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## Thermogravimetric analysis of respirable coal mine dust for simple source apportionment

Lizeth Jaramillo, Eleftheria Agioutanti, Setareh Ghaychi Afrouz, Cigdem Keles, and Emily Sarver

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### ABSTRACT

Resurgence of coal mine dust lung diseases in the central Appalachian region of the United States and elsewhere has spurred a range of efforts to better understand respirable coal mine dust (RCMD) exposures and sources. Thermogravimetric analysis (TGA) of RCMD samples can enable the dust mass to be fractionated into three main components: coal, non-carbonate minerals, and carbonates. These are expected to approximate, respectively, the three primary dust sources in many underground mines: the coal seam being mined, the surrounding rock strata (i.e., typically dominated by non-carbonate minerals) being drilled or mined along with the coal, and the rock dust products (i.e., typically made from carbonate-rich limestone or dolostone) being applied in the mine to mitigate explosibility hazards. As proof of concept, TGA was applied to respirable dust samples that were laboratory-generated from real source materials representing 15 mines. Except in the case of two mines, compositional results were generally consistent with expectations. TGA was also applied to RCMD samples collected in standard locations of 23 mines (including the 15 mines represented by the dust source materials). Results showed significantly different compositions with respect to sampling location and geographic region (i.e., within and outside of central Appalachia). To further interpret the RCMD results, a simple source apportionment model was built using the dust compositions yielded from the source materials analysis. Model results indicated that, on average, about twice as much dust was sourced from mining into rock strata than from mining the target coal seam. This finding is particularly important for mines extracting relatively large amounts of rock along with the coal or for mines that frequently encounter high-silica rock strata.



### KEYWORDS


Coal worker's pneumoconiosis; occupational health; progressive massive fibrosis; respirable dust

### Introduction

Respirable coal mine dust (RCMD) still represents a serious occupational health concern in the United States (U.S.) and elsewhere (Graber et al. 2017; McCall 2017; Blackley et al. 2018; Han et al. 2018; Hall et al. 2019a). The resurgence of severe and rapidly progressive disease, especially in the central Appalachian region, has spurred a range of efforts to better understand and monitor RCMD exposures in U.S. mines. While studies of lung radiographs and pathology have implicated excessive exposures to crystalline silica (Blackley et al. 2016; Hall et al. 2019b) and perhaps other silicates (e.g., Cohen et al. 2016; Jelic et al. 2017), the available mine monitoring data does not reveal a clear picture of when and under what conditions such exposures occurred (Doney et al. 2019; Agioutanti et al. 2020).

To gain further insight about RCMD exposures and better protect miners moving forward, a recent consensus report published by the National Academies of Science, Engineering and Medicine (NASEM 2018) offered numerous recommendations—one of which was to “characterize and quantify important source contributions to airborne RCMD, including rock dusting and extraction of rock strata adjacent to the mined coal seam” (NASEM 2018). Indeed, there are commonly three sources of dust in many coal mines (Sarver et al. 2019). Dust, including respirable size particles, is generated from (1) the coal strata itself during mining. It can also be generated from (2) the surrounding rock strata, which is typically drilled for installation of roof bolts and may also be mined along with the target coal. Incidentally, central Appalachia is known for “thin-seam” mining, in

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which relatively large amounts of rock—frequently shale, slate or sandstone, dominated by silicates and silica—are extracted with the coal to maintain adequate opening heights for equipment, ventilation, and personnel (Pollock et al. 2010, Schatzel 2009). Another common source of dust is (3) “rock dusting”, which refers to the application of fine, inert material—often high-purity limestone or dolostone dust—to coal mine surfaces in order to mitigate explosibility hazards (Luo et al. 2017). Numerous studies have demonstrated this practice can contribute to the RCMD concentration (LaBranche et al. 2021; Pokhrel et al. 2021; Sarver et al. 2021). It is noted that, in mines operating diesel equipment, engine emissions can also contribute significant particle numbers to the RCMD (Sarver et al. 2019), though due to the very fine size of diesel particulates they are generally not expected to contribute much to the RCMD mass (Birch and Noll 2004; Sarver et al. 2021).

Considering the above, an analytical method that effectively splits RCMD into coal, carbonates, and non-carbonate minerals mass fractions might enable simple dust source apportionment in mines where the aforementioned sources and typical source compositions hold true. To this end, the authors of the current work previously devised a thermogravimetric application for analysis of RCMD (Agioutanti et al. 2020). Thermogravimetric analysis (TGA) essentially seeks to correlate changes in sample weight within defined temperature ranges (in a controlled atmosphere) to specific sample components. For RCMD, Agioutanti et al. (2020) showed that: the coal component should oxidize nearly completely at relatively low temperature (<480 °C); the carbonate component (using calcium carbonate, CaCO<sub>3</sub>, as a proxy for high purity limestone) should decompose at higher temperature (480–800 °C) to CaO and CO<sub>2</sub>; and the non-carbonate component (using low-carbonate shale rock as a proxy) should be thermally stable across the entire temperature range of the TGA experiment (ambient–800 °C). Thus, after tracking the sample weight loss with temperature, a series of mass balance equations can be used to fractionate the dust into the three components of interest (i.e., coal, carbonates, and non-carbonate minerals). Agioutanti et al. (2020) developed the TGA application using respirable dust samples which were generated in the laboratory from a few well-characterized source materials (i.e., real coal and shale rock, and a limestone rock dust product).

Of course, it is important to acknowledge that RCMD sources are not pure, and variability in composition could affect the thermogravimetric behavior

of dust—and therefore interpretation of TGA results. As a more robust proof-of-concept for the TGA application devised by Agioutanti et al. (2020), it was applied in the current study to respirable dust samples generated from source materials representing 15 underground coal mines in the U.S. It was also used to analyze RCMD samples collected in standardized locations of these 15 mines, plus eight more U.S. mines (i.e., 23 in total). The RCMD results were compared on the basis of sampling location and geographic region (i.e., within or outside of central Appalachia), and a simple source apportionment model was used to interpret the major sources of the respirable dust.

