showed that, when reported on an ash-residue basis, the REE concentrations in the clean coal products were higher than those in the refuse streams, with values approaching 1,000 ppm_a (ash-basis) of total REEs. This unexpected finding suggests that the residual mineral matter remaining after the combustion of the clean coal products contain much higher concentrations of REEs than the mineral matter associated with the refuse streams. The low-ash fractions contained in the plant products were also found to contain larger ratios of heavy and critical REEs, which are scarcer and often more valuable. The concentration of heavy REEs was two-to-four times higher in the low-ash clean coal products than in the high-ash refuse products. This observation suggests that mineral impurities intimately associated with carbonaceous matter have elevated concentrations of heavy rare earth elements. A similar trend was observed for critical REEs (Y, Nd, Eu, Tb, and Dy) that are believed to be most vulnerable to supply shortages. The data from the study also showed that a linear correlation existed between La for many REEs of interest, although there were several notable exceptions for some high-value REEs (Lu, Pr and Tb).

CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Table 1. List of rare earth elements (plus scandium and yttrium).

	Chemical Element	Chemical Symbol	Atomic Number	Yttrium Group* (HREEs)	Cerium Group* (LREEs)	Crustal Abundance (ppm)**
	Scandium	Sc	21		x	22
	Yttrium	Y	39	x		33
Lanthanide Elements	Lanthanum	La	57		x	39
	Cerium	Ce	58		x	66.5
	Praseodymium	Pr	59		x	9.2
	Neodymium	Nd	60		x	41.5
	Promethium	Pm	61		x	--
	Samarium	Sm	62		x	7.1
	Europium	Eu	63		x	2.0
	Gadolinium	Gd	64	x		6.2
	Terbium	Tb	65	x		1.2
	Dysprosium	Dy	66	x		5.2
	Holmium	Ho	67	x		1.3
	Erbium	Er	68	x		3.5
	Thulium	Tm	69	x		0.52
	Ytterbium	Yb	70	x		3.2
	Lutetium	Lu	71	x		0.8

**Note: HREE = heavy REEs, LREE = light REEs (King, 2019).*

***From Lide, 2005. CRC Handbook of Physics and Chemistry, 85th Ed.*

Table 2. Classes utilized for particle size partitioning of samples collected from each coal preparation plant.

Plant ID	Particle Size Classes (mm)				
1	+31.75	131.75 +12.7	-12.7 +1	-1 +0.15	-0.15
2	--	+19.05	-19.05 +1	-1 +0.15	-0.15
3	+31.75	-31.75 +15.88	-15.88 +1	-1 +0.15	-0.15
4	+38.1	-38.1 +15.88	-15.88 +1	-1 +0.15	-0.15
5	+38.1	-38.1 +12.7	-12.7 +1	-1 +0.15	-0.15
6	+31.75	-31.75 +12.7	-12.7 +1	-1 +0.15	-0.15
7	--	+19.05	-19.05 +1	-1 +0.15	-0.15
8	--	+12.7	-12.7 +1	-1 +0.15	-0.15
9	+25.4	-25.4 +15.88	-15.88 +1	-1 +0.15	-0.15
10	+38.1	-38.1 +12.7	-12.7 +1	-1 +0.15	-0.15
11	+31.75	-31.75 +9.53	-9.53 +1	-1 +0.15	-0.15
12	+31.75	-31.75 +12.7	-12.7 +1	-1 +0.15	-0.15
13	+19.05	-19.05 +6.35	-6.35 +1	-1 +0.15	-0.15
14	--	+19.05	-19.05 +1	-1 +0.15	-0.15
15	+19.05	-19.05 +9.53	-9.53 +1	-1 +0.15	-0.15
16	+7.94	-7.94 +3.18	-3.18 +1	-1 +0.15	-0.15
17	+22.2	-22.2 +3.18	-3.18 +1	-1 +0.15	-0.15
18	+19.05	-19.05 +3.18	-3.18 +1	-1 +0.15	-0.15
19	+9.53	-9.53 +3.18	-3.18 +1	-1 +0.15	-0.15
20	+19.05	-19.05 +6.35	-6.35 +1	-1 +0.15	-0.15

Table 3. Classes utilized for density partitioning of samples collected from each coal preparation plant.

Plant ID	Float-Sink Specific Gravities (SGs)							
	1.3 SG	1.4 SG	1.5 SG	1.6 SG	1.7 SG	1.8 SG	1.9 SG	2.2 SG
1				x		x		
2				x		x		
3				x		x		
4	x	x	x	x	x	x	x	x
5			x	x		x		
6				x		x		
7				x		x		
8				x		x		
9			x	x		x		
10				x		x		
11				x			x	
12				x		x		
13				x		x		
14				x		x		
15				x		x		
16				x			x	
17				x		x		
18				x			x	
19			x	x			x	
20			x	x		x		

Table 4. Summary of tonnages and concentrations of total dry solids, ash and rare earth elements produced by the 20 coal preparation plants examined in the current study.

Stream	Total Mass		Ash		TREE		TREE (ppm$_w$)	TREE (ppm$_a$)
	Mt/yr*	%	Mt/yr*	%	t/yr	%		
Plant Feed	101.93	100.0	49.50	100.0	12,732	100.0	124.9	257.2
Clean Coal	44.92	44.1	4.17	8.4	1,995	15.7	44.4	478.4
Coarse Refuse	47.04	46.2	39.26	79.3	9,230	72.5	196.2	235.1
Fine Refuse	9.96	9.8	6.06	12.3	1,507	11.8	151.2	248.5

