Robert Cohen, M.D., F.C.C.P.
Clinical Professor
Environmental and Occupational Health Sciences
School of Public Health, 1068 SPHPI, MC-923
1603 W. Taylor Street, Room 1068
Chicago, Illinois 60612

ALPHA FOUNDATION FOR THE IMPROVEMENT OF MINE SAFETY AND HEALTH

Final Technical Report

Project Title: Characteristics of Dust and Risk Factors Associated with the Development of Rapidly Progressive Pneumoconiosis and Progressive Massive Fibrosis

Grant Number: AFC417-1

Organization: University of Illinois Chicago
School of Public Health
Division of Environmental and Occupational Health Sciences

Principal Investigator: Dr. Robert Cohen

Contact Information: Phone (312) 413-3944
Fax (312) 413-5287
Email bobcohen@uic.edu

Period of Performance: January 16, 2018 – January 15, 2022

1.0 Executive Summary

Background: Since the mid-1990s, research and surveillance reports have documented a significant increase in coal workers' pneumoconiosis (CWP), including the most severe forms of progressive massive fibrosis (PMF)[1] and rapidly progressive pneumoconiosis (RPP),[2] in U.S. coal miners. The central Appalachian coalfields appear to be particularly affected.[3,4] There are several possible explanations for these observations, including excessive exposures to total respirable dust, increased exposure to particular dust constituents (e.g., freshly fractured silica and silicates, or greater exposure to smaller particles) that may be the result of changing mining practices.[5,6] However, to date the causal links between specific exposure characteristics and the increase in cases of RPP and PMF have not been fully characterized. The main goal of this study is to characterize the biologically relevant exposures, based on lung tissue pathology and mineralogy linked to mine dust and miner exposure characteristics, associated with CWP in its most severe forms. In addition, we hypothesized that analysis of respirable dusts from representative contemporary coal mines would help identify mining conditions that can be linked to the type of dusts found in the lungs of miners affected with RPP/PMF.

Methods: We compared miners born between 1910 and 1930, "historical miners", to those born in or after 1930 "contemporary miners". Historical miners worked mainly with conventional mining technology that relied on drilling and blasting, whereas contemporary miners spent at least a substantial portion of their mining tenure working with mechanized equipment which employ high powered cutting heads to shear the coal from the mine face.[7,8] We used brightfield and scanning electron microscopy with energy dispersive x-ray spectroscopy (SEM/EDS) to analyze lung tissue specimens from materials archived as part of the National Institute for Occupational Safety and Health's (NIOSH) National Coal Workers' Autopsy study (NCWAS) as well as pathologic specimens from contemporary miners with PMF.[9] Lung pathology specimens from 85 coal miners with PMF were included for evaluation and analysis. We also characterized respirable dust particles from representative coal mine environments.

Results: We found a significantly higher proportion of silica-type PMF (57% vs. 18%, $p<0.001$) among contemporary miners compared to their historical counterparts. Mineral dust alveolar proteinosis (MDAP) was also more common in contemporary miners compared to their historical counterparts (70% vs. 37%, $p<0.01$). *In situ* mineralogic analysis showed the percentage (26.1% vs. 17.8%, $p<0.01$) and concentration (47.3×10^8 vs. 25.8×10^8 particles/cm^3, $p=0.03$) of silica particles was significantly greater in specimens from contemporary miners compared to their historical counterparts**.** The concentration of silica particles was significantly greater when silica-type PMF, MDAP, silicotic nodules, or immature silicotic nodules were present ($p<0.05$).

Dust analysis showed that silica particles appear smaller than other minerals including silicates. In central Appalachian mines, the silica and/or silicate content (number %) near the production face and/or in the return was also significantly higher than in mines outside of this region.

Conclusions: Pathology and mineralogy demonstrate that exposure to respirable crystalline silica appears causal in the unexpected surge in severe disease in contemporary miners. Dust analyses show silica is finer than other minerals and present in greater concentrations in central Appalachia. Our findings underscore the importance of controlling workplace silica exposure in order to prevent the disabling and untreatable adverse health effects afflicting US coal miners.

2.0 Contents

3.0 Problem Statement and Objective

Background Information:

Despite modern mining technology, enhanced dust control and ventilation practices, and regulations that have reduced overall dust levels, coal miners in the US are still at risk of developing chronic respiratory diseases such as CWP as well as other adverse health effects related to their occupational exposures.[10] Although remarkable progress was made in reducing CWP in coal miners in the three decades following passage of the Federal Coal Mine Health and Safety Act of 1969 (Coal Act), recently this trend has reversed (Figure 1).[11] The prevalence of the most severe form of the disease, PMF, also fell dramatically after implementation of the Coal Act and reached historic lows in the 1990s. Of great concern is the increased incidence of both RPP and PMF since that time. [2] Severe disease is now being seen in relatively young coal miners,[1] especially in central Appalachia (Figure 2).[3] For example, a 2016 report described a cluster of 60 cases of PMF identified in current and former coal miners at a single eastern Kentucky radiology practice between January 2015 – August 2016.[4] Coincident with this report, an investigation by National Public Radio pointed to hundreds of additional potential cases.[12,13]

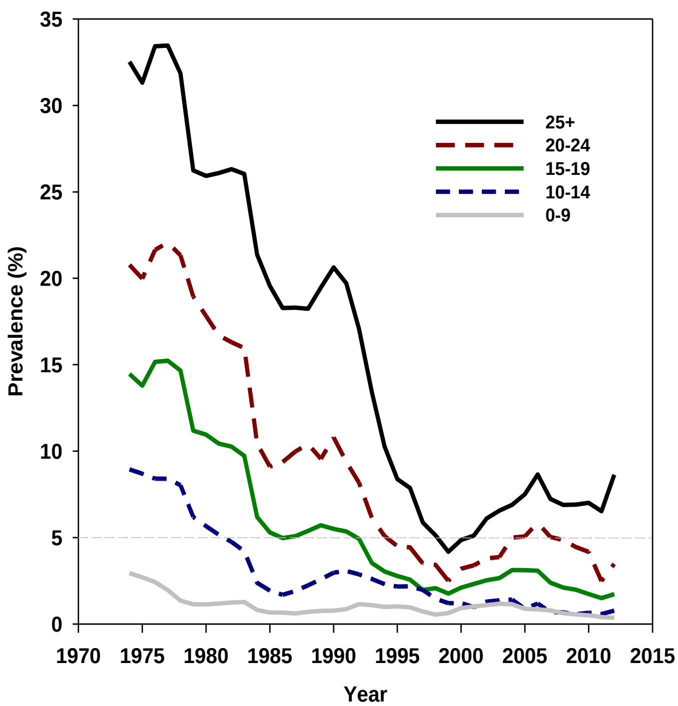


Figure 1. Graph showing steady decline (1974 - 1996) followed by unexpected rise in CWP prevalence (1996-2012).

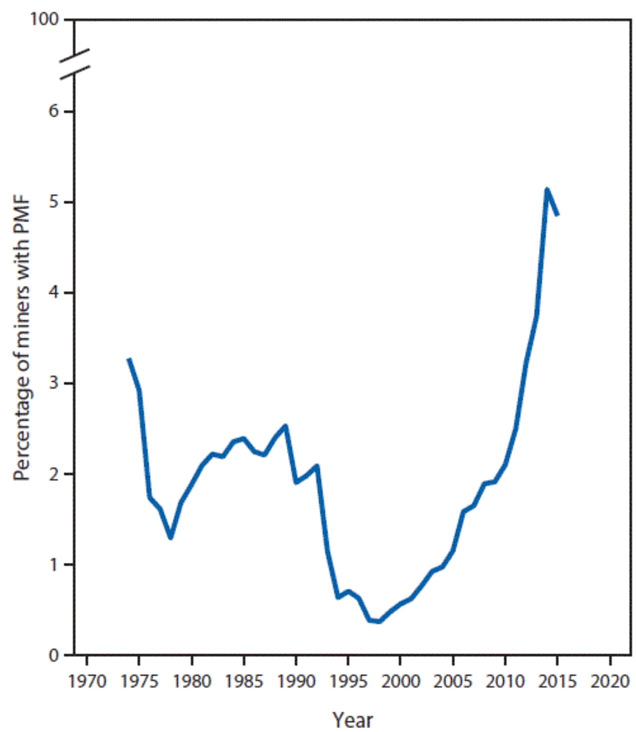


Figure 2 Increasing prevalence of PMF among working underground coal miners with 25 or more years of underground mining tenure (1974 - 2015) in Kentucky, Virginia, and West Virginia according to the NIOSH Coal Workers' Health Surveillance Program.