## Materials and methods

### Study mines

RCMD sampling was conducted in 23 underground coal mines between 2014–2020. Fifteen of the mines are located in central Appalachia (US Mine Safety and Health Administration [MSHA] districts 4, 5, 12), four are in northern Appalachia (MSHA districts 2, 3), two are in the mid-west (MSHA district 8), and two are in the west (MSHA district 9). Table 1 shows key details for each study mine, including the primary mining method, coal seam thickness and total mining height (i.e., the difference is the height of rock strata being mined along with the coal), and the primary rock strata type being mined during the RCMD sampling.

### Mine dust sampling

Across all 23 mines, a total of 129 RCMD samples were collected in the following standardized locations: in the intake (fresh) airway of the sampled mining section; in the return (exhaust) airway of the sampled section; just downwind of active coal production; adjacent to the feeder breaker, which crushes the raw coal and rock material before is loaded onto the haulage conveyor belt; and just downwind of an active roof bolting machine. Table 2 shows the number of samples collected in each location per mine. An effort was made to sample all locations in each mine, however this was not always possible due particular mine operations during sampling or limited time underground. Moreover, in some mines, multiple samples were collected in some of the locations during different sampling events (i.e., on different shifts or days).

All RCMD samples were collected by the authors' research team using standard sampling equipment

**Table 1.** Summary of study mines.

Mine	MSHA District	Region <sup>1</sup>	Mining method <sup>2</sup>	Coal thickness (m)	Total mining height (m)	Primary rock strata
1	4	CA	CM	0.9–1.5	1.5	Sandstone
2	4	CA	CM	0.9–1.2	1.7	Sandstone
3	4	CA	CM	1.4	1.8	Shale/sandstone
4	4	CA	CM	0.6–1.2	1.2	Sandstone
5	2	NA	LW	1.8–2.4	2.4	Sandy shale/slate
7	12	CA	CM	1.5–1.8	1.8–2.1	Sandy shale/slate
8	12	CA	CM	1.2–1.5	1.8	Shale
10	5	CA	CM	1–1.2	1.8–1.9	Shale/sandstone
11	5	CA	CM	0.8	1.2–1.3	Shale
12	5	CA	CM	0.8–1.5	1.9–2	Shale/sandstone
13	5	CA	LW	1.5–1.8	1.8–2	Sandstone/shale
14	5	CA	CM	0.6–0.9	1.5–1.9	Shale
15	4	CA	CM	0.9–1.2	2	Shale
16	3	NA	CM	2.7	2.1	Shale
17	3	NA	LW	1.5–1.8	2–2.3	Shale
18	3	NA	CM	0.7–0.8	1.5–1.8	Shale
19	8	MW	CM	1.8–2	1.9–2.3	Shale/limestone
20	8	MW	CM	1.8–1.9	1.8–2	Shale/limestone
21	12	CA	CM	0.8–1	2	Shale/sandstone
22	12	CA	CM	0.8	1.4	Shale
23	9	W	LW	4.3	4.3	Shale
24	9	W	LW	1.8	2.1–2.4	Shale/sandstone
25	12	CA	CM	0.9	2.4	Shale/sandstone

<sup>1</sup>CA: central Appalachia, NA: northern Appalachia, MW: mid-west, W: west

<sup>2</sup>LW: longwall, CM: continuous miner

**Table 2.** Number and location of respirable coal mine dust (RCMD) samples and primary dust source materials included in this study. RCMD samples include intake, return, production, bolter, and feeder samples while dust source materials include rock dust, run-of mine (ROM) coal and rock, and bolter dust samples.

Mine	RCMD samples					Dust source materials				Total	
	Sampling locations					Material					
	Intake	Return	Production	Bolter	Feeder	Rock dust	ROM coal	Rock strata			
								ROM rock	Bolter dust		
1	1	1	1	1	1	5	Source materials not collected				
2	1	1	1	1	1	5					
3	1	1	1	1	1	5					
4	1	1	–	1	1	4					
5	2	2	1	1	2	8					
7	1	1	1	1	1	5					
8	1	2	2	1	1	7					
10	1	1	1	2	2	7	–	–	1	1	2
11	–	1	1	1	1	4	1	–	1	–	2
12	1	1	–	1	1	4	1	1	1	–	3
13	2	2	–	1	1	6	1	1	–	1	3
14	1	–	1	1	1	4	1	1	1	1	4
15	1	1	1	1	1	5	1	1	1	–	3
16	–	1	1	1	1	4	1	1	–	1	3
17	2	2	1	1	–	6	1	1	–	1	3
18	1	1	1	1	1	5	1	1	1	1	4
19	1	2	1	1	1	6	1	1	1	1	4
20	1	1	1	1	2	6	1	–	–	1	2
21	1	1	1	1	1	5	–	1	1	1	3
22	1	–	1	–	1	3	1	1	1	1	4
23	3	1	–	–	–	4	1	1	1	1	4
24	3	2	–	–	1	6	1	–	–	1	2
25	1	4	8	1	1	15	Source materials not collected				
<b>Total</b>	<b>28</b>	<b>29</b>	<b>27</b>	<b>21</b>	<b>24</b>	<b>129</b>	<b>13</b>	<b>11</b>	<b>10</b>	<b>12</b>	<b>46</b>

and materials obtained from Zefon International (Ocala, FL). Each sampling train included an Escort ELF air pump operated at a flow rate of 2L/min, which was paired with a 10 mm nylon cyclone (i.e., identical to those used for Coal Mine Dust Personal

Sampler Units, CMDPSU). The cyclone should discard virtually all particles larger than 10 µm and yield a d50 of about 3.5 µm at the stated flow rate. This is consistent with the regulatory definition of respirable dust for U.S. coal mines (NASEM 2018). RCMD was

collected directly onto polycarbonate (PC) filters (37-mm, track-etched with nominal 0.4- $\mu\text{m}$  pore size), which were housed in two-piece styrene cassettes. Samples were typically collected over periods of about 2–4 hr.

