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Considerations for an Automated SEM-EDX Routine for Characterizing Respirable Coal Mine Dust

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Respirable dust in coal mining environments has long been a concern for occupational health. Over the past several decades, much effort has been devoted to reducing dust exposures in these environments, and rates of coal workers' pneumoconiosis (CWP) have dropped significantly. However, in some regions, including parts of Central Appalachia it appears that incidence of CWP has recently been on the rise. This trend is yet unexplained, but a possible factor might be changes in specific dust characteristics, such as particle composition, size or shape.

Prior work in our research group has developed a standardized methodology for analyzing coal mine dust particles on polycarbonate filter media using scanning electron microscopy with energy dispersive x-ray (SEM-EDX). While the method allows individual particles to be characterized, it is very time-intensive because the instrument user must interrogate each particle manually; this limits the number of particles that can practically be characterized per sample. Moreover, results may be somewhat user-dependent since classification of particle composition involves some interpretation of EDX spectra.

To overcome these problems, we aim to automate the current SEM-EDX method. The ability to analyze more particles without user bias should increase reproducibility of results as well as statistical confidence (i.e., in applying characteristics of the analyzed particles to the entire dust sample.) Some challenges do exist in creating an automated routine, which are primarily related to ensuring that the available software is programmed to differentiate individual particles from anomalies on the sample filter media, select and measure an appropriate number of particles across a sufficient surface area of the filter, and classify particle compositions similarly to a trained SEM-EDX user following a manual method. This paper discusses the benefits and challenges of an automated routine for coal mine dust characterization, and progress to date toward this effort.

Keywords: Coal workers' pneumoconiosis, Respirable dust, particle analysis, scanning electron microscopy, Automated SEM

1. Introduction

Coal mining operations generate dust which can be respired into the lungs of workers to cause occupational health diseases such as coal workers' pneumoconiosis (CWP). The mining industry saw dramatic reductions in CWP cases as dust standards and ventilation regulations in underground coal have improved over the past few decades under the Federal Coal Mine Health and Safety Act of 1969 [1]. The Act also established the Coal Workers' Health Surveillance Program through which NIOSH has witnessed first-hand the increase in CWP rates in the eastern United States, particularly Central Appalachia, since the mid-1990s [1-3]. This is of particular concern because the majority of cases have been reported in young coal miners and many of the cases are advanced [1-2]. Further research should be aimed toward determining the cause of increased incidence of CWP in Central Appalachia in order to improve miner health and safety [3]. Little is definitively known regarding the effects of specific dust characteristics (such as size, shape, and chemical composition) on lung disease occurrences in underground miners. Analyzing these dust particle characteristics using scanning electron microscopy (SEM) may be a good place to start. Automated SEM analysis could be particularly advantageous in collecting data from more dust samples at a faster rate.

Automated SEM-EDX analysis has historically been used for applications such as industrial process control and forensics [4]. However, SEM automated analysis hardware and software advancements have made it applicable to a variety of other applications, including mineral samples [4]. Automated SEM is able to analyze features such as inclusions in metals; porosity of geological samples, and samples containing wear debris from combustion engines [4]. Another application for mineral samples is the detection of an anomalous particle within a grouping of thousands of particles of other compositions [4]. This application might be particularly useful to the occupational health field for the analysis of dust samples containing atypical or hazardous particles.

Some work has been conducted in the realm of automated dust particle analysis. Deboudt et al. [5] performed automated SEM-EDX particle analysis on dust samples collected on the Atlantic coast of Africa. As in our project, this group collected airborne particulate samples on polycarbonate filters and ran an SEM at an accelerating voltage of 15kV. Using the Link ISIS Series 300 Microanalysis system developed by Oxford Instruments, this group was able to collect spectral data for individual particles with a 20 second acquisition time [5]. Even faster rates of data collection can be achieved though. Ritchie and Filip [6] recently undertook an effort

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to optimize the speed of automated particle analysis by SEM-EDX and demonstrated data collection at approximately three particles per second. They employed a structured query language database that stores millions of particle records and is able to simultaneously classify multiple particles to multiple categories [6]. The authors' research group does not necessarily need to be analyzing particles at that rate, though aiming to speed up analysis time by a few seconds and perhaps creating a comprehensive particle database could be beneficial toward research efforts.