Problem statement:

The scientific literature on the increase in RPP and PMF has focused mainly on chest imaging findings in these populations and on geographic and workplace characteristics. There is less information on the full spectrum of clinical findings (including in-depth exposure and medical history characterization, smoking, lung function, and particularly lung tissue pathology), and almost no insight into the mineralogy and toxicology of the dust implicated in these severe forms of CWP.

Main hypotheses:

Our overall hypothesis was that RPP and PMF are pathologically and mineralogically distinct from simple CWP and are associated with particular occupational risk factors, such as dust particle characteristics and exposure conditions. Specifically, we hypothesized that there would be significant differences in the mineralogic and pathologic profiles in lung tissues from contemporary coal miners with PMF/RPP compared to those with PMF that developed prior to institution of modern mining methods. In addition, we hypothesized that analysis of respirable dusts from representative contemporary coal mines would help identify relevant geologic deposits and mining conditions that can be linked to the type of dusts found in the lungs of miners affected with RPP/PMF. We further hypothesized that specific variables in the medical and occupational histories of miners with PMF/RPP would provide insight into additional activities and host factors that affect risk for these lung diseases. Together, this information would inform targeted prevention efforts to address recent increases in these deadly coal mine dust lung diseases.

The aims and objectives developed to address the solicitation focus area were:

Aim 1: Detailed clinical characterization of contemporary cases of RPP and PMF as well as historical referent cases from NCWAS biorepository.

Objective 1.1: Select miners with contemporary RPP and PMF from the registry
Objective 1.2: Select historical referent cases from NIOSH National Coal Workers' Autopsy Study (NCWAS) database

Aim 2: Characterize the lung tissue histology in coal miners with contemporary cases of RPP/PMF and compare these findings to lung tissue from historical cases from miners with PMF in the NCWAS biorepository.

Objective 2.1: Characterize pathologic and histologic features of dust-related lung disease
Objective 2.2: Characterize lung dust *in situ*
Objective 2.3: Characterize digested lung dust

Aim 3: Characterize respirable dust particles from representative coal mine environments.

Objective 3.1: Collect dust samples in representative mine environments
Objective 3.2: Characterization of mine dust

Aim 4: Analyze clinical, pathologic, mineralogic, and mine dust data from miners with RPP and PMF compared to historical referent cases from NCWAS to identify risk factors for severe disease and identify opportunities to design prevention strategies.

Objective 4.1: Statistical analysis of data from contemporary cases of RPP and PMF, historical referent cases, and current mine dust samples

4.0 Research Approach

Aim 1: Detailed clinical characterization of contemporary cases of RPP and PMF as well as historical referent cases from the National Coal Workers' Autopsy Study (NCWAS) biorepository.

Objective 1.1: Select miners with contemporary RPP and PMF from the registry

Task 1.1.1: Develop registry of RPP/PMF and identify those with complete data sets and informed consent.

Cases of RPP and PMF were identified through outreach to Black Lung Clinics, radiologists and other providers in areas with high rates of disease, and attorneys and lay advocates representing miners who have filed for disability benefits. Miners were also invited to participate in the registry through recruitment at Black Lung conferences and other miners' events. Deceased miners with RPP or PMF were included in the registry if their next of kin consented to participation. These cases were enrolled in the registry, and detailed clinical and historical exposure phenotyping was undertaken.

Task 1.1.2: Characterize occupational and health history of miners

To characterize the health and occupational exposure history of our subjects participating in the general registry, we developed a comprehensive study questionnaire that allowed data collection within 45 – 90 minutes. The data was entered into a REDCap database for analysis.

Task 1.1.3: Obtain lung tissue specimens from miners

A subset of miners in the registry had undergone lung biopsy or resection, usually for suspected lung cancer or at the time of lung transplantation. As part of consent for study participation, miners were asked to give permission to obtain samples of available lung tissue. In addition to recruiting living patients, the biorepository also included deceased miners from the registry. All had lung tissue available via biopsy, explant, or autopsy.

Objective 1.2: Select historical referent cases from the NIOSH NCWAS database

Task 1.2.1: Scan NCWAS database and select referent cases

In April 2018, team members (including the principal investigator, lead study pathologist, and the study epidemiologist) visited the NIOSH Morgantown, WV office to review NCWAS cases accessioned between 1971 and 1996 to determine their suitability as historical referent cases for the study registry's RPP cases. These cases had been classified by NIOSH pathologists as part of prior studies. A secondary objective was to determine if any RPP cases had been accessioned into the program between 1996 and 2013 given that these cases had not been classified and entered into the database. The case reviews were classified with the assistance of Dr. Anne Hubbs, chief veterinary pathologist, and Dr. Marlene Orandle, associate veterinary pathologist, at the NIOSH Morgantown laboratories. Dr. Francis Green reviewed the 387 PMF cases, giving priority to those that had been accessioned between 1996 and 2013.

From July 28 to August 4, 2018, the principal investigator (Dr. Robert Cohen), and lead study pathologist (Dr. Francis Green), visited the NIOSH laboratories in Morgantown to review NCWAS cases accessioned between 1971 and 1996 to determine their suitability as historical referent cases for the study registry's RPP cases. Additional visits took place in October 2018 and January 2019 in order to review cases accessioned between 1996 and 2013, cases which had not previously been classified by NIOSH pathologists. All PMF cases were reviewed for type of PMF to determine if there has been a change in the type of PMF in recent decades. We found three major histologic types of PMF; those with a predominantly silicotic pattern, those that were predominantly composed of coal dust, and a group with a mixture of the two types. A preliminary analysis of a subset of 325 PMF cases (1971-1996) were classified by type. This initial analysis showed no clear change in type from 1971-1996 but an increase in silicotic type PMF after this date. Based on this preliminary study we refined the diagnostic criteria for types of PMF and broadened our analysis to include seven pathologists, two groups of two working jointly, and three pathologists working independently. This resulted in five classifications for each specimen using the revised diagnostic criteria. Additional cases accessioned between 1996 and 2013 were also added with the result of 456 potential PMF cases, 401 cases had both histologic slides and corresponding blocks of the PMF lesion. We selected 76 NCWAS cases for the study based on birth year, mining tenure, state of mining, and confirmation of PMF. Of these 62 were historical cases with birth year prior to 1930, and 14 were considered contemporary with their birth year in or after 1930. The pathologists were blinded to the results of the other groups.

Task 1.2.2: Abstract occupational and health history data on referent cases

The study team has obtained occupational history data for all NCWAS cases. The data includes total years of coal mine employment, years of surface and/or underground employment, the state in which the miner worked as a surface and/or underground miner and a description of the principal (i.e., longest-held) coal mining job and coal mining job last held by the miner. Examples of job descriptions within the NCWAS data set include "roof bolters and helpers"; "continuous mining machine operators and helpers"; "electricians and helpers, wiremen, mechanics, general repairmen"; and "superintendent, assistant foremen, section bosses, grade foremen." The NCWAS data set also includes detailed smoking history data, including ever/never smoker status; smoking status at time of death; typical number of cigarettes per day; and total pack-years smoked. The study team has obtained copies of the original data collection forms completed by miners' next of kin to confirm and, as needed, correct occupational and smoking history data.

Task 1.2.3: Obtain lung tissue from NCWAS historical comparison cases

Ten initial samples from 1990 – 1996 were identified in the first case review and received in 2018 to pilot test the histologic and mineralogic analysis processes before proceeding with formal selection of the remaining historical referent cases. We received all historical comparison samples by January 2021.

<u>Aim 2</u>: Characterize the lung tissue histology in coal miners with contemporary cases of RPP/PMF and compare these findings to lung tissue from historical cases from miners with PMF in the NCWAS biorepository.

Objective 2.1: Characterize pathologic and histologic features of dust-related lung disease

Task 2.1.1: Selection of tissue blocks for characterization

Blocks were chosen from a review of case details including radiology reports, mining tenure, and pathology reports. Cases from all sites were processed and relabeled in order to maintain blinding of the pathology team as to whether a case was historic or contemporary. The selected blocks were then processed at a collaborating study laboratory, with sections cut for both *in-situ* scanning electron microscopy/energy dispersive x-ray spectroscopy (SEM-EDS) and for bulk digestion for particle analysis. In addition, digitally scanned images obtained from slides made from the blocks were distributed to study pathologists for scoring.

Task 2.1.2: Classification and grading via bright field and polarized light microscopy

A standardized form was created in the REDCap web-based data collection tool, and used to record the number of slides, type of specimen (biopsy, wedge resection, explant, and autopsy), and adequacy of the pathologic materials. The form also included a detailed grading schema for all pathologic lesions associated with mineral dust exposure. The precise definitions and scoring measures for histologic lesions were further clarified and incorporated into a revised form. We developed a new classification system characterizing the PMF lesion type, with "coal"-type PMF having ≤25% silicotic nodules by area in the image(s) reviewed; "silica"-type PMF having >75% silicotic nodules; and "mixed"-type PMF having >25% and ≤75% silicotic nodules.