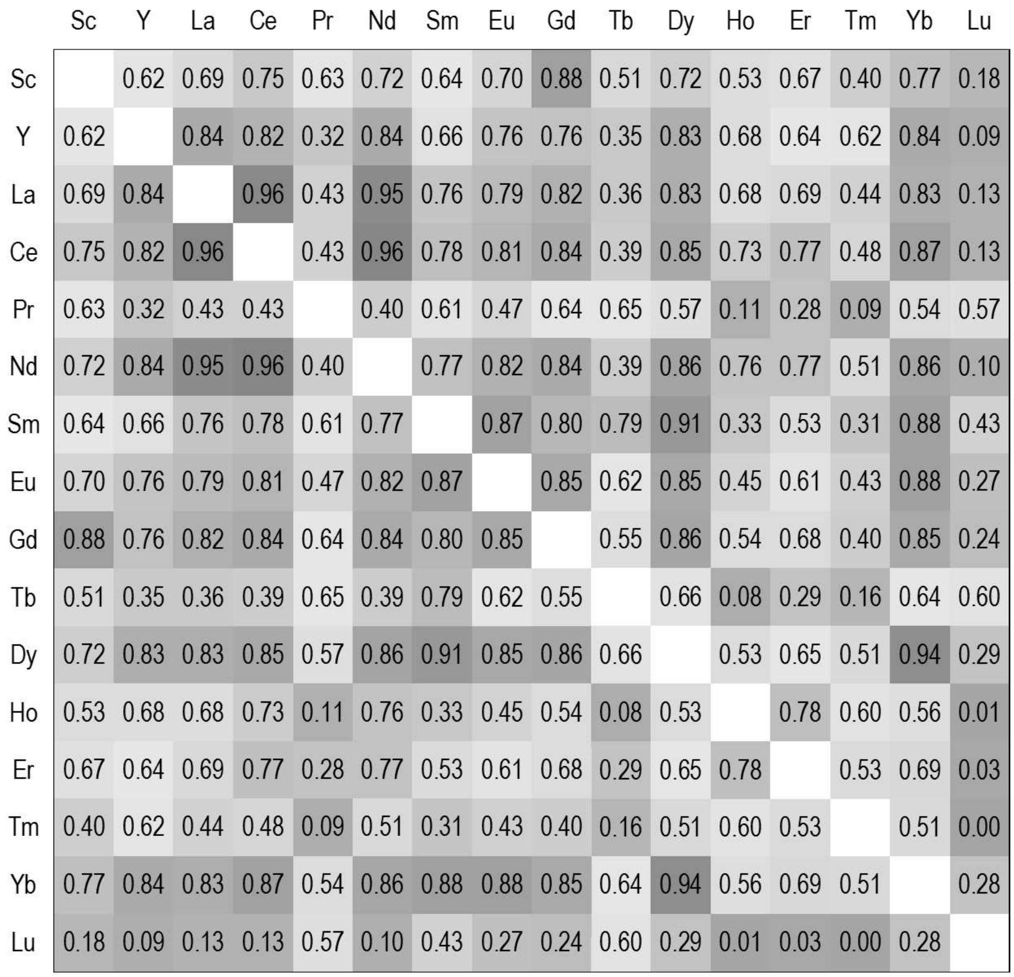
**Mt/yr = million metric tonnes per year*

Table 5. Correlation data for fractional ash content versus total rare earth concentration for size and density classes generated from the products collected at each coal preparation plant.

Plant ID	Constant “K”	Constant “m”	R^2	Fit?
11	270.0	0.66	0.98	Excellent
13	254.8	0.76	0.98	
15	275.2	0.90	0.96	
16	285.5	0.74	0.96	
3	235.2	0.70	0.94	
14	238.5	0.81	0.94	
12	306.6	0.87	0.90	Good
2	269.5	0.67	0.89	
4	242.7	0.73	0.88	
9	235.5	0.57	0.88	
5	227.1	0.75	0.87	
19	225.4	0.63	0.85	
20	175.2	0.87	0.84	
8	243.8	0.71	0.83	
18	227.6	0.77	0.81	
1	184.0	0.58	0.79	Poor
6	200.1	0.75	0.76	
10	178.6	0.65	0.73	
17	252.3	0.60	0.63	
7	228.0	0.54	0.62	

Maximum	306.6	0.90	0.98
Minimum	175.2	0.54	0.62
Mean	237.8	0.71	0.85
Median	237.0	0.72	0.88

Table 6. Linear coefficient of determination (R^2) values showing the associations between various REEs contained in the products from 20 coal preparation facilities.



	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Sc		0.62	0.69	0.75	0.63	0.72	0.64	0.70	0.88	0.51	0.72	0.53	0.67	0.40	0.77	0.18
Y	0.62		0.84	0.82	0.32	0.84	0.66	0.76	0.76	0.35	0.83	0.68	0.64	0.62	0.84	0.09
La	0.69	0.84		0.96	0.43	0.95	0.76	0.79	0.82	0.36	0.83	0.68	0.69	0.44	0.83	0.13
Ce	0.75	0.82	0.96		0.43	0.96	0.78	0.81	0.84	0.39	0.85	0.73	0.77	0.48	0.87	0.13
Pr	0.63	0.32	0.43	0.43		0.40	0.61	0.47	0.64	0.65	0.57	0.11	0.28	0.09	0.54	0.57
Nd	0.72	0.84	0.95	0.96	0.40		0.77	0.82	0.84	0.39	0.86	0.76	0.77	0.51	0.86	0.10
Sm	0.64	0.66	0.76	0.78	0.61	0.77		0.87	0.80	0.79	0.91	0.33	0.53	0.31	0.88	0.43
Eu	0.70	0.76	0.79	0.81	0.47	0.82	0.87		0.85	0.62	0.85	0.45	0.61	0.43	0.88	0.27
Gd	0.88	0.76	0.82	0.84	0.64	0.84	0.80	0.85		0.55	0.86	0.54	0.68	0.40	0.85	0.24
Tb	0.51	0.35	0.36	0.39	0.65	0.39	0.79	0.62	0.55		0.66	0.08	0.29	0.16	0.64	0.60
Dy	0.72	0.83	0.83	0.85	0.57	0.86	0.91	0.85	0.86	0.66		0.53	0.65	0.51	0.94	0.29
Ho	0.53	0.68	0.68	0.73	0.11	0.76	0.33	0.45	0.54	0.08	0.53		0.78	0.60	0.56	0.01
Er	0.67	0.64	0.69	0.77	0.28	0.77	0.53	0.61	0.68	0.29	0.65	0.78		0.53	0.69	0.03
Tm	0.40	0.62	0.44	0.48	0.09	0.51	0.31	0.43	0.40	0.16	0.51	0.60	0.53		0.51	0.00
Yb	0.77	0.84	0.83	0.87	0.54	0.86	0.88	0.88	0.85	0.64	0.94	0.56	0.69	0.51		0.28
Lu	0.18	0.09	0.13	0.13	0.57	0.10	0.43	0.27	0.24	0.60	0.29	0.01	0.03	0.00	0.28	