### **Primary source materials and lab-generated dust samples**

During the RCMD sampling visits at 15 of the mines (Mines 10–24), 46 bulk samples of primary dust source materials were also collected (Table 2). These included: bulk “run-of-mine” (ROM) material, which was pulled from the production belt and later hand separated into coal and rock materials; materials taken directly from the dust collection system of roof bolter machines; and grab samples of rock dust products, taken from the duster machine storage bin or new product sacks. While these materials should approximately represent the coal strata, rock strata from the ROM rock or roof bolter dust, and rock dust product sources contributing to the RCMD during mine sampling, they were not expected to be pure. For example, since the delineation of coal and rock strata is imperfect, the coal materials were expected to contain some non-carbonate minerals and the rock strata materials were expected to contain some coal. Moreover, the rock dust products were expected to contain small fractions of non-carbonate minerals due to impurities in the products themselves.

From each of the 46 source materials, samples of respirable dust were generated in the laboratory. The bolter dust and rock dust materials did not require pre-processing since they were already powdered. However, the ROM coal and rock materials were pulverized and sieved to minus 230 mesh ( $<63\ \mu\text{m}$ ) in the laboratory prior dust sample generation. For each material, a small mass of the bulk powder was loaded into a sealed enclosure and was made airborne by applying compressed air pulses during the dust sample collection time. The aforementioned sampling equipment (pump and cyclone) was used to collect the respirable-sized dust onto the same PC filters used for the RCMD sampling. Sample weights were generally between 800–1500  $\mu\text{g}$ . Sample mass was determined by weighing the filter before and after dust loading using a microbalance (Sartorius MSE6.6S, Gottingen, Germany).

### **Dust sample analysis**

All 129 RCMD samples and 46 lab-generated samples of source materials were analyzed using the

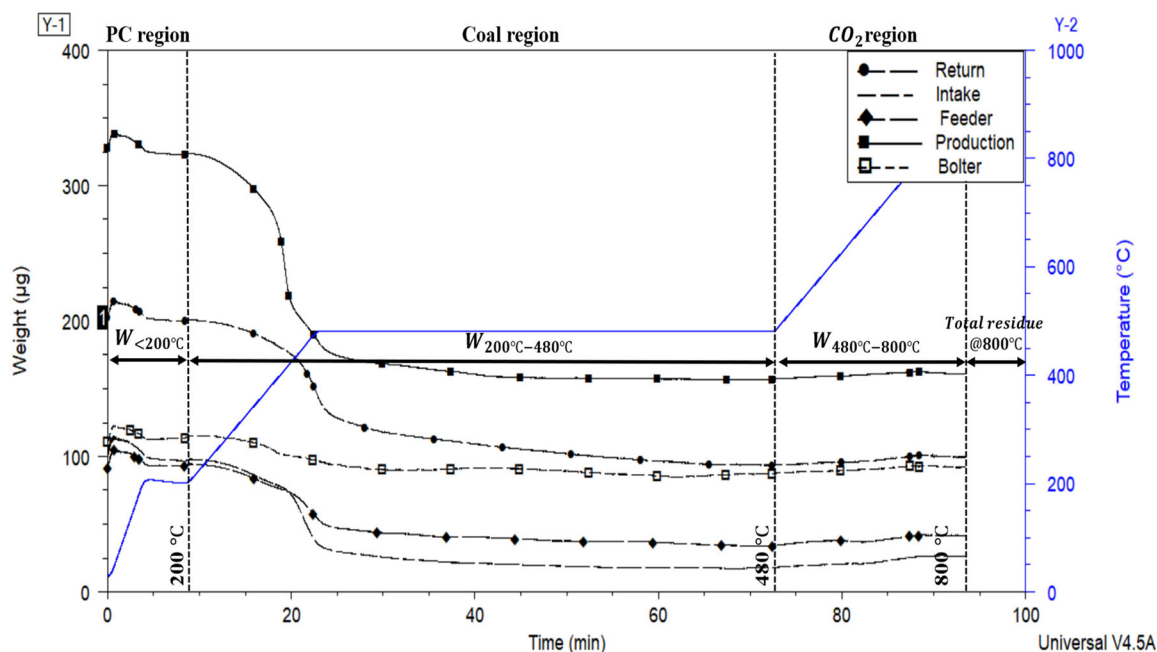
thermogravimetric application described by Agioutanti et al. (2020). Briefly, each sample filter was placed inside a clean glass tube and submerged in 5–10 mL of isopropyl alcohol (IPA). The tube was placed in an ultrasonic bath at 30 °C for 3 min. Next, the filter was carefully removed and the tube was centrifuged to settle the dust. Then, the IPA was allowed to evaporate substantially before the remaining contents of the tube (dust suspension in IPA) was pipetted into a clean, tared TGA pan (pure platinum). After the IPA had completely evaporated, the pan was loaded onto the same TGA instrument (Q500 TGA; TA Instruments, New Castle, DE) used by Agioutanti et al. (2020).