Other researchers have also worked on multi-frame particle analysis using the SEM. Fritz, Camus, and Rohde [7] have done work with automated microscope stage analysis that can cover hundreds of frames in one run to ensure total sample coverage. For applications in the mining industry, this ability is particularly attractive for particle sizing and the analysis of respirable dust particles. Collecting data for particles over the entire sample can be necessary for statistical significance [7]. The authors' research group also plans to employ automated multi-frame analysis.

2. Previously Developed Standard Dust Characterization Method

Prior work in the authors' research group has developed a standardized methodology for analyzing coal mine dust particles on a polycarbonate filter [8-10]. This method was developed using an FEI Quanta 600 FEG environmental scanning electron microscope (ESEM) (FEI, Hillsboro, OR) equipped with a Bruker Quantax 400 EDX spectroscope (Bruker, Ewing, NJ). The ESEM is operated under high vacuum conditions at a voltage of 15kV with a spot size of 5.0µm and at the optimal working distance of 12-13mm. Bruker Esprit software is used to collect spectra results for the classification of individual particles. The "spot" analysis function of the ESEM software is used in conjunction with the EDX software to generate elemental spectra.

Six compositional classification schemes were developed for coal mine dust particles based on peak elemental spectra heights of aluminum, calcium, carbon, copper, iron, magnesium, oxygen, potassium, silicon, sodium, sulfur, and titanium. The six classifications are "alumino-silicate," "carbonaceous," "carbonate," "heavy mineral," "mixed carbonaceous," and "quartz." Any particles that do not fit into these categories are termed "other." Data on particle size and shape is also collected in this dust characterization method. The long and intermediate dimensions of each particle are measured using the line measurement tool provided in the ESEM imaging software. The shape is qualitatively classified based on user interpretation as either "angular," "rounded," or "transitional."

The sample analysis routine begins by focusing the SEM at 10,000x magnification to provide optimal resolution for analyzing particles in the desired size range of 0.5-8.0µm in diameter. Two horizontal lines are drawn 2µm apart, centered on the screen and spanning the width of the screen, using a line measurement tool, as depicted in Figure 1.

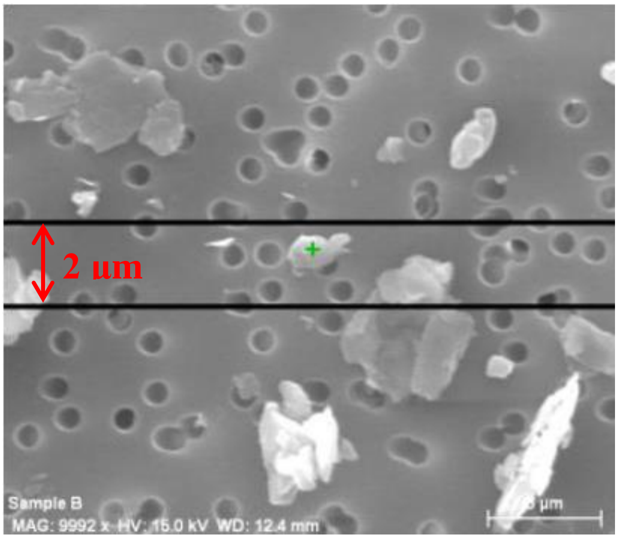


Fig. 1. Example of the horizontal lines drawn 2 µm apart for particle selection. Only particles touching this region are to be selected for analysis [8].

The stage is moved so that the first field to be analyzed is three screen shifts from the outer, left edge of the filter, 2.25mm (one quarter of the filter diameter) down from the top of the filter. Moving from left to right and top to bottom each particle with a long dimension greater than 0.5µm intersecting the space between the two horizontal lines and falling completely within the field of view is analyzed. Up to ten particles meeting the specifications are characterized per field in order to ensure that at least ten fields are analyzed and increase the representativeness of the results. Figure 2 shows a backscatter detector image of a typical field of ten particles that would be analyzed.