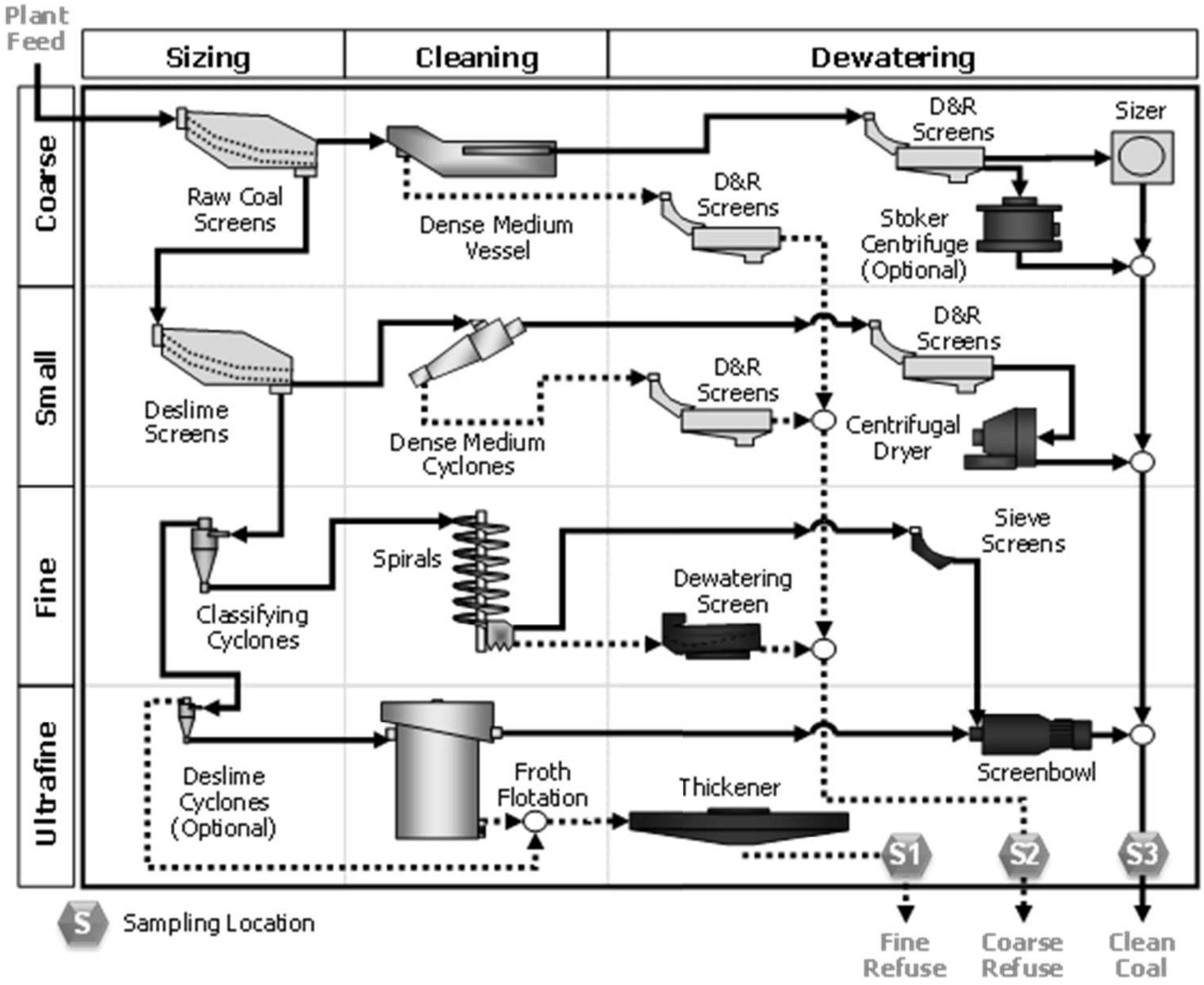


Figure 1. Generic flowsheet showing locations of plant sampling points.