TGA samples were generally processed in sets of six, with empty pans included at the beginning and end of each set (i.e., eight pans per run, with dust samples in pans 2–7). As described by Agioutanti et al. (2020), the empty pans (1 and 8) are used for QA/QC (i.e., to check and correct for thermal drift). For each sample, the TGA output was a thermogram; representative thermograms for several RCMD and lab-generated samples from the source materials are shown in Figures 1 and 2, respectively. Thermal Advantage software (TA Instruments, New Castle, DE) was used to determine the sample weight loss in each specific temperature region (i.e., ambient–200 °C, 200–480 °C, 480–800 °C) and the final weight of the sample residue at 800 °C. Then, the coal, carbonates, and non-carbonate minerals mass percentages were estimated using the mass balance equations derived by Agioutanti et al. (2020), which also account for sample weight loss due to any interference associated with the dust recovery procedure (e.g., residue from the PC filter itself).

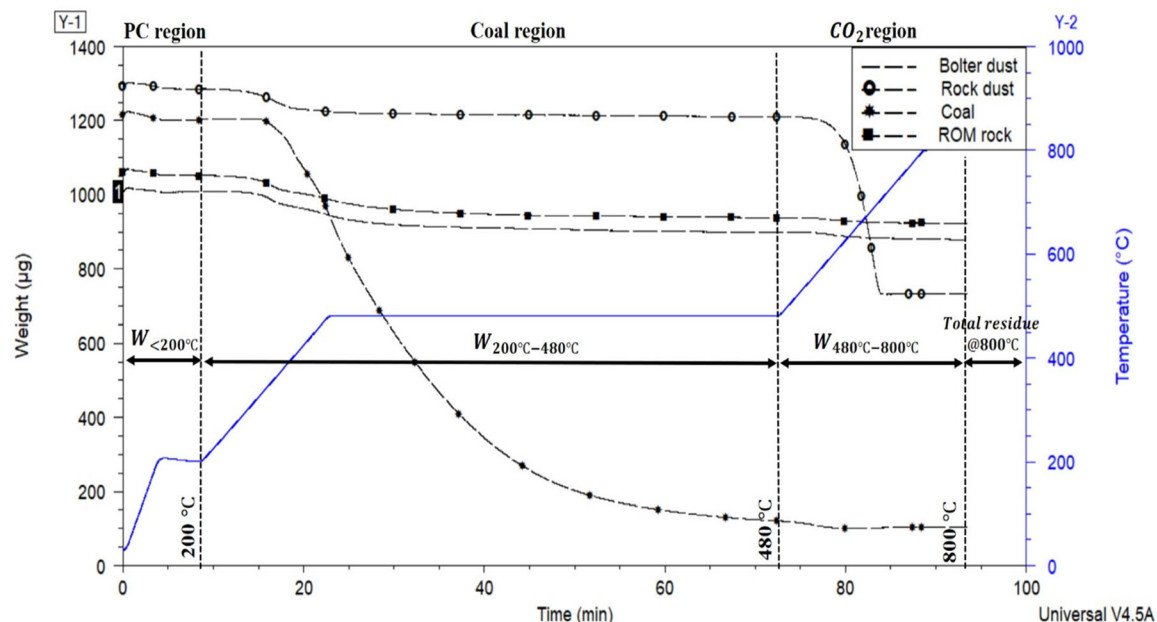
It is noted that the TGA results from Mines 10–25 were included in previous reports aimed at comparing methods for monitoring specific RCMD constituents (see Pokhrel et al. 2022 and 2021). They are included here to enable discussion of source apportionment for the whole dust mass.

### **Data analysis**

The TGA results for the RCMD samples were analyzed to evaluate trends with respect to sampling location, mine region, and mining method. In cases where multiple samples were collected in a given location for a particular mine, the individual sample results were averaged to yield a single set of results for that location in that mine (e.g., the results from the two intake samples in Mine 5 were averaged to yield Mine 5-Intake results). This was done to avoid biasing toward



**Figure 1.** Representative thermograms of RCMD samples collected in the five standard locations (i.e., return, intake, feeder, production, bolter) in a single mine.