Diagnostic definitions and severity grading were further refined by consensus through group conference calls conducted by video-conferencing. Six-hundred polarized light and corresponding brightfield digital photomicrographs from 92 cases were taken. We found that digital photomicrographs permit a better characterization of birefringent particles and also provide resolution of fine detail of black particles than whole-slide digital imaging. A subset of these images were selected after a consensus conference review and were then used as standard images to assist in the grading of the birefringent particle abundance and types within PMF lesions.

Agreement on final classification was achieved via video conference calls and produced a consensus grading for all cases. The original grading data for either individual pathologists or groups of pathologists was retained for further analysis of inter- and intra-observer variability.[14] A total of 85 cases were ultimately selected for pathology classification. These included 23 contemporary and 62 historical cases.

Objective 2.2: Characterize lung dust *in situ*

Task 2.2.1: Perform *in situ* mineralogic analysis

We obtained tissue samples from evaluable cases and completed analysis of 50 specimens. All of these samples were de-identified and processed for blinded reading by study pathologists using brightfield and polarized light microscopy as described above, followed by characterization of the mineralogy by *in situ* analysis and by lung tissue digestion.

Samples were analyzed manually by SEM-EDS using the developed morphometric protocol for quantitative particle analysis, which was optimized using the tissue samples received on our first case. Each sample analysis takes approximately 8-10+ hours. For each sample, we recorded the field size (area), the number of fields searched, numbers of particles counted per field and the number of individual particles analyzed using EDS. The long axis diameter of individual particles was also recorded, along with EDS spectra of individual particles. Hematoxylin and eosin-stained microscopic slides were received for each case and incident light and polarized light microscopic images representative of the particle distribution were uploaded for review by other pathologists.

Objective 2.3: Characterize digested lung dust

Task 2.3.1: Determine the relative mass concentration of total lung dust and specific constituents

Specific Constituents: An overview of the methods to digest lung tissue and collect the particulate matter and to characterize the particulate matter using SEM-EDS are described in Lowers et al., 2018.[15] However, alterations to that method were needed as the particle load of the lung samples examined for this project were greater than those examined during the method development described in Lowers et al., 2018.[15] Specific changes included 1) use of a 20 mm formalin fixed, paraffin embedded tissue scroll as opposed to 60-100 mm thick scrolls; 2) the dried tissue was transferred to 25 mL of bleach and reacted for 2 hours; 3) the suspension was filtered through a 25 mm diameter 0.1 mm track etch polycarbonate filter; 4) particles less than 0.1 um in the longest dimension were excluded; and 5) the stopping criteria were set to 2000 particles or 110 fields of view. The automated particle analysis was acquired on an FEI1 field emission scanning electron microscope operated at 15 kV, spot size 5, working distance of 11 mm, objective aperture set to 30 mm, and the magnification set to 3000 for a 48-mm-wide field of view. The energy dispersive spectra were acquired with an Oxford Instruments1 x-Max 50 mm^2 silicon drift detector. EDS spectra and particle morphology were processed using Oxford Instruments Aztec® Micoanalysis Software suite.

A summary of the data collection process is displayed in Figure 3. The upper left pane shows the backscattered image with brightness and contrast set so that the filter is almost black and particles are brighter. The particle selection thresholds were set to ignore the filter and select all pixels that are brighter than the filter. The lower left pane shows part of the classification tree used to assign labels to each particle. The classification was confirmed manually. The particles' classification, area, aspect ratio, breadth, length, and perimeter of the particles was recorded in a table as shown on the right pane of Figure 3. Data tables for each sample were joined to one Excel file that contains all chemical and morphological data for all samples. To compare the particles identified in the digested lung tissue to mine dust results, particles were also classified using the parameters described in Sarver et al., 2021.[16]

Specific constituents were normalized to volume of tissue (cm^3) by multiplying the tissue thickness by the tissue area. Tissue areas were determined by Jeremy Hua (National Jewish Health) using the bioformats plugin to ImageJ.

Relative mass concentration: To determine the relative mass concentrations of selected elements from the digested tissue, bleach filtrates and particulate matter collected from the method described above were submitted for inductively coupled plasma mass spectrometer analysis (ICP-MS).

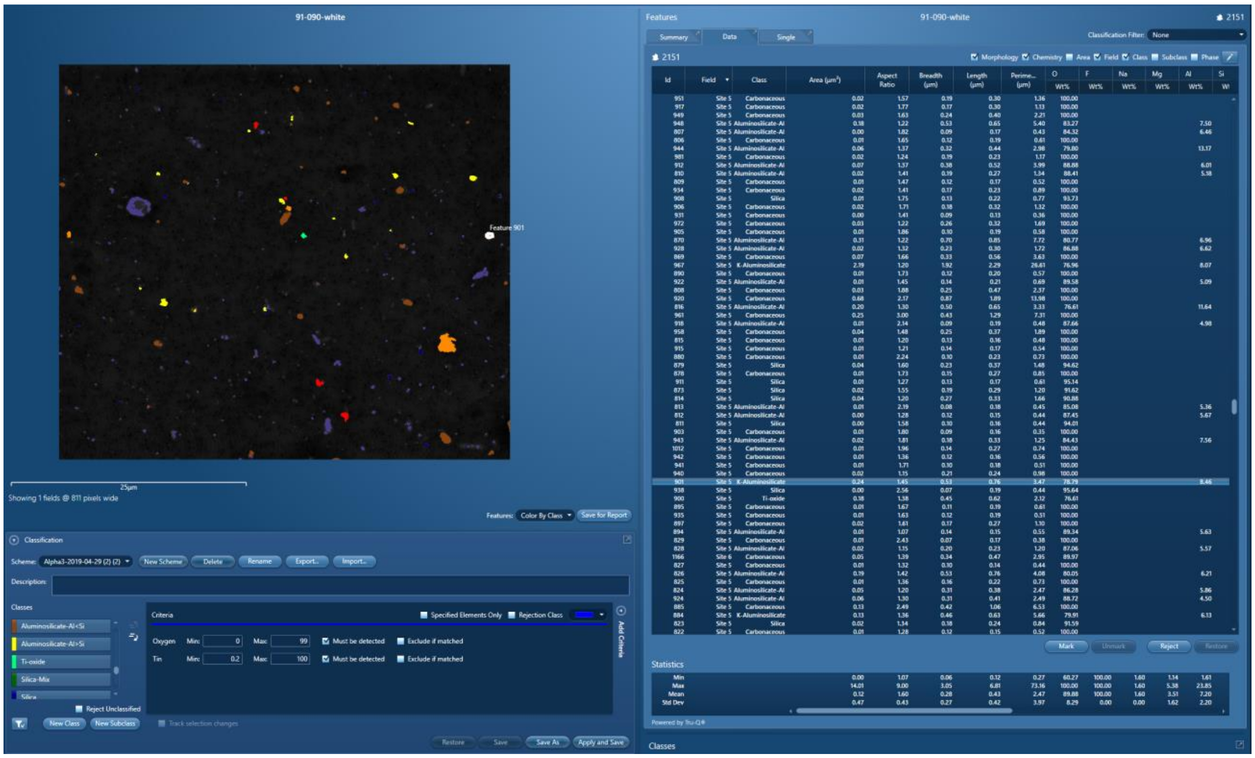


Figure 3 Screen capture of the data collection process to record the chemical, morphological, and classification parameters of particles released from digested lung tissue.

Bleach filtrates preparation: Samples were logged for unique identification and tracking. Samples, approximately 30 mL, were received in plastic bottles. Initially, each solution was transferred to 30 mL Teflon vials (Savillex) and heated on a hot block at 120 °C to evaporate the solvent. As solvent evaporated, greenish-yellow molten salt matrix was obtained - a highly alkaline NaOH-NaOCl matrix. At this point, concentrated nitric acid (HNO_3, trace metal grade, Fisher Scientific) was added drop-wise on the alkaline salt to neutralize the contents. Addition of HNO_3 caused strong effervescence that completely seized upon adding 5-6 mL of HNO_3 indicating all NaOH-NaOCl was neutralized. The contents were heated at 120 °C to evaporate the excess acids until near dryness, yielding a bright white salt matrix (e.g., NaCl-$NaNO_3$). In cases, salts matrix was incompletely neutralized (e.g., showing yellow color), additional HNO_3 was added, and contents were heated to near dryness. The dried sample in 30 mL Teflon vessel was dissolved with 2 mL of 10% (v/v) trace metal grade HNO_3 and transferred to acid-cleaned 15 mL test tubes and diluted to 15 mL with deionized water (18.2 MΩ-cm). The acidity of the final solution was around 1% (v/v) HNO_3.