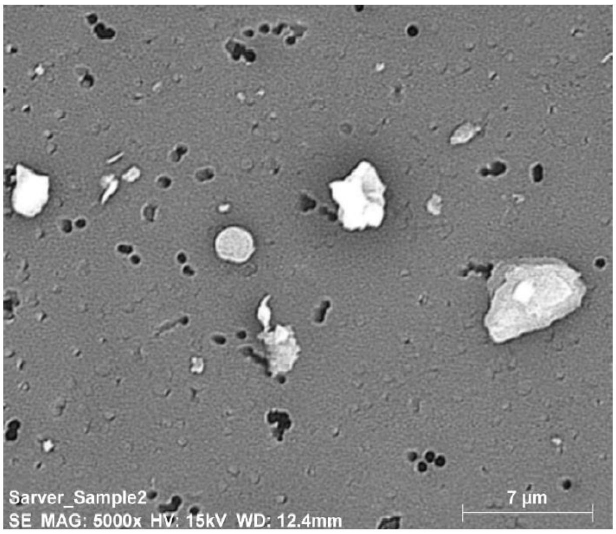


Fig. 2. Example of a typical field of particles to be analyzed (at 10,000x magnification).

Once the first field has been analyzed, the next field of view to the right is analyzed. If fewer than 100 particles have been analyzed upon reaching the edge of the first row, the stage is shifted so that the field of view is 4.5mm (one half of the filter diameter) from the top of the filter and the same procedure for the previous row is followed. If fewer than 100 particles have been analyzed upon reaching the edge of the second row, the stage is shifted once more so that the field of view is 6.75mm (three quarters of the filter diameter) from the top of the filter, following the previous procedure. Figure 3 depicts this analysis routine in terms of filter navigation under the SEM.

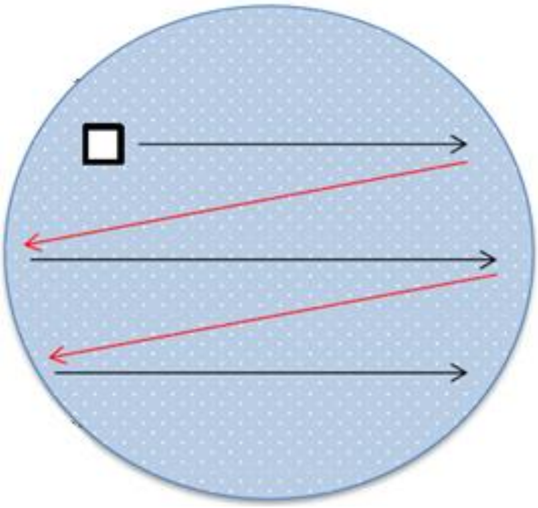


Fig. 3. Illustration of a 9 mm diameter polycarbonate filter and navigation routing for SEM-EDX analysis. The box represents the first frame in which particles are selected for characterization; the black arrows define the directions for successive screen shifts between characterization frames. When one horizontal line of analysis is complete (black arrows), the red arrows define shifting back to the left side of the filter to continue analysis on the next horizontal line [8].

The manual method has the capacity to analyze 100 particles on one sample in 75-90 minutes, depending on user experience and sample characteristics (e.g., particle density); despite the wealth of information that can be obtained, the method is clearly too time-consuming to be practical for a large number of samples.

3. Automation of the Standard Dust Characterization Method

Considering the need to significantly speed up particle characterization, efforts to automate the above routine have recently been initiated. This work is being developed using the same ESEM-EDX system previously mentioned, and several special features available for add-on to Bruker’s Esprit software. A major benefit to automation is that the software can characterize particles at a magnification that is ten times lower than the magnification required for the standard dust characterization method. Figure 4 shows a backscatter detector image of a typical field of particles that would be analyzed using the automated routine.

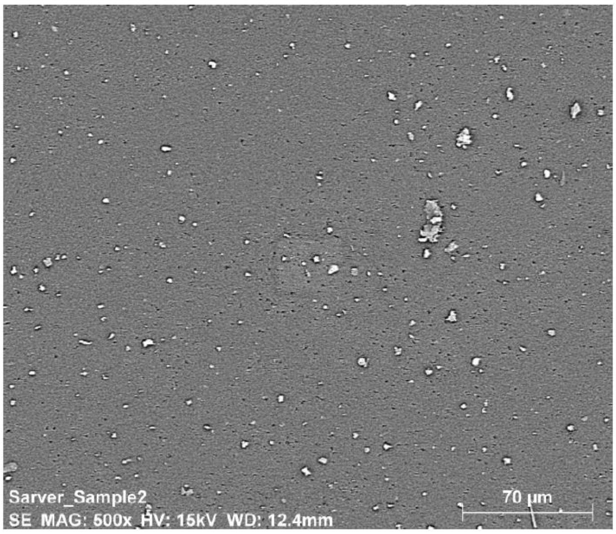


Fig. 4. Example of a typical field of particles (at 1,000x magnification).