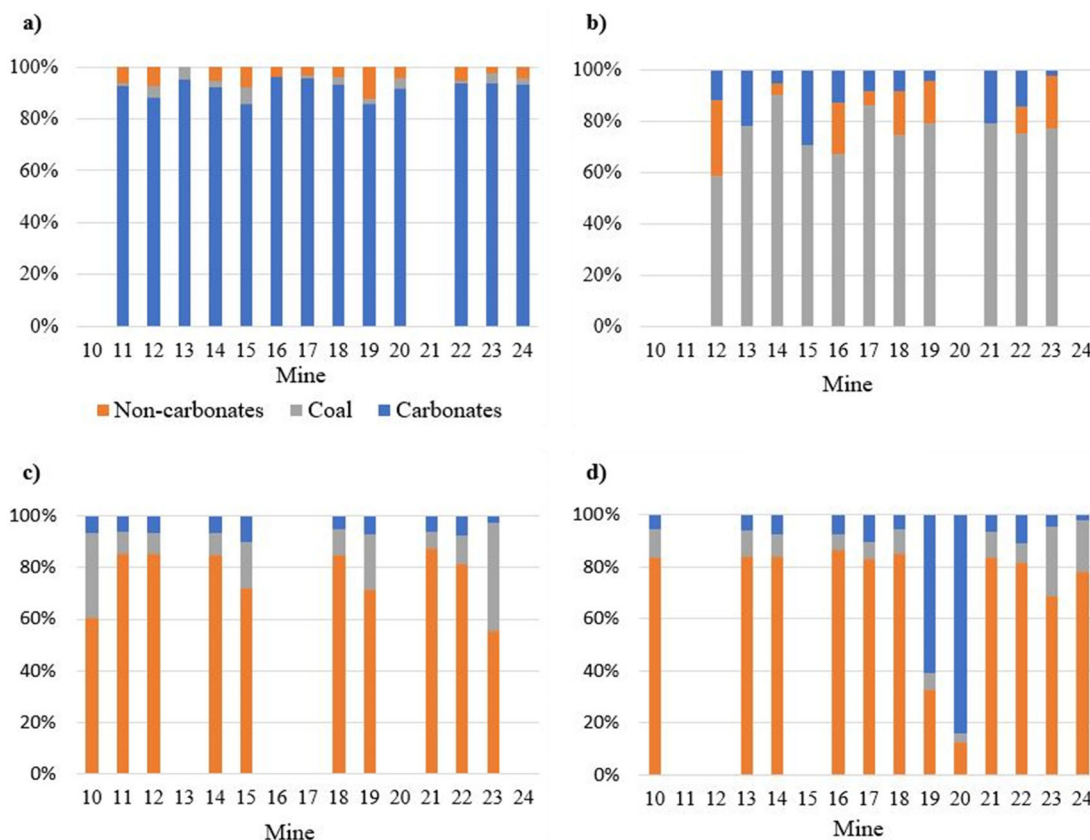


**Figure 2.** Representative thermograms of respirable dust samples generated from the primary source materials (i.e., rock dust, ROM coal, ROM rock, bolter dust) from a single mine.

mines with relatively many samples. All told, results from the 129 RCMD samples collapsed to represent 101 unique sampling locations (i.e., mine  $\times$  location). The 101 results were grouped by sampling location (i.e., I, R, P, B or F, per Table 2); and then within a certain region or mining method (continuous miner or longwall). Given the relatively small numbers of samples from regions other than central Appalachia, regional analysis only considered two general groups:

mines in central Appalachia or those outside of central Appalachia.

To test for significant differences between groups means for each dust component fraction, one-way analysis of variance (ANOVA) was conducted using a 90% confidence level (i.e.,  $\alpha = 0.1$ ). The null hypothesis was that no significant difference exists between means, and the following assumptions were checked: population distribution is approximately normal and



**Figure 3.** TGA-derived component fractions for all 46 respirable dust samples generated from dust source materials: (a) rock dust, (b) ROM coal, (c) ROM rock, and (d) roof bolter.

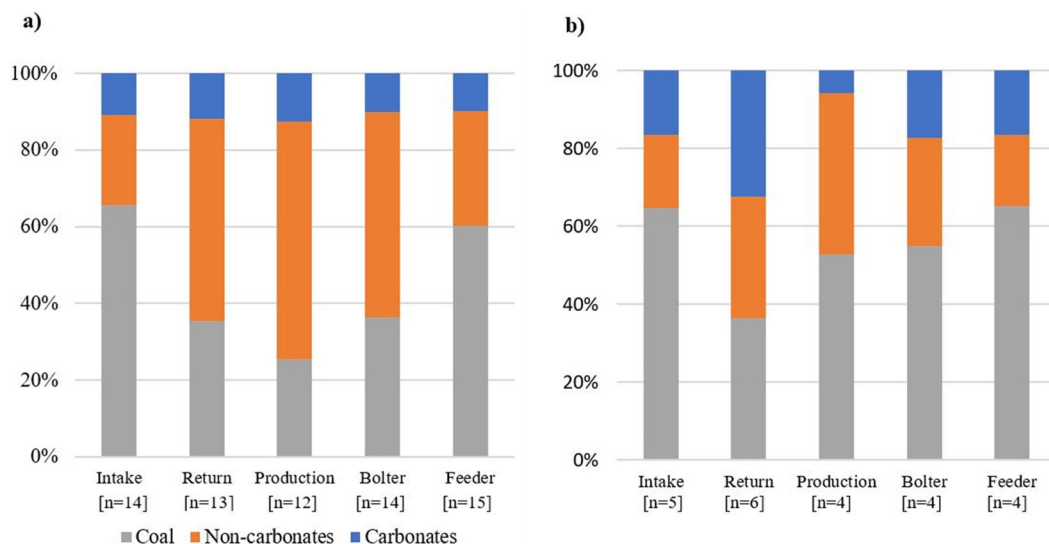
residuals have constant variance. In some cases, the distribution of the residuals was not normal, so logarithmic transformations were applied to those data.

## Results and discussion

### Dust generated from source materials

TGA results for all 46 samples generated from dust source materials are shown in Figure 3 (data are tabulated in the Supplemental Information, Table S1). As expected, the samples generated from all 13 rock dust products represented in this study showed high fractions of carbonates. On average, the carbonate mass fraction in these samples was 92%, and most of the balance was attributed to non-carbonate minerals. For the 11 samples generated from ROM coal, the coal fraction was on average 76%. For seven of these samples, the balance was dominated by non-carbonate minerals as was expected based on imperfect separation between the coal and rock strata (i.e., typically shale and/or sandstone per Table 2). The other four samples (Mines 13, 15, 21, and 22) showed more carbonates, though previous analyses of respirable dust generated from these same ROM coal materials by

Fourier Transform infrared spectroscopy (FTIR) and scanning electron microscopy with energy dispersive X-ray (SEM-EDX) did not indicate significant carbonate content (Pokhrel et al. 2021). It is possible that specific mineral impurities in the coal could partially decompose in the 480–800 °C region (i.e., where weight loss is attributed to CO<sub>2</sub> evolution from carbonates); for example, high-purity kaolinite is expected to lose some of its mass (<15%) due to dehydroxylation around 500 °C (Ptáček et al. 2010). A close inspection of the thermograms from the four samples in question indicated that some weight loss was still occurring end of the long isotherm at 480 °C. To determine the responsible constituent, further analysis (e.g., by X-ray diffraction) would be needed. Nevertheless, this sort of non-ideal behavior for several of the dust samples generated from ROM coal materials underscores the variability in RCMD dust sources—and the benefit of characterizing primary source materials for a particular mine before interpreting TGA results from RCMD samples. Of the 10 samples generated from ROM rock materials, the non-carbonate minerals mass fraction averaged 77%, with most of the balance attributed to coal as expected. Higher coal content in two of the samples



**Figure 4.** Average TGA-derived component fractions for RCMD samples (excluding Mines 19 and 20) by sampling location in: (a) central Appalachia and (b) outside of central Appalachia.