Particulate matter preparation: Samples were logged for unique identification and tracking. Samples were received in plastic bottles containing suspended particles in approximately 100 mL water along with polycarbonate (PC) filter. Initially, bottles were placed in ultrasonic bath and sonicated for 1 hour to release attached particles from the PC filter. After sonication, the PC filter was removed from the solution/suspension and rinsed into its own bottle. The suspension was shaken well and half of the solution was transferred to 70 mL Teflon vial (Savillex) and then placed on a hot block and heated at 130 ℃ to evaporate the solvent. As solvent evaporated, remaining solution was added to the vial and continued heating until a 0.5 to 1 mL solution remained. At this stage, 2 mL nitric acid (HNO_3, trace metal grade, Fisher Scientific) 0.5 mL hydrochloric acid (HCl, trace metal grade, Fisher Scientific) and 0.5 mL hydrofluoric acid (HF, trace metal grade, Fisher Scientific) were added to the vials. Lids were closed and contents were digested 140 ℃ for 4h. After digestion, lids were opened, and acids were evaporated at 120 ℃ to near dryness. Then, 1 mL HNO_3 and 0.5 mL perchloric acid ($HClO_4$, trace metal grade, Fisher Scientific) were added to the vials. Lids were closed and contents were digested at 130 ℃ for 2h. After digestion, contents were evaporated to near dryness at 120 ℃ to get rid of excess acids. Then 2 mL of 10% (v/v) HNO_3 was added to each vial, dissolved the residue and transferred to 15 mL acid-cleaned polypropylene test tube. Volume was completed to 5 mL with deionized water (18.2 MΩ-cm). Final acidity of the samples was around 4% (v/v) HNO_3.

Instrumental analysis: Elemental analysis of the prepared sample solutions was performed with inductively coupled plasma mass spectrometry (ICP-MS). Solutions were directly analyzed for ICP-MS determinations on a Perkin Elmer NexION 2000B ICPMS instrument. All samples were analyzed from undiluted solutions (5 mL volume), except those that were treated with bleach. Bleach digests were analyzed from 10-fold diluted solutions. The quartz-based sample introduction system (spray chamber and nebulizer and injector) was replaced with HF-inert teflon nebulizer and spray chamber (Savillex) and sapphire injector (Perkin Elmer) to accommodate the analysis of HF-containing sample solution. Instrumental operating conditions for ICP-MS are summarized in Table 1.

Table 1. ICP-MS instrument operating parameters and conditions.

Parameter	ICP-MS
RF power (W)	1500
Plasma argon flow (L/min)	13.0
Auxiliary argon flow (L/min)	0.85
Nebulizer flow (L/min)	0.86
Sample flow rate (mL/min)	0.5
Spray Chamber	PFA
Spray chamber temperature (ºC)	Ambient ~ 22
Scan mode	Peak hopping
Dwell time (ms)	30
Point/peak	1
Scans/peak	3
Scans/replicate	5
Measurement modes	KED and Standard

Calibration and quality control/assurance: The ICP-MS was calibrated with aqueous multielement standards in 2% HNO_3. Number of calibrants varied from 5 to 6 in addition to a calibration blank and required a R^2 of at least 0.995. Quality control/assurance for the instrumental determinations were carried out by analysis of several USGS (T-series), and NIST water reference materials. These include T-215, 221 and 239, and SRM 1640 (Trace Elements in Natural Water). Sub-samples of the reference materials were analyzed by ICP-MS.

Blanks, blank correction and experimental results: Reagent solutions including deionized water, bleach, H_2O_2, HNO_3, HCl, and HF used for sample preparation and processing were analyzed for trace metal impurities. Method blanks (n=6) were also prepared and analyzed for trace metal impurities at the same dilution factor as the samples. Reagent-corrected concentrations were then corrected for the method blanks to calculate the final elemental concentrations in experimental samples.

Task 2.3.2: Determine the size distributions of lung dust particles

As described above, morphological parameters including, area, aspect ratio, breadth, length, and perimeter, of each particle were determined during automated particle analysis using the Oxford Instruments Aztec® Micoanalysis Software suite. Table 2 describes the software parameters.

Table 2. Morphology parameters as defined by Oxford Instruments Aztec®1

Area (μm^2)	The area of the grain in μm^2.
Perimeter (μm)	The perimeter of the grain in μm.
Length (μm)	The longest linear dimension of the grain calculated from the maximum Feret diameter (μm).
Breadth (μm)	The shortest linear dimension of the grain calculated from the minimum Feret diameter (μm).
Aspect Ratio	The ratio of the major to minor axis of the elliptical fit.

[1]Aztec EBSD Data Analysis User Guide, Oxford Instruments NanoAnalysis, Part No. 51-1720-467

<u>Aim 3</u>: Characterize respirable dust particles from representative coal mine environments

Objective 3.1: Collect dust samples in representative mine environments

Task 3.1.1: Dust sampling

Respirable dust samples were obtained from 15 mines on this project (Mines 10-24 in Table 3). We also had samples available from another 10 mines that were obtained on other projects (Mines 1-8 were sampled on an earlier Alpha Foundation-funded project; Mine 9 was sampled on an unfunded project; Mine 25 was sampled on a CDC/NIOSH project). Table 3 provides a summary of key details for all 25 mines. In total, 16 mines are located in central Appalachia (i.e., the RPP/PMF hotspot region); of the others, five are located in northern Appalachia, two are in the Illinois basin, and two are in the western region.

Table 3. Summary of key details and samples collected per mine. (Taken from Sarver et al., 2021.)

Mine No.	MSHA District	Prod. method[1]	Diesel status[2]	Roof/floor strata[3]	Average strata height mined			Number of sample sets per location[4]					Total sets
					Coal (m)	Rock (m)	Rock/ Total	B	F	I	P	R	
1	4	CM	U	SN	1.2	0.3	0.20	3	1	1	1	1	7
2	4	CM	U	SN	1.1	0.6	0.36	2	1	1	1	1	6
3	4	CM	U	SH/SN	1.4	0.5	0.25	1	1	1	1	1	5
4	4	CM	U	SN	0.9	0.3	0.25	1	1	1	0	1	4
5	2	LW	N	SN/SH/SL	2.1	0.3	0.13	1	3	2	1	3	10
6	3	LW	D	SH	2.0	0.5	0.19	1	3	6	1	4	15
7	12	CM	D	SN/SH/SL	1.7	0.3	0.15	2	3	2	4	3	14
8	12	CM	U	SH	1.3	0.5	0.29	2	3	3	3	4	15
9	5	CM	D	SH	0.6	0.4	0.39	3	4	3	2	2	14
10	7	CM	D	SH/SN	1.1	0.8	0.42	2	2	1	1	1	7
11	5	CM	D	SH	0.8	0.5	0.40	1	1	0	1	1	4
12	5	CM	D	SH/SN	1.3	0.7	0.34	1	0	1	1	1	4
13	5	LW	D	SN/SH	1.6	0.3	0.13	1	1	2	0	2	6
14	5	CM	D	SH	0.8	0.9	0.52	1	1	1	1	0	4
15	4	CM	U	SH	1.0	0.9	0.48	1	1	1	1	1	5
16	3	CM	D	SH	2.1	0.0	0.00	1	1	0	1	1	4
17	3	LW	D	SH	1.6	0.5	0.24	1	0	2	1	2	6
18	3	CM	U	SH	0.8	0.9	0.54	1	1	1	1	1	5
19	8	CM	D	SH/LM	1.9	0.2	0.08	1	1	1	1	2	6
20	8	CM	D	SH/LM	1.8	0.1	0.04	1	2	1	1	1	6
21	12	CM	D	SH/SN	0.9	1.1	0.55	1	1	1	1	1	5
22	12	CM	D	SH	0.8	0.6	0.45	0	1	1	1	0	3
23	9	LW	D	SH	4.3	0.0	0.00	0	0	3	1	1	5
24	9	LW	D	SH/SN	1.8	0.5	0.20	0	1	3	0	2	6
25	12	CM	N	SH/SN	0.2	0.5	0.67	1	1	1	1	1	5
Total sets								30	35	40	28	38	171

[1]Primary production method: CM = continuous miner, LW = longwall
[2]Diesel status: D = diesel equipment in the mine, N = no diesel equipment in the mine, U = diesel equipment status unknown
[3]Roof/floor strata being mined/drilled: SN = sandstone, SH = shale, SL = slate, LM = limestone
[4]Sampling Location: B = bolter, F = feeder, I = intake, P = production, R = return
MSHA, Mine Safety and Health Administration

Briefly, the sampling protocol was to collect area dust samples in key locations of each mine: in the intake (I) and return (R) just outby of the production face, just downwind of active roof bolting (B) and coal cutting (P), and near the feeder breaker (F). In each location, several replicate samples were collected to allow for multiple analyses. Sampling was done using Mine Safety and Health Administration (MSHA) -approved air pumps and Dorr-Oliver cyclones to capture only the respirable-sized particles. The dust was collected onto polycarbonate filters.