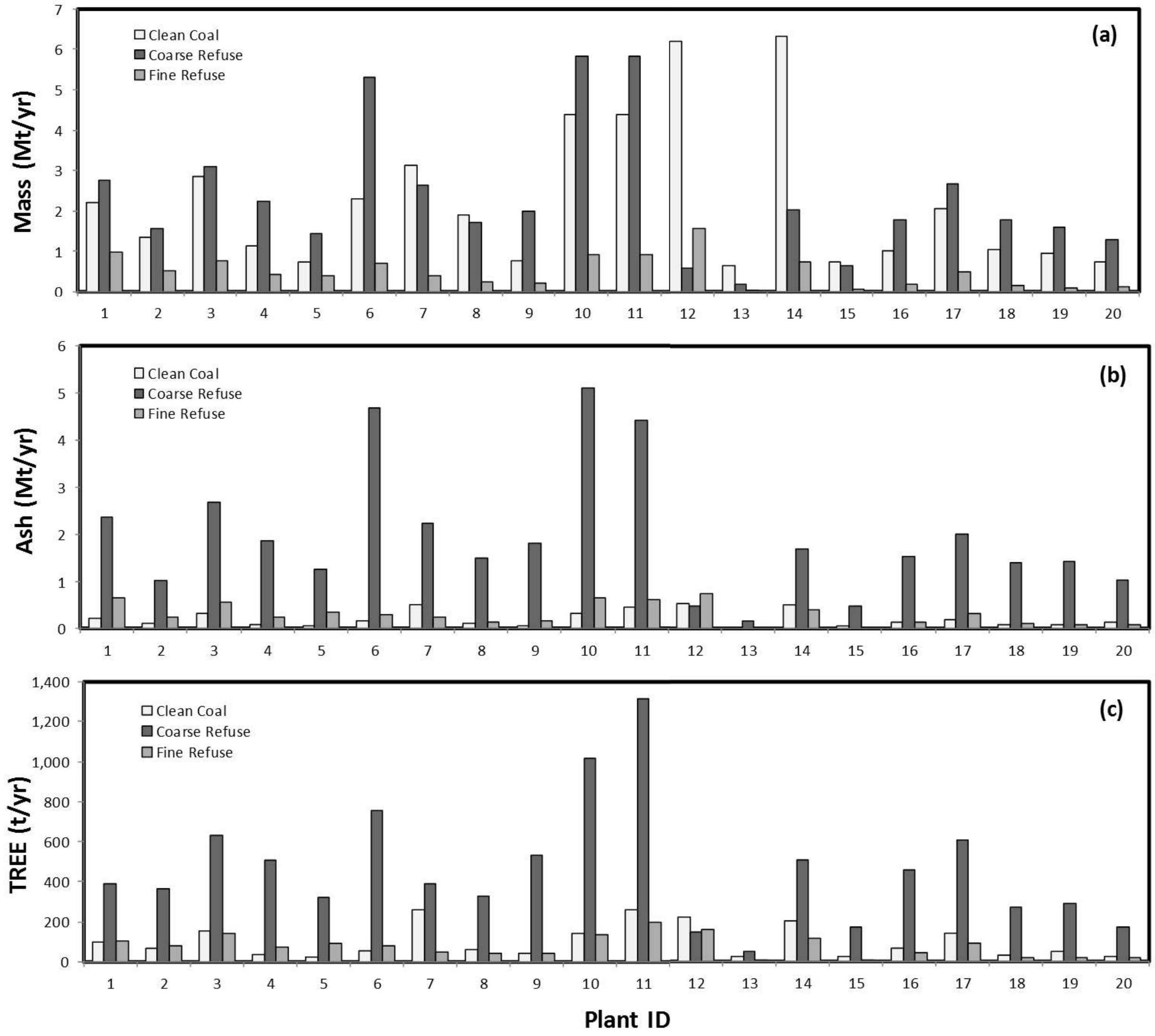


Figure 2. Annual tonnages of total mass, ash and rare earth elements contained in the clean coal, coarse refuse and fine refuse streams generated by 20 eastern U.S. coal preparation plants.

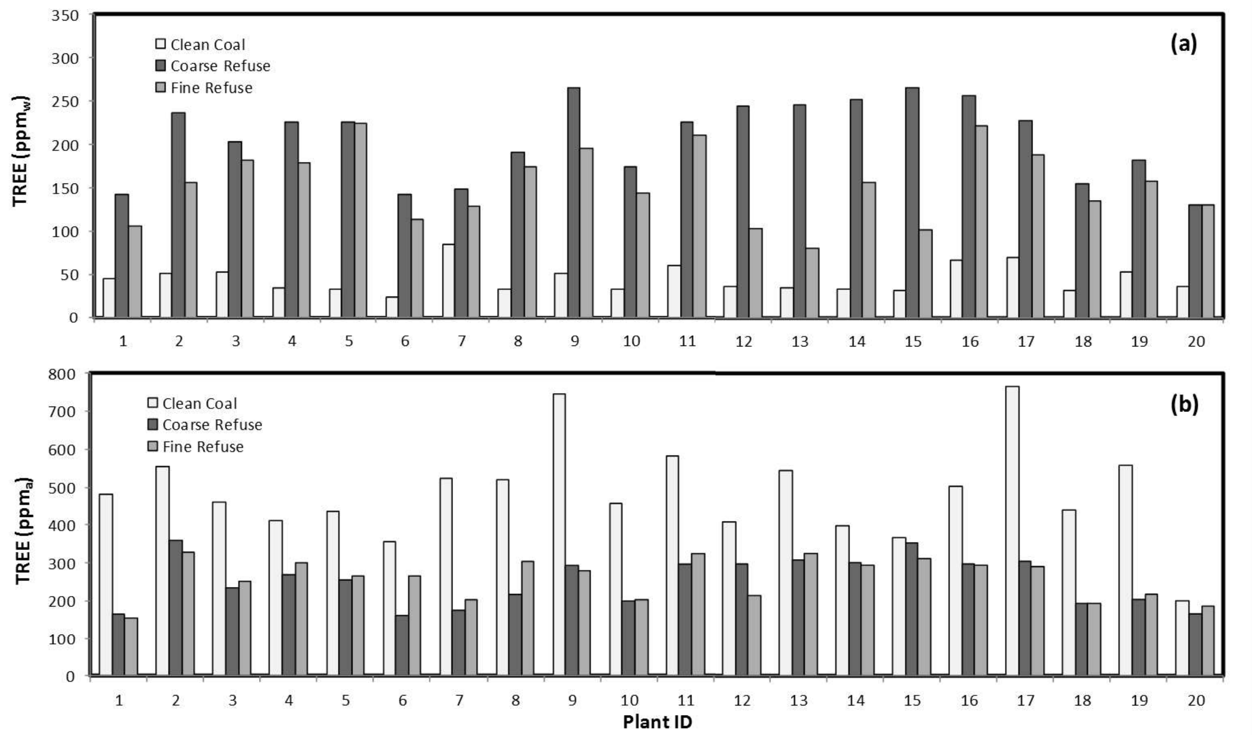


Figure 3. **Concentrations of rare earth elements contained in the clean coal, coarse refuse and fine refuse streams generated by 20 eastern U.S. coal preparation plants. Values reported on (a) a whole-sample basis and (b) an ash-residue basis.**