(Mines 10 and 23) could be related to organic content in the rock itself or impurities introduced as the ROM materials was hand sorted. Of the 12 samples generated from the material taken from roof bolter dust collection systems, 10 showed high non-carbonate minerals fractions (72% on average). The other two samples generated from roof bolter materials (Mines 19 and 20) showed high carbonates (61 and 84%, respectively). This is attributed to the limestone roof strata encountered in these two mines (Table 1); the limestone and coal strata were separated by a thin layer of shale such that the limestone was not significantly included in the total mining height, but it was drilled for roof bolting. None of the other mines included in this study are known to encountered carbonate-rich strata.

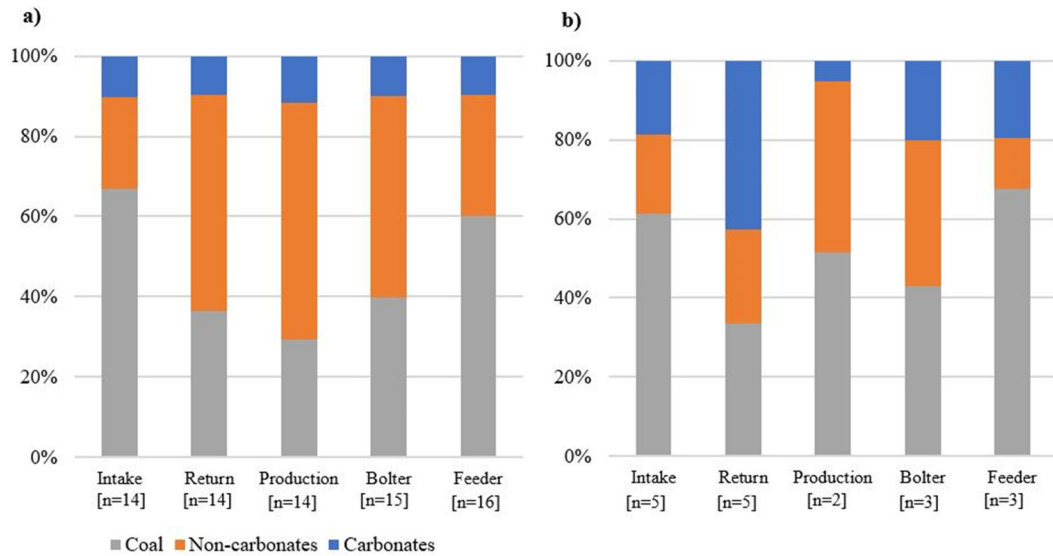
### Mine dust

TGA results for all 129 RCMD samples are given in Table S2. Figures 4 and 5 summarize the results by sampling location and mine region or mining method, respectively, and Tables 3 and 4 present the related ANOVA findings. (Since the limestone rock strata in Mines 19 and 20 confound the ability to roughly associate the carbonates fraction with rock dusting activities, results from these two mines are excluded from the figures and ANOVA, reducing the number of unique sampling locations from 101 to 91.)

Results in the intake and feeder locations are similar between the samples collected in central Appalachian mines vs. others, and indeed no statistical differences were found in these data (Table 3). Results in these locations are also similar when comparing continuous miner

and longwall operations, though the longwall samples tend to show more carbonates. The relatively high fraction of coal in the feeder samples is consistent with the primary dust generating activity in this location (i.e., crushing the ROM material). RCMD analysis using a preliminary TGA application (Phillips et al. 2017) and electron microscopy techniques to investigate particle mineralogy (Johann-Essex et al., 2017; Sarver et al. 2019; 2021) have also indicated relatively higher coal content in the feeder location than other sampling locations represented here. Results from the intake location are also generally consistent with those prior studies. Notably, in mines operating diesel equipment, particulates from engine emissions can contribute substantially to the RCMD concentration in the intake airways, where dust concentrations are typically low overall (Sarver et al. 2019; 2021)—and diesel particulates could contribute to the TGA-derived coal fraction in such samples.

Figures 4 and 5 illustrate some key differences in the bolter, production, and return RCMD sampling locations. For the production and bolter locations, the TGA-derived coal fraction was lower and non-carbonate minerals fraction was higher for samples from central Appalachian mines than from other mines. (Most of these differences were found to be significant per Table 3.) This makes sense based on coal seam thickness and total mining heights shown in Table 1. Mines in central Appalachia were generally cutting more roof rock and less coal in terms of the total mining height—and this means most of the bolting was also in rock—whereas in mines outside of central Appalachia, the coal thickness was closer to the total mining height. Since most of the central Appalachian



**Figure 5.** Average TGA-derived component fractions for RCMD samples (excluding Mines 19 and 20) by sampling location in: (a) continuous miner and (b) longwall mines.

**Table 3.** Summary statistics for comparison of mean TGA-derived component fractions for RCMD samples collected in standard locations in central Appalachia vs. outside of central Appalachia (excluding Mines 19 and 20).

	Intake (n <sub>CA</sub> = 14) (n <sub>O</sub> = 5)		Return (n <sub>CA</sub> = 13) (n <sub>O</sub> = 6)		Production (n <sub>CA</sub> = 12) (n <sub>O</sub> = 4)		Bolter (n <sub>CA</sub> = 14) (n <sub>O</sub> = 4)		Feeder (n <sub>CA</sub> = 15) (n <sub>O</sub> = 4)	
	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference
Coal	0.9253	–	0.9323	–	0.0086	CA < O	0.1384	–	0.6846	–
Carbonates	0.4550	–	0.0106	CA < O	0.6249	–	0.1206	–	0.2945	–
Non-carbonates	0.3932	–	0.0991	CA > O	0.0190	CA > O	0.0727	CA > O	0.3577	–

CA = central Appalachia, O = outside of central Appalachia

**Table 4.** Summary statistics for comparison of mean TGA-derived component fractions for RCMD samples collected in standard locations in continuous miner vs. longwall operations (excluding Mines 19 and 20).