Objective 3.2: Characterization of mine dust

Task 3.2.1: Determine the mass concentration of metals and trace elements
For dust samples obtained for this project (i.e., from Mines 10-24), mass concentrations of potentially bio accessible and total acid-soluble metals and trace elements were determined by sequential digestions in simulated lung fluid (SLF) and strong acid, followed by ICP-MS analysis. Details of the specific methods followed are available in the Appendix (Section 11.1). Samples from Mines 1-8 had been previously analyzed using similar methods on a separate Alpha Foundation project, and results are available in Sarver et al., 2019.[17] Mine 9 did not have a sample available for digestion.

Task 3.2.2: Determine the size and mineralogy distribution of dust particles

Dust samples from all 15 mines sampled on this project, plus the other 10 mines shown in Table 3, were analyzed by SEM-EDS using two automated routines to collect particle size and elemental data across the entire range between 100-10,000 nm. The elemental data was used to infer particle mineralogy. Details of the SEM-EDS analysis are also available in the Appendix (Section 11.1).

Aim 4: Analyze clinical, pathologic, mineralogic, and mine dust data from miners with RPP and PMF compared to historical referent cases from NCWAS to identify risk factors for severe disease and identify opportunities to design prevention strategies.

Objective 4.1: Statistical analysis of data from contemporary cases of RPP and PMF, historical referent cases, and current mine dust samples

Task 4.1.1: Integration of all study data

This study used individual and mine-level data to analyze differences in historical and contemporary RPP/PMF cases. Where possible, we linked data for epidemiologic analyses, including linking pathology findings to the mineralogy (*in situ* and digestion results) findings. Ultimately, we were unable to link mine-level data to the historical and contemporary cases which precluded an investigation of mine-specific predictors of RPP/PMF. We compiled a database of all individual data pertaining to miners in the registry and NCWAS including demographic, occupational exposure history, clinical, histopathological, and mineralogic data.

Task 4.1.2: Complete case characterization of contemporary RPP and PMF cases

Using the integrated data described in Task 4.1.1, we performed descriptive analyses to describe the occupational exposure, clinical, histopathological, and mineralogic characteristics of miners with severe disease. We examined regional differences in histopathology and mineral type, quantity, morphology, and the presence of metals.

Task 4.1.3: Case-control study of risk factors for severe disease comparing contemporary RPP and PMF cases to NCWAS historical referent cases

We identified risk factors for severe disease by comparing the demographic and occupational characteristics of contemporary (cases) and historical PMF cases (controls). Using data collected from NCWAS and the registry cases where available, we explored differences in longest job held and smoking characteristics between contemporary (born 1930+) and historic (born 1910-1929) cases of PMF. Jobs were classified into high risk designated occupations (DOs, including roof bolters, continuous miner or cutting machine operators, loaders), those not designated high risk, or unknown based on Mine Safety and Health Administration regulations (30CFR§70.208). We used Chi-Square or Fisher Exact tests in our analyses of categorical variables and t-tests for continuous variables.

We compared the relationship between pathologic and mineralogic features of historical and contemporary cases of severe disease by evaluating which particles were associated with PMF types and selected pathologic features (e.g. silica-type PMF, mineral dust associated alveolar proteinosis, immature silicotic nodules). We used SAS (version 9.4; SAS Institute, Cary, NC) for all analyses. Categorical variables were compared between historical and contemporary groups using Fisher's exact test. Continuous variables such as the weighted percentages of particles and particle concentration were examined across historical and contemporary status as well as PMF type using t-tests with pooled or Satterthwaite results as appropriate. ANOVA Tukey's pairwise comparison were used to compare mean differences in continuous variables across multiple groups. Levene's test was used to assess homoschedasticity; in cases of unequal distribution we used the Welch's test for ANOVA testing. A p-value ≤ 0.05 was considered significant. In those cases where multiple comparisons were employed we also provided a Bonferroni corrected p-value.

Task 4.1.4: Identification of severe disease risk factors and recommendations for preventive strategies

Pathologic, mineralogic, occupational, and dust sample findings were synthesized to arrive at final conclusions regarding risk factors for RPP/PMF among contemporary miners compared to their historical counterpoints.

5.0 Research Findings and Accomplishments

Aim 1: Detailed clinical characterization of contemporary cases of RPP and PMF as well as historical referent cases from NCWAS biorepository.

We screened 1,129 records from the clinics database and referrals and found 433 with possible PMF/RPP. There were 279 of these that either declined or were unavailable, leaving 154 who consented and were enrolled into our general registry, (Figure 4). Of these, we identified and received specimens from 16 contemporary registry cases, seven of which were excluded from analysis (Table 4). In total, we accessioned 95 contemporary and historical pathology specimens from NCWAS, of which 76 cases were included for final analysis (Table 5). Thus, we acquired 23 contemporary cases and 62 historical cases (85 total evaluable cases with confirmed PMF) for pathology and mineralogy analyses (Table 5).

Figure 4. Flow chart of cases accessioned into the study.

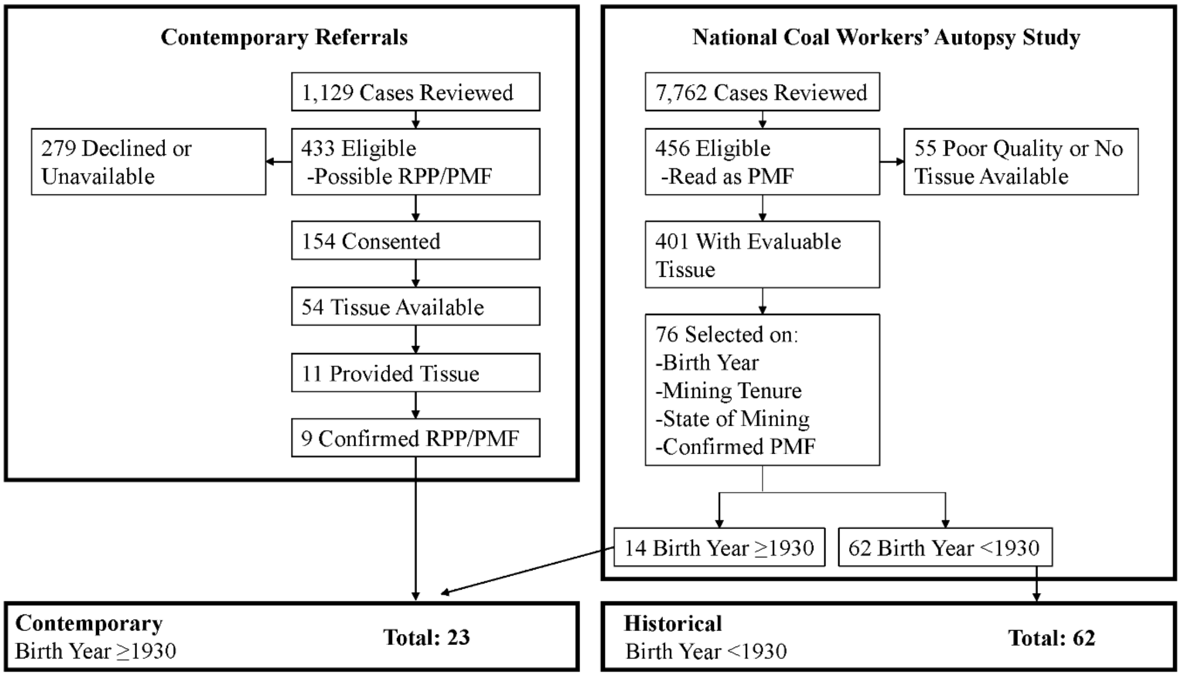


Table 4. Cases excluded and included from analyses.

Source	Excluded Cases*	Included Cases	Total
NCWAS	19	76	95
Registry	7	9	16
Total	24	85	111

*Reasons for exclusion were: non-PMF lesions after expert review, inadequate specimen, or poor quality specimen.

Table 5. Contemporary and historical comparison cases for pathology and mineralogy analyses.

Source	Contemporary*	Historical*	Total
NCWAS	14	62	76
Registry	9	0	9
Total	23	62	85

*Contemporary cases were those with a date of birth from 1930 onward; historical cases were those with a date of birth before 1930.