This image comes from the same sample as Figure 2; however, by being able to analyze particles at 1,000x versus 10,000x magnification, many more particles can be analyzed per frame. Once the first frame is selected, the imaging tool in the Esprit software is used to pull the image of the frame from the SEM software and import it for analysis. A special feature in Esprit allows for rules and filters to be applied to the image so that the software is programmed to identify dust particles. Here, a binary image can be created and settings can be adjusted so that the software distinguishes particles as white and the filter as black. Figure 5 depicts the binary imaging process in Esprit.

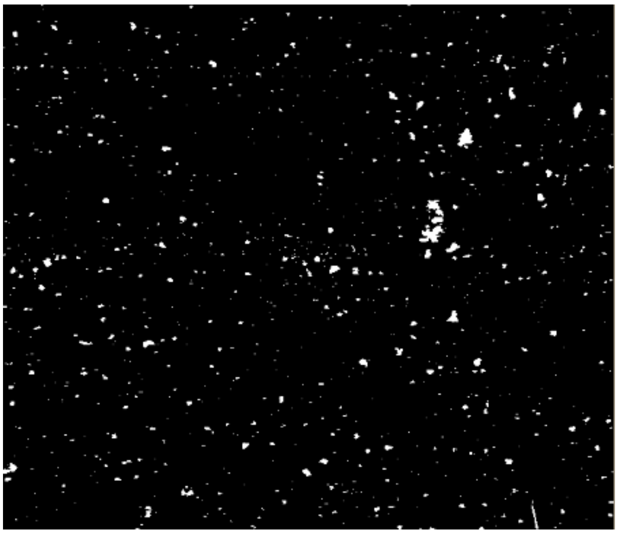


Fig. 5. Example of a binary image of a typical particle field (at 1,000x magnification). This is the same particle field as in Figure 4.

Once all settings have been adjusted for particle identification, particle sizing can be conducted. Figure 6 depicts the particle field once all the particles have been sized. The software outputs data such as length, width, and shape factor to characterize the particles by size and shape, as shown in Figure 7.

This single frame contains 171 particles that can be analyzed, 71 more particles than would have been analyzed in up to ten frames using the standard dust characterization method. This particle sizing routine minimizes user interpretation of the long and intermediate particle dimensions which was required for the standard dust characterization method.

Once sizing is completed, the particle classification scheme can be implemented. The Esprit software allows for particle classification based on the weight percent of elements detected in spectral analysis. Therefore, rules can be set for the maximum or minimum elemental weight percentages required for various particle classification categories. We have currently developed rudimentary rules and particle classification categories to demonstrate the utility of automated analysis and its potential for respirable dust particles from coal mining environments. The preliminary categories are based on typical elemental weight percentages observed for particles classified using the manual method. Figure 8 displays chemistry results for some particles identified in Figure 6, showing the weight percentages of specific elements considered by the classification rules. Figure 9 shows particle classification results in a bar chart as a useful visual tool.

It should be noted that the chemistry classification categories are not currently developed enough to accurately classify every particle (i.e., as it would be classified manually). This is especially true for carbonaceous particles because they can contain small weight percentages of elements other than carbon and oxygen. This does not allow them to be classified under the original carbonaceous category that only sets parameters for carbon and oxygen weight percentages. Therefore, in this preliminary work, a second carbonaceous classification category was created with additional elemental weight percentage rules (i.e., “carbonaceous II” in Figure 9). By doing this, carbonaceous particles that were previously not classified are classified in the second carbonaceous category. A particle chemistry analysis feature in the Esprit software can classify each particle detected in the frame.