	Intake (n <sub>CA</sub> = 14) (n <sub>O</sub> = 5)		Return (n <sub>CA</sub> = 14) (n <sub>O</sub> = 5)		Production (n <sub>CA</sub> = 14) (n <sub>O</sub> = 2)		Bolter (n <sub>CA</sub> = 15) (n <sub>O</sub> = 3)		Feeder (n <sub>CA</sub> = 16) (n <sub>O</sub> = 3)	
	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference	P-value	Mean difference
Coal	0.6582	–	0.7888	–	0.1367	–	0.8306	–	0.5796	–
Carbonates	0.2682	–	< 0.0001	CM < LW	0.6962	–	0.0475	CM < LW	0.0747	CM < LW
Non-carbonates	0.4874	–	0.0241	CM > LW	0.4307	–	0.4280	–	0.2236	–

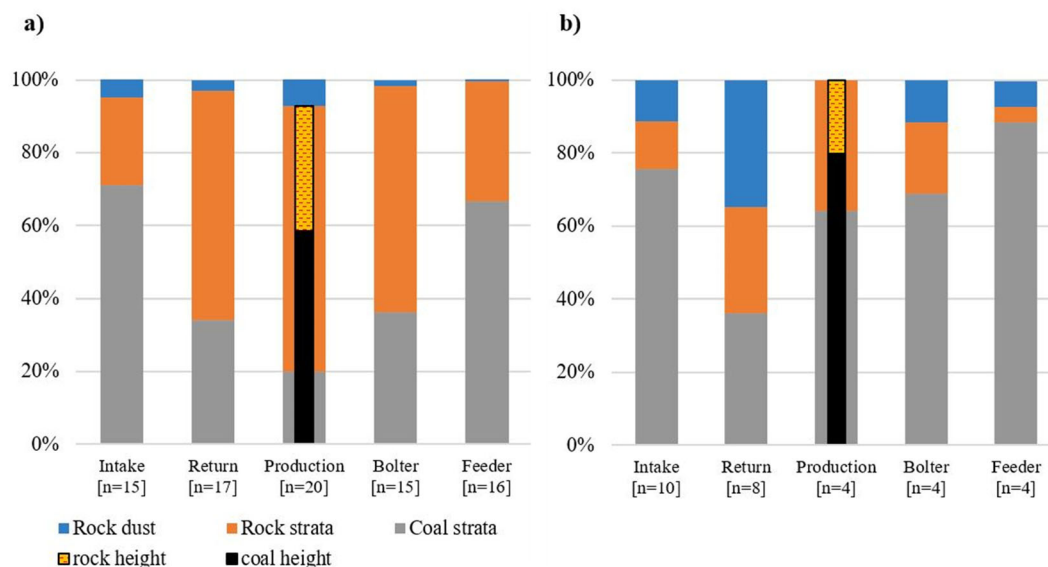
CM = continuous miner, LW = longwall

mines were operating continuous miners, and most of the other mines were using longwalls, the trends in production sample components are similar between Figures 4 and 5.

In the return location, samples from the central Appalachian mines again showed high fractions of non-carbonate minerals. This is consistent with the expectation that dust generated at the production face (i.e., typically by a continuous miner in this region) and/or during roof bolting should dominate the RCMD in the return airway. However, most mines outside of central Appalachia were operating longwalls, with significant rock dusting in the return and in other areas of the mine. This is reflected in the return sample results

(Figures 4 and 5), which show significantly higher carbonate fractions (and lower non-carbonate fractions) for the mines outside of central Appalachia and for longwall mines (Tables 3 and 4). Carbonate fractions were sometimes relatively high in the intake, bolter and feeder locations too for this region, though these differences were not found to be statistically significant on a regional basis (Table 3). When comparing mining methods, the bolter and feeder locations did show significantly more carbonates in longwall mines than in continuous miner operations.

Given that TGA is a mass-based technique and the recovered RCMD samples masses were variable (i.e., from a minimum of just 10 µg to nearly 4,000 µg



**Figure 6.** Average source apportionment results for RCMD samples (excluding Mines 19 and 20) by sampling location in: (a) central Appalachia and (b) outside of central Appalachia.

across all 129 samples), the above statistical analysis was also performed after filtering the TGA results to exclude very low mass samples (i.e., recovered dust mass  $<55\mu\text{g}$ ). This threshold yielded data from 69 unique sampling locations (i.e., about 76% of the 91 unique locations represented in Tables 3 and 4). Tables S3 and S4 present the updated ANOVA results. Since return and production samples were largely unaffected by the data filtering to exclude low mass samples (i.e., just one return and one production sample were excluded), the ANOVA findings are the same for these locations based on both the regional (Table S3 compared to Table 3) and mining method analysis (Table S4 compared to Table 4). Findings for the intake location were also the same (i.e., no significant differences based on region or mining method), despite the fact that 8 of 19 intake samples were considered low mass.

For the feeder and bolter locations, exclusion of low mass samples (6 of 19 and 6 of 18, respectively) produced some additional findings: In central Appalachia, feeder samples showed significantly lower carbonates fractions and bolter samples showed significantly lower coal fractions than samples from other mines (Table S3). However, the bolter samples from continuous miner operations no longer showed lower carbonate fractions than those from longwall mines (Table S4).