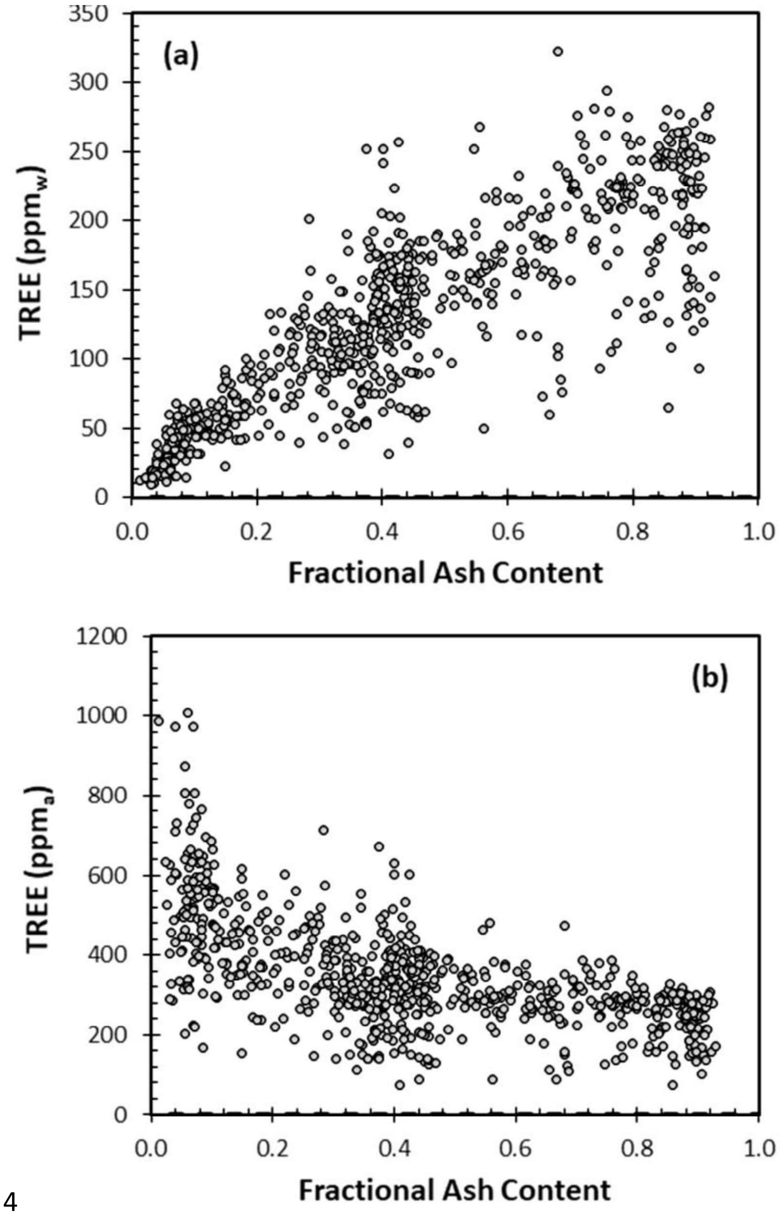


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Figure 4. Total concentration of rare earth elements (TREE) versus fractional dry ash content for various particle size and density fractions within the products generated by 20 eastern U.S. coal preparation plants. Values reported on (a) a whole-sample basis and (b) an ash-residue basis.

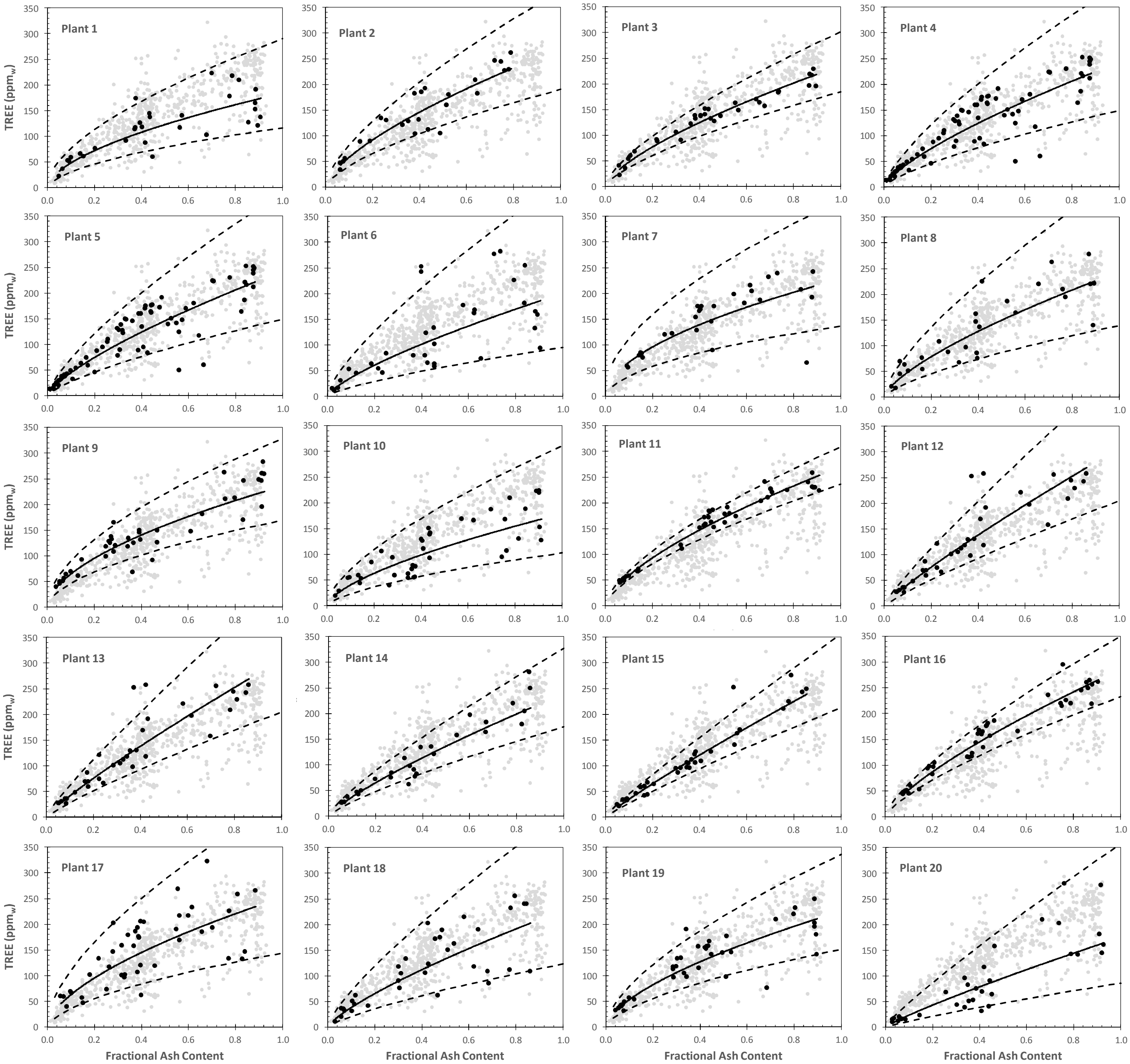

Figure 5. Regression analyses relating total rare earth element (TREE) concentration on a whole-sample basis versus fractional ash content for partitioned samples from each of the 20 coal preparation facilities. Solid curve represents a power equation fit and dashed lines represent 90% prediction bands.