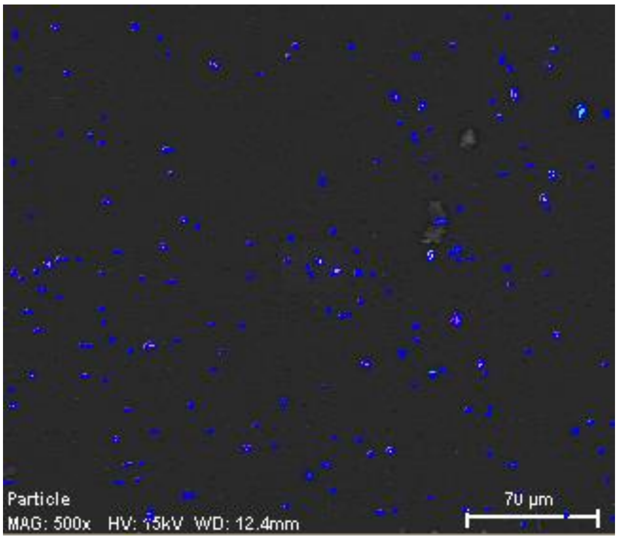
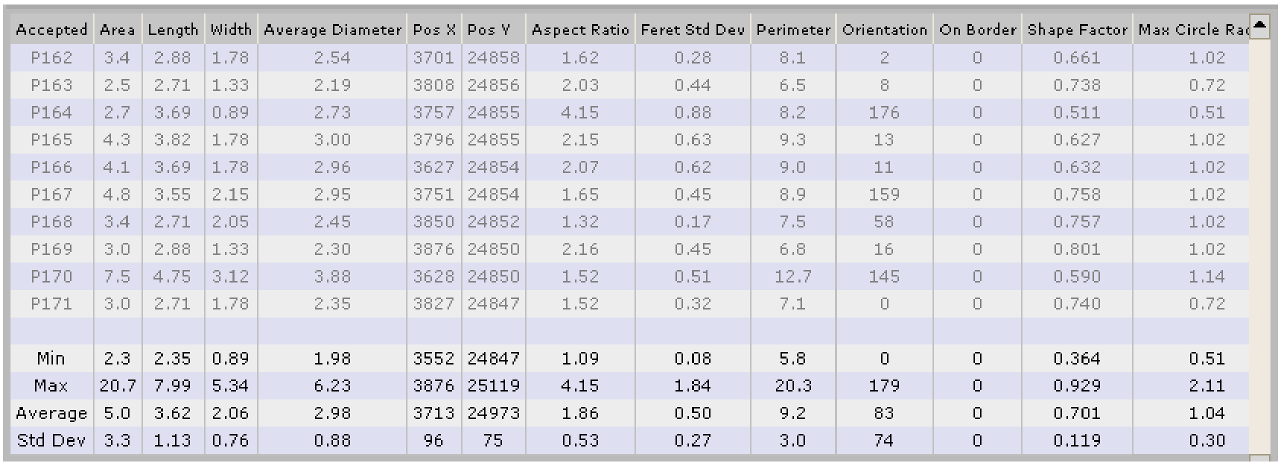
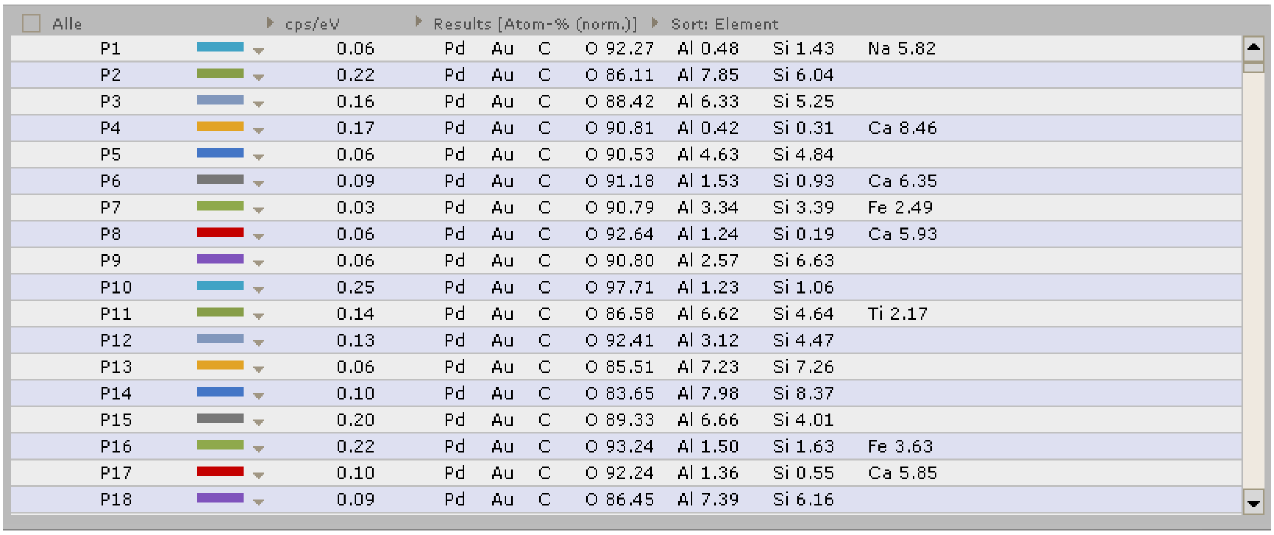