### RCMD source apportionment

Figure 3 serves as a sort of proof-of-concept that the three TGA-derived components for RCMD samples (coal, non-carbonate minerals, and carbonates) can be

largely associated with the three primary dust sources in many mines (i.e., the coal strata, rock strata, and rock dusting products). However, each source material contains some impurities. For example, the respirable dust samples generated from ROM coal materials here show some mineral content and, as such, a portion of the mineral content in the RCMD samples might actually be sourced from the coal seam. Likewise, dust generated from the rock strata materials and rock dust products also showed impurities that can affect interpretation of the RCMD sample results. To account for these impurities and enable more accurate interpretation, the TGA-derived results for the source material samples were used to build a simple RCMD source apportionment model. (The relevant model equations are given in Table S5.)

The model was built at the individual mine level using the TGA component fractions determined for the dust generated from the ROM coal (to represent coal strata), rock dust product, and ROM rock (to represent rock strata); a decision was made to use the ROM rock rather than the roof bolter material to represent rock strata since the latter might have accumulated over substantial time (i.e., including well before the RCMD samples were collected). In cases where a particular source material was not available from a given mine, the average component fractions for that material type on a regional basis were used. (For example, since ROM coal was not available from Mine 10, the average TGA results for all ROM coal materials in central Appalachia were used to represent ROM coal in Mine 10.) The model was applied to the RCMD sample results in order to apportion the dust

in each unique sampling location (i.e.,  $n=91$ , as Mines 19 and 20 were again excluded due to the limestone roof strata being a possible confounder for carbonates content).

Results of the source apportionment exercise are shown in Figure 6 on a regional basis. (The results are also shown for continuous miner vs. longwall operations in Figure S1.) Since the TGA component fractions associated with the dust source materials were generally consistent with expectations, Figure 6 appears similar to Figure 5. The relatively smaller height of most blue bars in Figure 6 (vs. Figure 5) is attributed to the fact that some carbonates content has now been apportioned to the coal strata in many mines (i.e., per the results shown in Figure 3). Overall, rock dust appears as minor source of the RCMD, except for in the return location of mines outside of central Appalachia and in longwall mines.

Along with the RCMD source apportionment results, Figure 6 (and Figure S1) shows the average relative heights of coal and rock at the production face during the RCMD sampling. The disproportionate contribution of rock strata to the respirable dust in this location is striking. On average, rock represented 37% of the total mining height in central Appalachian mines included in this study, yet 78% of the of the RCMD sourced from the mine strata (coal + rock) is attributed to the rock. In the mines outside of this region, only about 20% of the total mining height was in rock, but 36% of the RCMD sourced from the mine strata is attributed to the rock. This analysis suggests rock generates about 2x as much respirable dust as coal during mining (i.e., by a continuous miner drum or longwall shearer), or that controls applied at the mine face (e.g., water sprays, scrubbers) are more effective for respirable coal particles than for mineral dust particles. Notably, a previous study using SEM-EDX to characterize RCMD in the same mines studied here similarly found that particles likely sourced from rock strata (i.e., silicates and silica) were much more abundant (number %) than expected based on the relative rock vs. coal mining heights (Sarver et al. 2021); using the SEM-EDX data, the rock strata seemed to produce about 2–3x as much respirable dust as coal.

On the other hand, the results shown in Figure 6 for the feeder breaker location tell a different story. As mentioned above, the feeder breaker is used to crush the ROM material that has just been mined at the production face. In the central Appalachian mines, the relative contribution of rock and coal strata to the RCMD in the feeder location is similar to the relative

heights of rock and coal being mined at the face; and in the outside mines, it appears the rock contributes less dust than might be expected based on its relative height at the face. Comparing the production and feeder results underscores the fundamentally different dust generation and/or control mechanisms in these locations.

## Conclusions

There are commonly three primary sources of respirable coal mine dust (RCMD): the coal strata being mined, the rock strata being mined along with the coal or drilled for roof bolting, and the rock dusting products being applied to mitigate explosibility hazards. This study demonstrates that application of thermogravimetric analysis (TGA) to RCMD can enable fractionation of the dust into three mass components (i.e., coal, non-carbonate minerals, and carbonates), and that these can allow simple dust source apportionment in many mines. A baseline or periodic source apportionment exercise could provide valuable knowledge for mines to assess their local conditions and evaluate how changes in geologic or mining conditions impact RCMD composition. It should be noted that, since source material qualities vary from mine to mine, some knowledge of the primary source materials a priori is key for interpretation of the RCMD component fractions derived from TGA. For example, in mines that encounter carbonate-rich rock strata, this circumstance might confound any attempt to associate the RCMD carbonates fraction with rock dusting activities. Moreover, non-carbonate mineral constituents in dust source materials that partially decompose in the thermal region where only  $\text{CO}_2$  evolution is desired could also result in an apparently high carbonates fraction, and should therefore be taken into account.

Because real dust source materials were available for many of the mines studied here, it was possible to conduct a RCMD source apportionment exercise. Results highlight the inordinate contribution of rock strata to the RCMD present just downwind of the mine production face. This emphasizes a need to improve the effectiveness of dust controls specifically for reducing rock strata-sourced dust during mining.

Results presented here should also inform discussions of thin-seam mining practices, more broadly. Indeed, powerful mining equipment and efficient materials separation processes have enabled the economic extraction of significant rock along with thin coal seams for decades. However, the evolving

understanding of respirable dust generation and related exposure and health implications—not to mention the costs of environmental controls to manage coal waste materials (i.e., the rock rejected in the processing plant)—might warrant new consideration of advanced mining technologies. Technologies capable of safely targeting thin seams of coal without necessitating so much rock extraction could have a variety of benefits.

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## Data availability statement

The authors confirm that the data supporting the findings of this study are available within the article and its [supplementary materials](#).

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