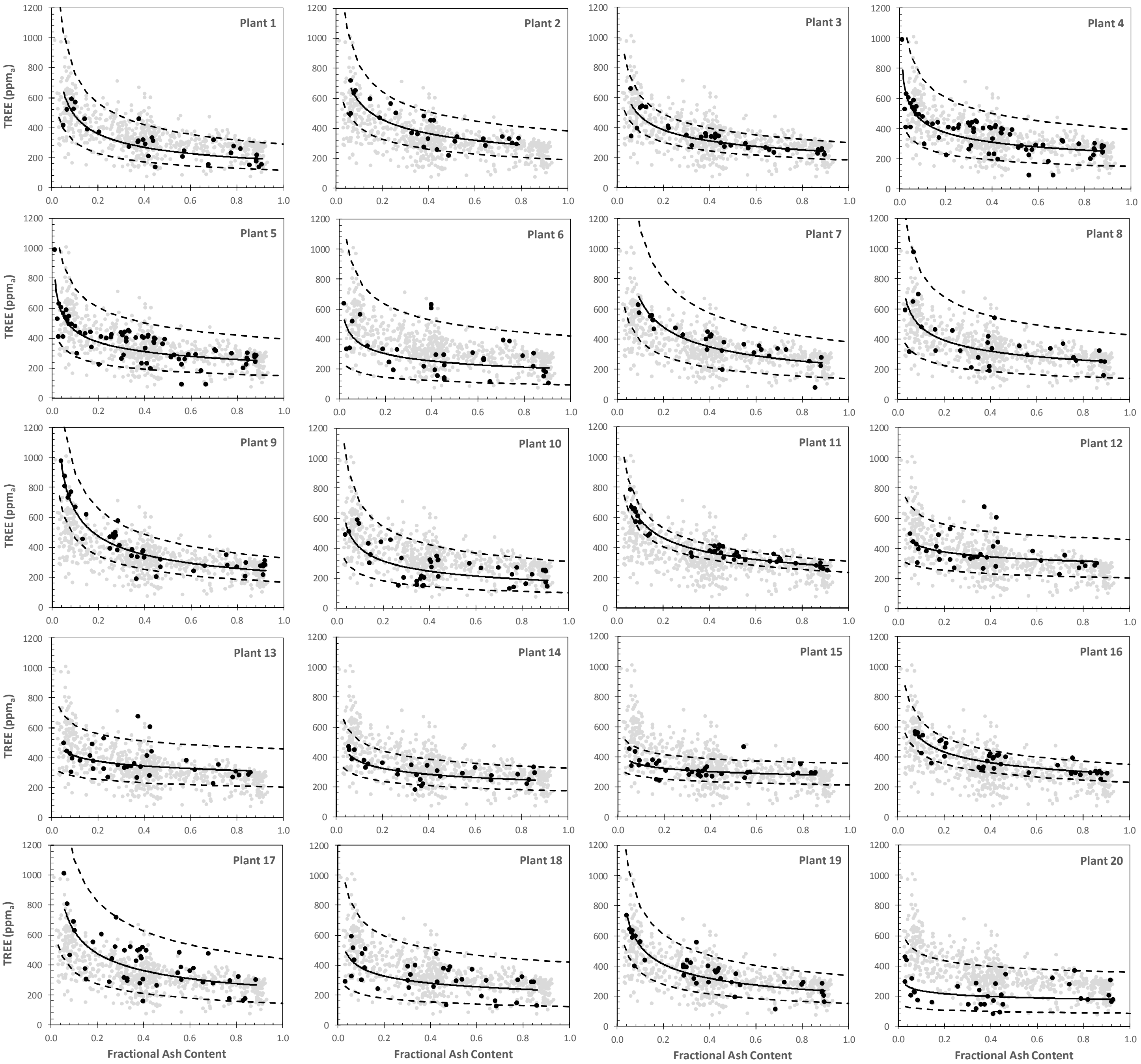

Figure 6. **Regression analyses relating total rare earth element (TREE) concentration on an ash-residue basis versus fractional ash content for partitioned samples from each of the 20 coal preparation facilities. Solid curve represents a power equation fit and dashed lines represent 90% prediction bands.**