Fig. 6. Example of the particle sizing process in Esprit. This is the same particle field as in Figure 4. Each particle found that is accepted for analysis is outlined in blue while undergoing size classification.



Accepted	Area	Length	Width	Average Diameter	Pos X	Pos Y	Aspect Ratio	Feret Std Dev	Perimeter	Orientation	On Border	Shape Factor	Max Circle Rac
P162	3.4	2.88	1.78	2.54	3701	24858	1.62	0.28	8.1	2	0	0.661	1.02
P163	2.5	2.71	1.33	2.19	3808	24856	2.03	0.44	6.5	8	0	0.738	0.72
P164	2.7	3.69	0.89	2.73	3757	24855	4.15	0.88	8.2	176	0	0.511	0.51
P165	4.3	3.82	1.78	3.00	3796	24855	2.15	0.63	9.3	13	0	0.627	1.02
P166	4.1	3.69	1.78	2.96	3627	24854	2.07	0.62	9.0	11	0	0.632	1.02
P167	4.8	3.55	2.15	2.95	3751	24854	1.65	0.45	8.9	159	0	0.758	1.02
P168	3.4	2.71	2.05	2.45	3850	24852	1.32	0.17	7.5	58	0	0.757	1.02
P169	3.0	2.88	1.33	2.30	3876	24850	2.16	0.45	6.8	16	0	0.801	1.02
P170	7.5	4.75	3.12	3.88	3628	24850	1.52	0.51	12.7	145	0	0.590	1.14
P171	3.0	2.71	1.78	2.35	3827	24847	1.52	0.32	7.1	0	0	0.740	0.72
Min	2.3	2.35	0.89	1.98	3552	24847	1.09	0.08	5.8	0	0	0.364	0.51
Max	20.7	7.99	5.34	6.23	3876	25119	4.15	1.84	20.3	179	0	0.929	2.11
Average	5.0	3.62	2.06	2.98	3713	24973	1.86	0.50	9.2	83	0	0.701	1.04
Std Dev	3.3	1.13	0.76	0.88	96	75	0.53	0.27	3.0	74	0	0.119	0.30

Fig. 7. Example of the particle sizing results for some particles shown in Figures 4-6. The accepted particles are all numbered and listed in order in the first column. Other particle properties are provided for each particle in subsequent columns. At the bottom of the results page, minimum, maximum, average, and standard deviation values are provided for each particle property.