Objective 1.1: Select miners with contemporary RPP and PMF from the registry

Task 1.1.1: Develop registry of RPP/PMF and identify those with complete data sets and informed consent.

One-hundred fifty-six individuals consented to participate in the general registry. The majority of participants were recruited from West Virginia (n = 41), Colorado (n = 39), Kentucky (n = 32) and Virginia (n = 30). The remaining participants were recruited from Wyoming (n = 4), Tennessee (n = 3), Pennsylvania (n = 2), Arizona (n = 1), Illinois (n=1), North Carolina (n = 1), South Dakota (n = 1), and Texas (n = 1).

Task 1.1.2: Characterize occupational and health history of miners

One-hundred thirty-one medical history interviews and 62 occupational history interviews have been fully completed with an additional 32 partially completed occupational history interviews as part of the general registry. We completed 16 medical history interviews and nine occupational history interviews on those subjects from whom we have received pathology specimens.

We found that contemporary miners (those born after 1930, working predominantly with modern mining techniques) had worked significantly less time underground than historic miners and in different jobs. This work was presented at the American Thoracic Society meeting in 2021 and a manuscript is currently in progress to report and expand on these findings.

Task 1.1.3: Obtain lung tissue specimens from miners

Lung tissue was obtained from 16 subjects in the registry, of which seven cases were found to be non-PMF after expert review and were not included in the analyses.

Objective 1.2: Select historical referent cases from NIOSH National Coal Workers' Autopsy Study (NCWAS) database

Task 1.2.1: Scan NCWAS database and select referent cases

We identified 387 PMF cases in the NIOSH NCWAS database for the historical comparison period 1971 – 1996. Of the 387 potential PMF cases, 332 had both histologic slides and corresponding blocks of the PMF lesion. A secondary objective of our review of the NCWAS cases was to determine if any PMF/RPP cases had been accessioned into the program after 1996, the year that NIOSH stopped reviewing cases for entry into their database because of a lack of a pathologist to evaluate human tissue. We found that 69 additional post-1996 PMF cases were identified for a total of 401 cases. The addition of the post-1996 cases was very important as it augmented the number of contemporary cases of PMF/RPP for use in this study.

From the 401 cases in the NIOSH NCWAS database, we selected cases by birth year and state in which coal mining was performed to capture potential differences in geography and exposures experienced related to time periods in which mining was performed.

Task 1.2.2: Abstract occupational and health history data on referent cases

The study team obtained occupational history data for all NCWAS cases. The data included total years of coal mine employment, years of surface and/or underground employment, the state in which the miner worked as a surface and/or underground miner and a description of the principal (i.e., longest-held) job and job last held by the miner. Examples of job descriptions within the NCWAS data set include "roof bolters and helpers"; "continuous mining machine operators and helpers"; "electricians and helpers, wiremen, mechanics, general repairmen"; and "superintendent, assistant foremen, section bosses, grade foremen." The NCWAS data set also included detailed smoking history data, including ever/never smoker status; smoking status at time of death; typical number of cigarettes per day; and total pack-years smoked. The study team obtained copies of the original data collection forms completed by miners' next of kin to confirm and, as needed, correct occupational and smoking history data. Because of changes in the form used to collect occupational history data and the staff responsible for NCWAS data entry over time, several discrepancies were identified between the NCWAS database and paper forms. For this reason, two study team members familiar with mining terminology (Leonard Go and Lauren Zell-Baran) independently reviewed each of the original principal and last job descriptions and classified them into the available list of job codes. Any disagreements were reviewed, and consensus reached.

Task 1.2.3: Obtain lung tissue from NCWAS historical comparison cases

We obtained lung tissue from NIOSH for 62 NCWAS historical comparison cases and 14 contemporary NCWAS cases (Table 5).

<u>Aim 2</u>: Characterize the lung tissue histology in coal miners with contemporary cases of RPP/PMF and compare these findings to lung tissue from historical cases from miners with PMF in the NCWAS biorepository.

Objective 2.1: Characterize pathologic and histologic features of dust-related lung disease

Task 2.1.1: Selection of tissue blocks for characterization

After our selection process we classified tissue from 85 cases for pathology, (Table 5).

Task 2.1.2: Classification and grading via bright field and polarized light microscopy

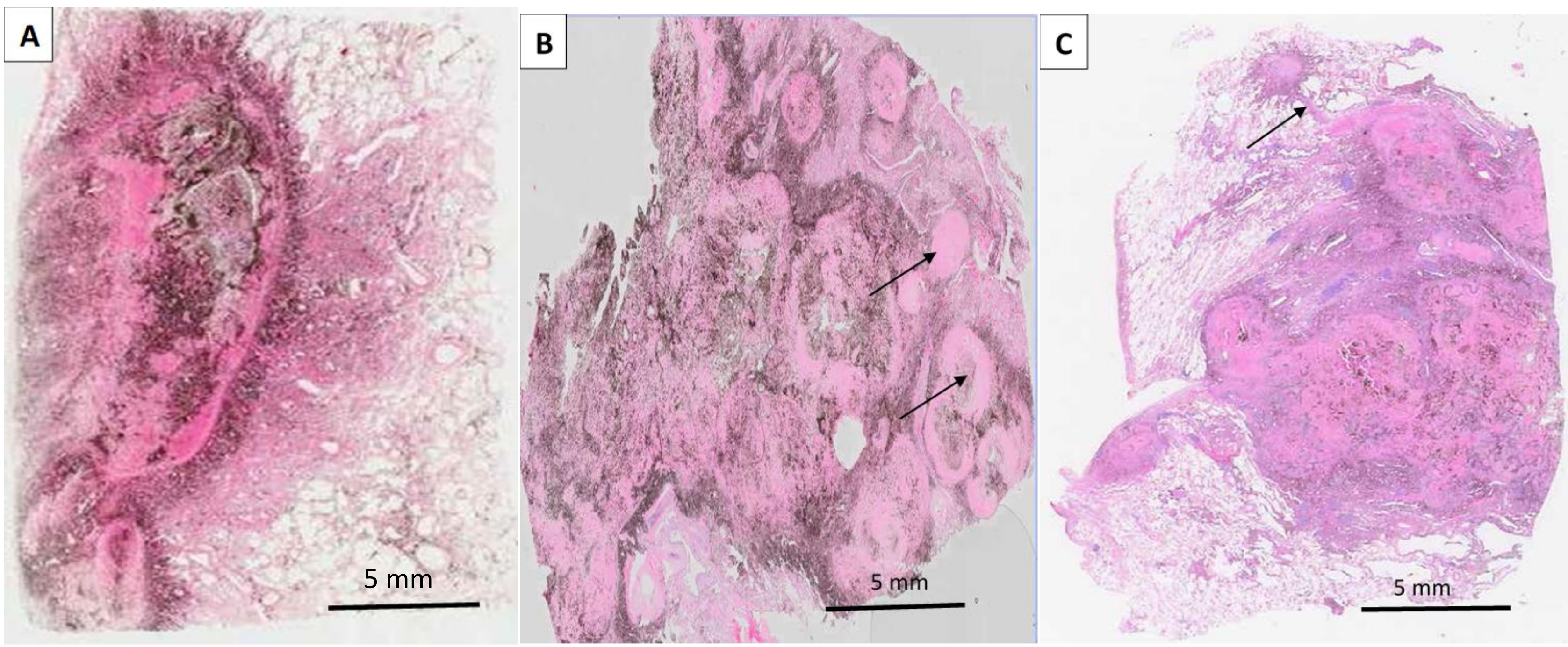
Our lead study pathologist (FG) initially classified the cases of PMF from the NCWAS for the period 1971 to 1996 (n=387 cases) of which 332 had histologic slides and paraffin blocks. These preliminary findings showed a significant increase in the proportion of silica-type PMF identified in cases accessioned after 1990 (40%, versus 24% of cases accessioned prior to 1990). This was followed by a systematic review of these cases by all study pathologists who independently re-classified these cases of PMF. Photomicrographs of examples of the types of PMF used in this classification are shown below, (Figure 5). These data showed a significantly higher prevalence of silica-type PMF (57% vs. 18%, $p < 0.001$) among contemporary miners compared to their historical counterparts. In contrast, coal miners born before 1930 had a significantly higher prevalence of both coal-type PMF (50% vs. 17%, $p < 0.001$) and mixed-type PMF (33% vs. 26%, $p < 0.001$) (Table 6).

Figure 5. Representative examples of coal-, mixed- and silica-types of PMF, (hematoxylin and eosin stains).