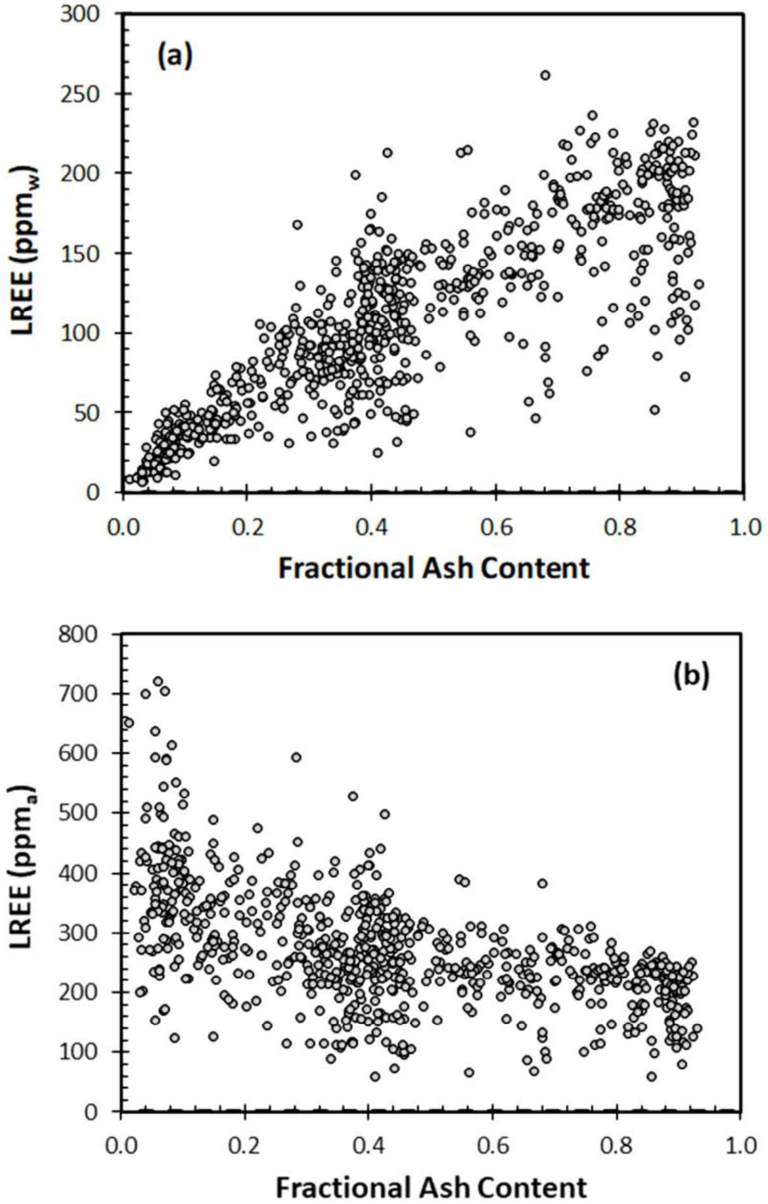

Figure 7. Concentration of light rare earth elements (LREE) versus fractional dry ash content for various particle size and density fractions within the products generated by 20 eastern U.S. coal preparation plants. Values reported on (a) a whole-sample basis and (b) an ash-residue basis.

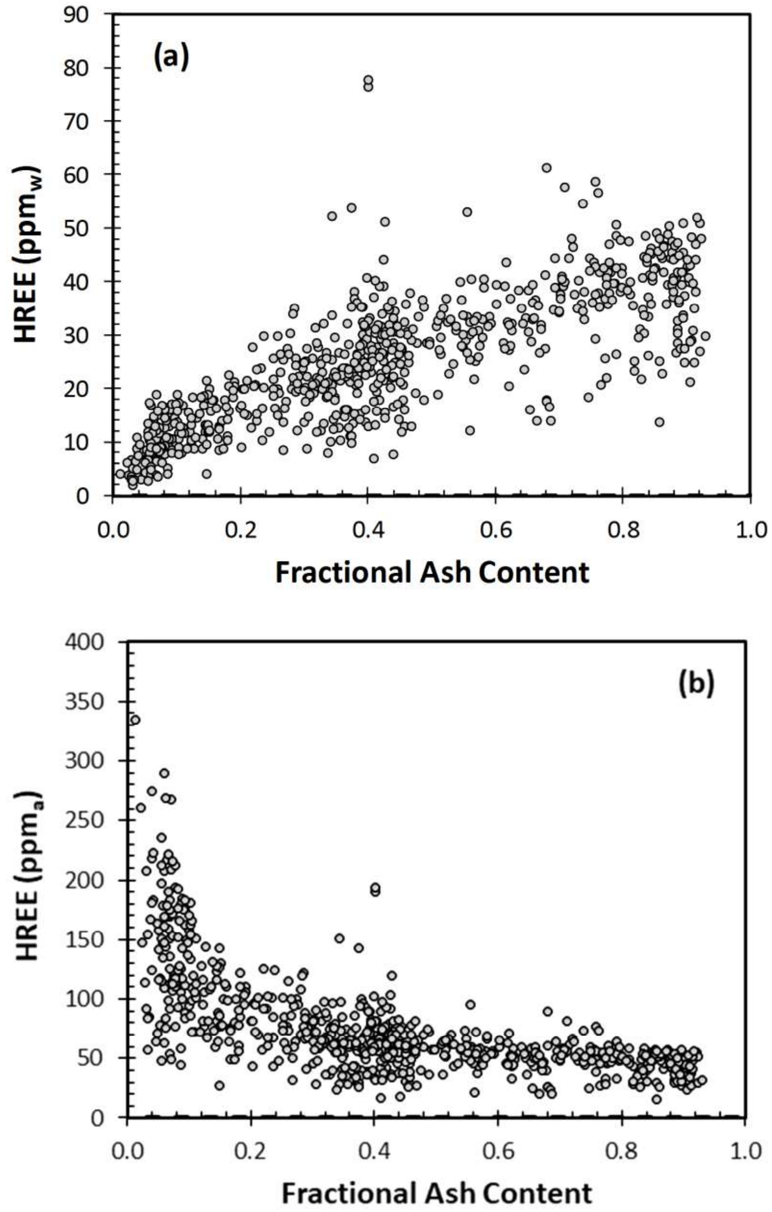

Figure 8. **Concentration of heavy rare earth elements (HREE) versus fractional dry ash content for various particle size and density fractions within the products generated by 20 eastern U.S. coal preparation plants. Values reported on (a) a whole-sample basis and (b) an ash-residue basis.**