☐ Alle		cps/eV	Results [Atom-% (norm.)]				Sort: Element		
P1	🟦	0.06	Pd	Au	C	O 92.27	Al 0.48	Si 1.43	Na 5.82
P2	🟩	0.22	Pd	Au	C	O 86.11	Al 7.85	Si 6.04	
P3	🟦	0.16	Pd	Au	C	O 88.42	Al 6.33	Si 5.25	
P4	🟧	0.17	Pd	Au	C	O 90.81	Al 0.42	Si 0.31	Ca 8.46
P5	🟦	0.06	Pd	Au	C	O 90.53	Al 4.63	Si 4.84	
P6	⬛	0.09	Pd	Au	C	O 91.18	Al 1.53	Si 0.93	Ca 6.35
P7	🟩	0.03	Pd	Au	C	O 90.79	Al 3.34	Si 3.39	Fe 2.49
P8	🟥	0.06	Pd	Au	C	O 92.64	Al 1.24	Si 0.19	Ca 5.93
P9	🟪	0.06	Pd	Au	C	O 90.80	Al 2.57	Si 6.63	
P10	🟦	0.25	Pd	Au	C	O 97.71	Al 1.23	Si 1.06	
P11	🟩	0.14	Pd	Au	C	O 86.58	Al 6.62	Si 4.64	Ti 2.17
P12	🟦	0.13	Pd	Au	C	O 92.41	Al 3.12	Si 4.47	
P13	🟧	0.06	Pd	Au	C	O 85.51	Al 7.23	Si 7.26	
P14	🟦	0.10	Pd	Au	C	O 83.65	Al 7.98	Si 8.37	
P15	⬛	0.20	Pd	Au	C	O 89.33	Al 6.66	Si 4.01	
P16	🟩	0.22	Pd	Au	C	O 93.24	Al 1.50	Si 1.63	Fe 3.63
P17	🟥	0.10	Pd	Au	C	O 92.24	Al 1.36	Si 0.55	Ca 5.85
P18	🟪	0.09	Pd	Au	C	O 86.45	Al 7.39	Si 6.16	

Fig. 8. Example of the particle chemistry results for some particles shown in Figures 4-6. Each analyzed particle is listed in order in the first column. Information regarding the counts and weight percentages of detected elements are provided for each particle in subsequent columns.