A: Coal-type PMF lesion (≤25% silicotic nodules). This lesion consists of one large nodule fused to two smaller nodules below. There is substantial collagen with varying orientation surrounded by a rim of coal dust-laden histiocytes with fibrotic extensions into the adjacent parenchyma. There is prominent central necrosis with large quantities of dust. Mature or immature silicotic nodules are not seen, with the possible exception of the small collagenized nodule at bottom left.

B: Mixed-type PMF (>25% and ≤75% silicotic nodules). This PMF lesion is composed of fused nodules, some with features of coal dust nodules, others showing features of mature silicotic nodules (arrows). Some of the nodules show central necrosis, and there is extensive necrosis with cavitation on the left side of the lesion. Black coal dust pigment is prominent in all areas.

C: Silica-type PMF (>75% silicotic nodules). This lesion is composed almost entirely of mature silicotic nodules. Silicotic nodules are also seen in the adjacent parenchyma with bridging fibrosis (arrow) to the PMF lesion. Black coal mine dust is markedly less apparent than in the other PMF types.



Study pathologists graded all 401 cases of PMF. This included 332 cases accessioned into the NCWAS prior to 1997 and 69 cases accessioned in 1997 or later. Analysis of this data showed good agreement among pathologists for pathologic features of PMF.[14]

Specimens had varying amounts of lung parenchymal tissue surrounding the PMF lesions. Six specimens had so little non-PMF parenchyma that neither coal macules and nodules nor mature and immature silicotic nodules could be evaluated. Despite these differences in the quantity of lung parenchymal tissue areas surrounding PMF lesions, we found a trend toward an increased prevalence of both mature silicotic nodules ($p = 0.17$) and immature silicotic nodules ($p = 0.11$) in contemporary miners. See Figure 6 for an example of an immature silicotic nodule. Compared to contemporary miners, miners born before 1930 had a significantly higher prevalence of coal macules (93% vs 60%, $p < 0.01$), with a trend towards increased coal nodules in surrounding lung parenchyma (78% vs. 58%, $p = 0.08$) (Table 6).

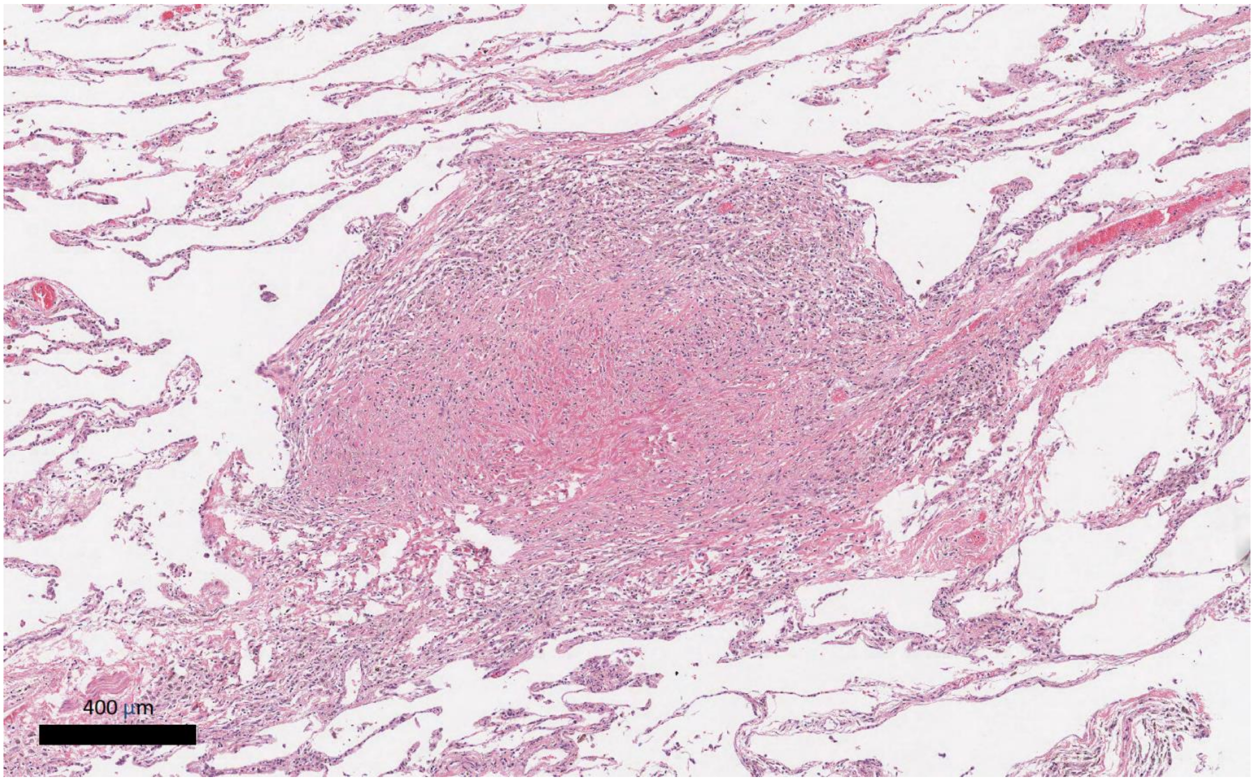


Figure 6. Immature silicotic nodule, (hematoxylin and eosin stain). The nodule is composed of central collagen bundles lacking the characteristic central whorling of a mature silicotic nodule. The periphery is composed of fibrohistiocytic cells with prominent lymphocytes. The latter extend into the adjacent lung interstitium. Note: These nodules should not be confused with granulomas which differ from immature (and mature) silicotic nodules in that they are composed of activated histiocytes and do not have the central collagen bundles.

Findings consistent with mineral dust-related alveolar proteinosis (MDAP) were significantly more prevalent in contemporary miners compared to their historical counterparts (70% vs. 37%, $p < 0.01$) (Table 6). To confirm these visual assessment findings, 10 specimens with MDAP on H&E stains were stained with PAS-D, and all were positive. MDAP in these specimens was confined to focal involvement of alveolar spaces adjacent to the PMF lesions (Figure 7).

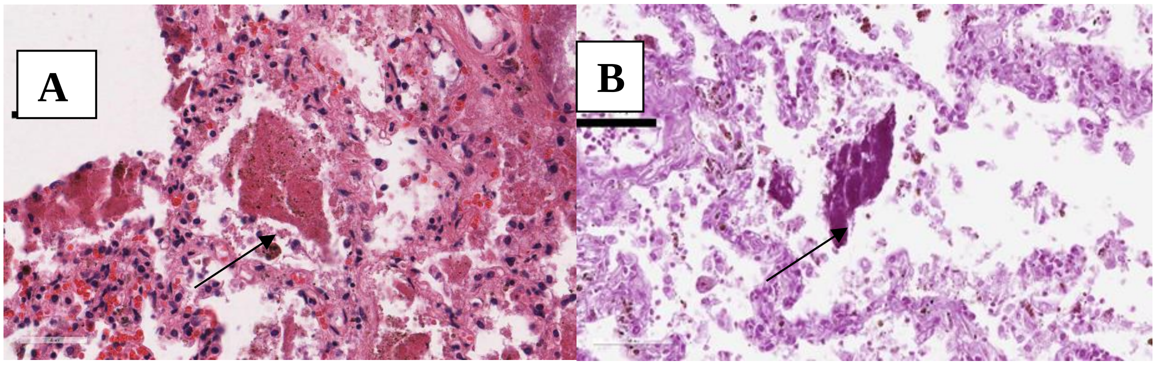


Figure 7. Example of mineral-dust alveolar proteinosis (MDAP).
This feature was characterized by the finding of scattered alveoli containing dark pink, finely granular, lipo-proteinaceous material (A), that stained with PAS (B). Characteristic cracking artefact (arrows) was also seen.

Analysis by mining region showed that the significantly increased prevalence of silica-type PMF, MDAP, and the trends towards increased profusion of mature and immature silicotic nodules were largely seen in miners who worked in the central Appalachian states of Virginia, West Virginia, and Kentucky. Similar differences between historical and contemporary miners were seen outside of central Appalachia, however the numbers were small and were not statistically significant (Table 6).

Table 6. Pathologic findings in historical compared to contemporary coal miners with PMF, including all cases and findings by US geographic region.