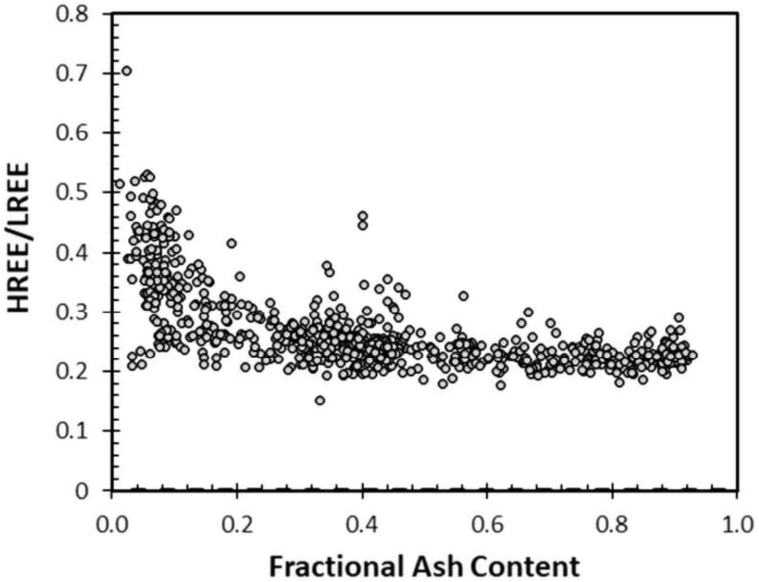

Figure 9. Ratio of heavy-to-light rare earth elements (HREE/LREE) versus fractional dry ash content for various particle size and density fractions within the entire set of plant products examined in this study.

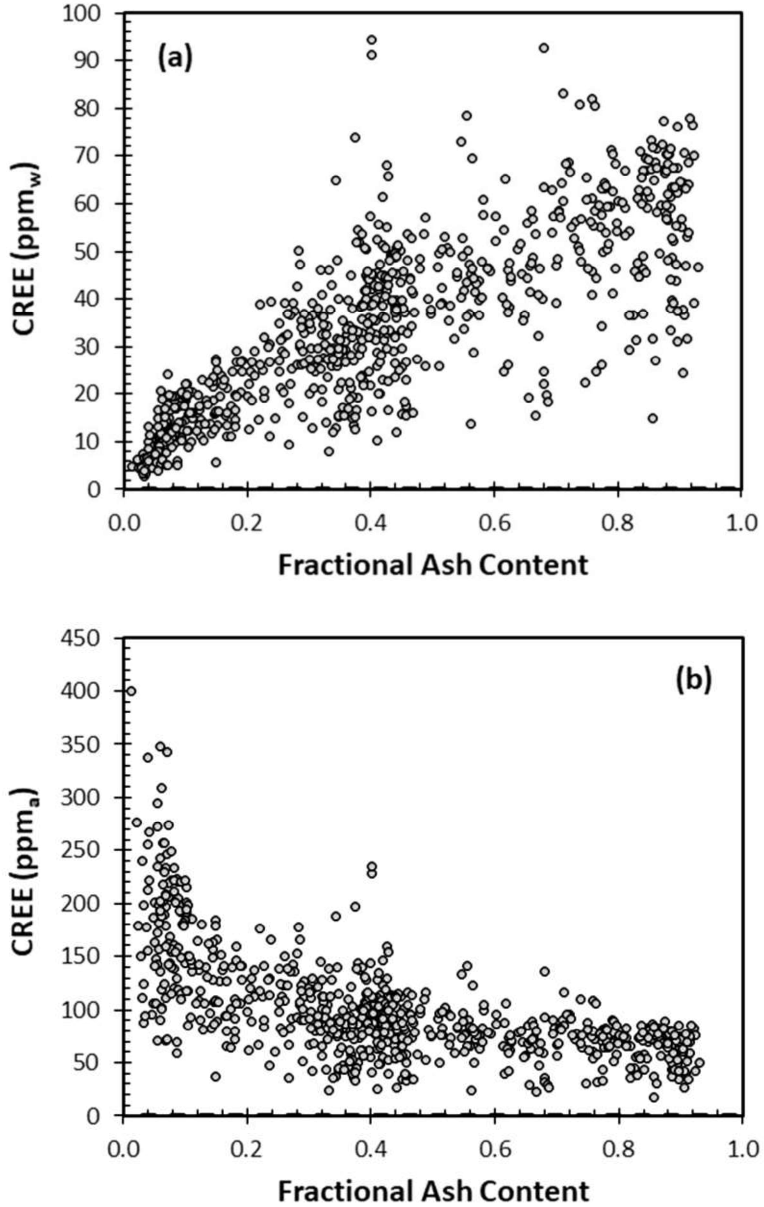

Figure 10. Concentration of critical rare earth elements (CREE) versus fractional dry ash content for various particle size and density fractions within the products generated by 20 eastern U.S. coal preparation plants. Values reported on (a) a whole-sample basis and (b) an ash-residue basis.

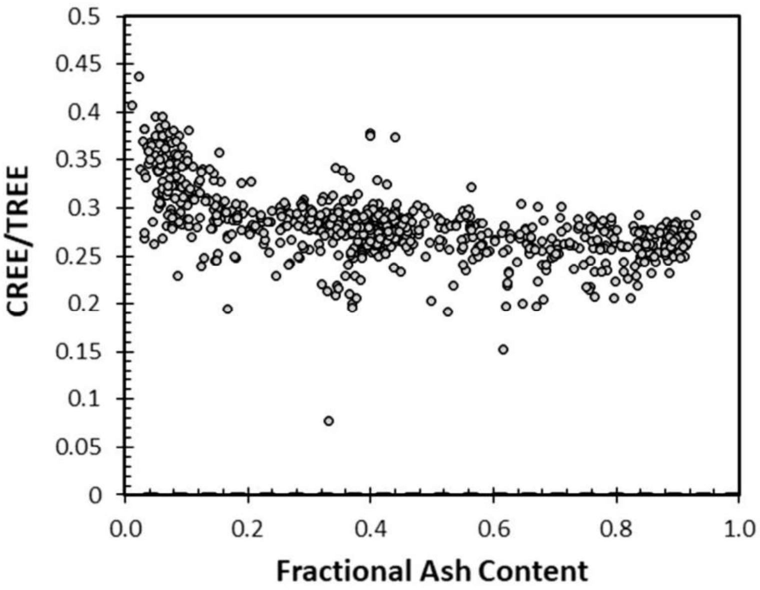

Figure 11. Ratio of critical-to-total rare earth elements (CREE/TREE) versus fractional dry ash content for various particle size and density fractions within the entire set of plant products examined in this study.