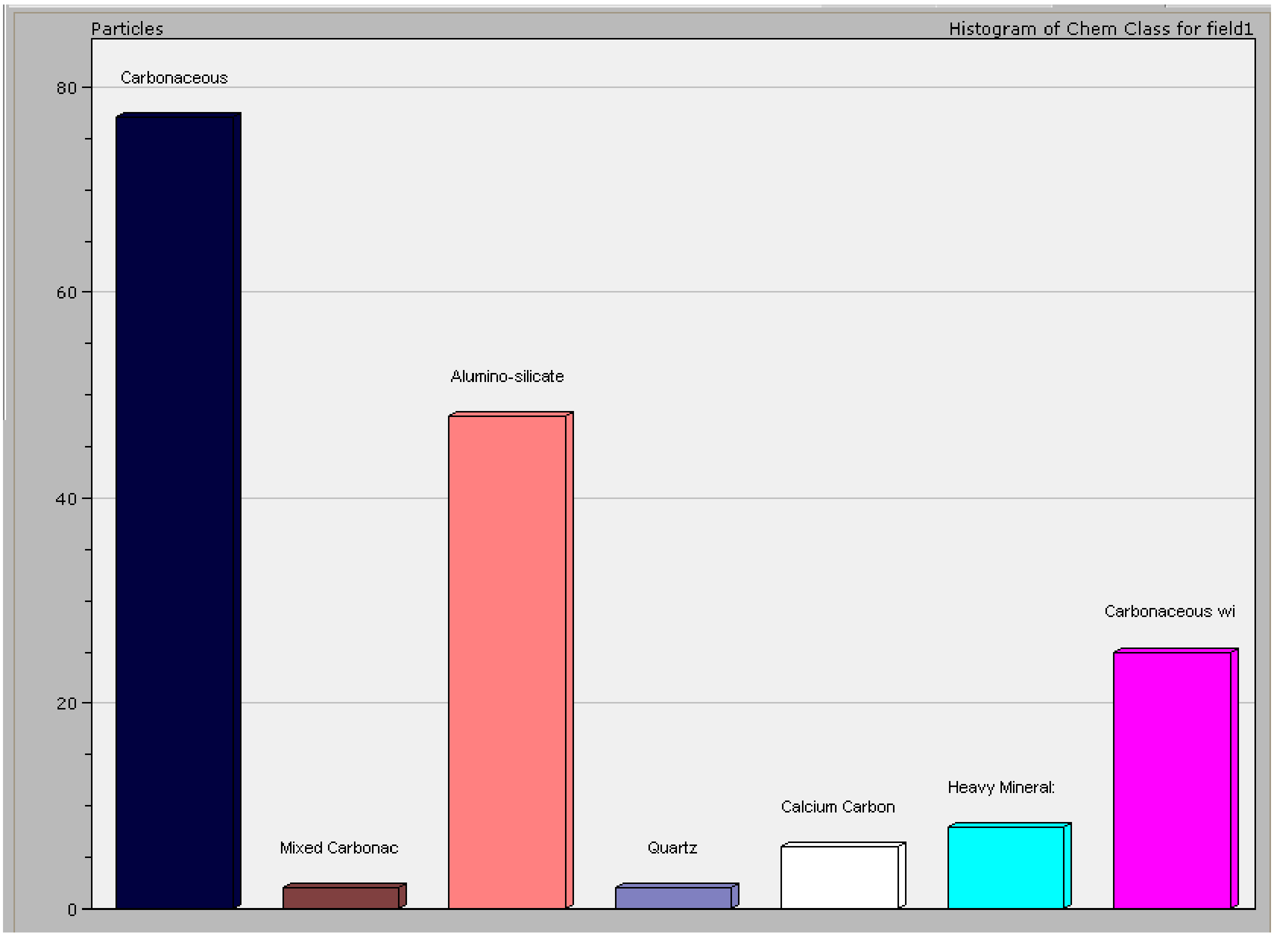


Fig. 9. Example of the particle classification results for particles shown in Figures 4-6. Once each particle is classified based on the specified chemical classification parameters, the Esprit software outputs a histogram with the particle frequency for each class (carbonaceous, mixed carbonaceous, alumino-silicate, quarts, carbonate, heavy mineral, and carbonaceous II)

The automated analysis at the particle density demonstrated in Figure 4 takes just over ten seconds per particle which allows for total analysis of this frame of 171 particles in approximately 30 minutes. At this rate, 171 dust particles are being analyzed two or three times faster than just 100 dust particles using the standard dust characterization method. It is expected that with several modifications in basic operating parameters of the SEM-EDX system, the efficiency can be greatly increased. The majority of particles in this frame were classified as carbonaceous while many particles were classified as alumino-silicates and fewer were classified as heavy minerals, carbonates, mixed carbonaceous, and quartz.

Another special feature in the Esprit software allows for the automation of the microscope stage movement from frame to frame. The user is able to designate the starting frame position and the ending frame position along the sample filter. Once the entire area of the filter to be analyzed is determined, particle sizing and chemistry can be run frame by frame and the software will export the data after completion. This tool can even be used to run multiple samples consecutively so that up to 16 samples could be analyzed in one run.

4. Discussion

It seems as though developing an automated particle analysis routine is a step in the right direction for our research. The standard dust characterization method is too time-intensive for the amount of particles that can practically be analyzed per sample because the user must interrogate each particle manually. A major advantage to automated particle analysis is the amount of time saved in the lab due to quick, electronic characterization of particles. This is also applied to data entry where automated analysis automatically exports data into Microsoft Excel while data obtained from the standard method must be manually entered. Moreover, results from the standard dust characterization method may be somewhat user-dependent since classification of particle composition and shape currently involves some interpretation of EDX spectra. Other benefits of an automated particle analysis routine are a significant increase in the number of particles than can be analyzed per sample and minimization of user interpretation to acquire results. The ability to analyze more particles without user bias should increase reproducibility of results as well as statistical confidence in obtaining results from a representative portion of the sample.

However, some challenges do exist in creating an automated routine, including training the available software to appropriately make multiple decisions such as those involving differentiation of individual particles from anomalies on the filter media, selection of particles for analysis, and classification of particle composition. Another challenge arises due to the filter media being comprised of carbon and carbon being a key element in the classification of carbonaceous, mixed carbonaceous, and carbonate minerals. We are working to determine whether or not carbon should be deconvoluted for proper mineral identification and other key elements be used to classify these particles in its place. We have determined that gold and palladium should definitely be deconvoluted before spectral analysis because the sample coating is comprised of these elements and can interfere with the chemical identification of the dust particles.

5. Conclusion

There is much work still to be done to refine the particle classification parameters in order to properly automate the particle analysis. Our goal is to determine and implement the correct particle classification categories and their appropriate rules for our samples. To do this, we plan to:

- collect spectral data on many particles from our samples
- determine the appropriate elemental weight percentage thresholds for the classification parameters
- determine which elements should be deconvoluted
- modify the currently developed particle classification categories so that they are able to classify all particles encountered
- ensure that the software is properly classifying all particles

We aim to program the software to classify particles in the same manner that the user would classify them, but without the human error.

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