Finding	**All Regions** Historical n=62	Contemporary n=23	P value	**Central Appalachia†** Historical n=39	Contemporary n=17	P value	**Rest of the US** Historical n=23	Contemporary n=6	P value
PMF Type									
Silica	11 (18)	13 (57)	<0.001	5 (13)	10 (59)	<0.001	6 (26)	3 (50)	0.28
Mixed	20 (33)	6 (26)	<0.001	10 (26)	4 (24)	<0.001	10 (44)	2 (33)	0.28
Coal	31 (50)	4 (17)	<0.001	24 (66)	3 (18)	<0.001	7 (30)	1 (17)	0.28
Surrounding lung parenchyma*									
Silicotic Nodules	20 (33)	10 (52)	0.17	12 (32)	7 (50)	0.33	8 (35)	3 (60)	0.35
Immature Silicotic Nodules	11 (18)	7 (37)	0.11	6 (16)	4 (29)	0.43	5 (22)	3 (60)	0.12
Coal Macules	55 (93)	12 (60)	<.01	34 (92)	10 (67)	0.02	22 (96)	2 (40)	0.01
Coal Nodules	47 (78)	11 (58)	0.08	33 (89)	8 (57)	0.01	14 (61)	3 (60)	1.00
MDAP**	22 (37)	16 (70)	<0.01	14 (36)	12 (71)	0.02	8 (35)	4 (67)	0.20

All values are presented as n (%).
* Six cases had only PMF lesions without evaluable parenchyma. Total evaluated = 79.
**MDAP: mineral dust-related alveolar proteinosis
†Note: Central Appalachia refers to the states of Virginia, West Virginia and Kentucky.

Objective 2.2: Characterize lung dust *in situ*

Task 2.2.1: Perform *in situ* mineralogic analysis

In situ mineralogic analysis was performed on lung tissues from 17/23 (74%) contemporary miners and 33/62 (53%) historical comparisons. There was a greater total concentration of mineral particles in lung specimens of contemporary compared to historical miners, although this difference was not statistically significant (180 x 10^8 vs. 149 X 10^8 particles/cm^3, Table 7). Most notably, the percentage (26.1% vs. 17.8%, p<0.01) and concentration (47.3 x 10^8 vs. 25.8 X 10^8 particles/cm^3, p=0.03) of silica particles was significantly greater in specimens from contemporary miners compared to their historical counterparts**.**

Compared to historical miners, there was a lower percentage of aluminum silicate (SiAl and SiAlK) particles in contemporary miners, likely due to the increased percentage of silica. However, we found no significant difference in the concentration of aluminum silicate particles between groups (Table 7). There were no other significant changes noted in the percentages or concentrations of the other most common particles including titanium (Ti) or less commonly found metals.

Table 7. In situ *lung mineralogy findings in historical versus contemporary coal miners with PMF: Percentages and concentrations by particle type*

	Historical Mean (SD) n=33	**Contemporary Mean (SD)** n=17	**P value***
Total particle concentration†	149 (77)	180 (156)	0.43
Particle Type:			
Silica (Si)			
% of Particles	**17.8 (10.0)**	**26.1 (10.0)**	**0.007****
Particle concentration†	**25.8 (19.7)**	**47.3 (37.0)**	**0.036**
Aluminum Silicates (SiAl and SiAlK)			
% of Particles	**74.6 (9.9)**	**66.2 (9.9)**	**0.006****
Particle concentration†	112 (60.5)	120 (118)	0.78
Titanium (Ti)			
% of Particles	5.8 (3.0)	6.2 (2.3)	0.68
Particle concentration†	8.8 (7.4)	11.4 (10.7(	0.32

* P < 0.05 in bold.

** P < Bonferroni correction for testing 6 comparisons value of 0.0085.

†Particle concentrations are particles X 10^8 per cm^3 of tissue.

Note: Data for particles comprising < 5% not shown, therefore percentages in Table 7 do not total 100%.

Objective 2.3: Characterize digested lung dust

Task 2.3.1: Determine the relative mass concentration of total lung dust and specific constituents

In total, 112 samples were analyzed including 14 blank sample preparations. Consensus readings ruled out some samples as not containing PMF and several more were not analyzed by USGS due to timing of sample receipt. This left a total of 65 samples to evaluate, including 24 coal type PMF, 24 mixed-type PMF, and 17 silicotic PMF. Of these 65 samples, 15 were categorized as contemporary miners and 50 were categorized as historical miners. There were over 60 different particle types identified through the analysis including carbonaceous particles, silicate minerals, and metals and metal oxides (Table 8). To aid in data analysis and to have a more direct comparison to the mine dust analysis, the particles were classified using the scheme described in Sarver et al., 2021.[16] As seen in Table 8, most of the particles classified by USGS fall into the appropriate Sarver et al., 2021 category.[16] Particle classification disagreements between the USGS and Sarver et al. (2021) schemes are likely due to EDS spectra that represented a mixture of phases. For all data results presented, the classification scheme of Sarver et al., 2021 was used.[16]

Table 8. Comparison of particle classification used by USGS to the classification scheme presented in Sarver el al., 2021[16].

USGS \ Sarver et al.	C	MC	ASK	ASO	SLO	S	M	CB	O	NoVT
Ag_phase	NA	1	1	NA	NA	NA	NA	NA	NA	NA
Al_oxide	14	17	NA	NA	NA	NA	NA	NA	1921	NA
Al_oxide_mix	NA	NA	7	NA	198	NA	4	NA	165	NA
Al_P_phase	NA	1	NA	NA	8	NA	NA	NA	285	NA
Al_S_phase	6	9	NA	NA	NA	NA	NA	NA	50	NA
Als_Al_gt_Si	1	23	3995	NA	437	NA	NA	NA	8	NA
Als_Al_lt_Si	1	38	17945	NA	193	90	NA	NA	2470	NA
Au_phase	NA	1	NA	NA	NA	NA	NA	NA	NA	NA
Ba_phase	NA	2	1	NA	1	1	NA	NA	1	NA
C_Cl	660	1384	NA	NA	NA	NA	NA	NA	NA	54
C_S	NA	6	NA	NA	NA	NA	NA	NA	NA	NA
Ca_Al_oxide	NA	1	NA	NA	NA	NA	NA	NA	5	NA
Ca_Al_silicate	NA	NA	NA	NA	43	1	NA	NA	2	NA
Ca_Mg_phase	NA	1	NA	NA	NA	NA	NA	3	2	NA
Ca_Mg_silicate	NA	NA	NA	NA	13	3	NA	NA	NA	NA
Ca_P_phase	NA	NA	NA	NA	NA	NA	NA	NA	39	NA
Ca_P_phase_mix	NA	NA	NA	NA	18	NA	NA	2	11	NA
Ca_phase	1	9	NA	NA	NA	NA	NA	NA	285	NA
Ca_phase_mix	NA	1	NA	NA	20	NA	NA	1	20	NA
Ca_S	NA	1	NA	NA	NA	NA	NA	NA	3	NA
Ca_silicate	NA	1	NA	NA	13	2	NA	NA	1	NA
Ca_Ti_oxide	NA	NA	NA	NA	NA	NA	NA	NA	3	NA
CaCl2	NA	2	NA	NA	NA	NA	NA	NA	29	NA
Carbonaceous	5541	33333	NA	NA	NA	NA	NA	NA	8	NA
Carbonaceous_No_O	4256	NA	NA	NA	NA	NA	NA	NA	NA	NA
Cr_phase	181	789	2	NA	3	56	NA	NA	16	61
Cu_phase	5	38	1	NA	1	5	NA	NA	6	7
Fe_Cr	NA	NA	NA	NA	41	NA	NA	NA	233	NA
Fe_Cu_Mn_Zn_Ni_Cr	NA	6	NA	NA	12	1	1	NA	38	NA
Fe_Ni	NA	NA	NA	NA	6	NA	NA	NA	1751	NA
Fe_Ni_Cr	NA	NA	NA	NA	NA	NA	NA	NA	128	NA
Fe_oxide	NA	NA	NA	NA	72	3	NA	NA	4877	NA
Fe_oxide_mix	NA	NA	NA	NA	1097	NA	12	NA	237	NA
Fe_silicate	NA	NA	NA	NA	12	NA	NA	NA	NA	NA
Fe_Ti_oxide	NA	NA	NA	NA	1	NA	NA	NA	47	NA
Fe_Ti_oxide_mix	NA	NA	NA	NA	17	NA	18	NA	1	NA
Feldspar_K	NA	NA	NA	NA	2	265	NA	NA	295	NA
Feldspar_Na	NA	NA	13	NA	4	116	2	NA	171	NA

Sarver et al. / USGS	C	MC	ASK	ASO	SLO	S	M	CB	O	NoVT
Feldspar_Plag	NA	NA	1	NA	2	2	NA	NA	18	NA
K_Als	NA	NA	14634	NA	1720	19	3	NA	2052	NA
Mg_Fe_Als	NA	NA	4	NA	1474	11	NA	1	54	NA
Mg_Fe_phase	NA	NA	NA	NA	NA	NA	NA	NA	1	NA
Mg_Fe_Silicate	NA	NA	NA	NA	6	1	NA	NA	NA	NA
Mg_phase	NA	NA	NA	NA	1	NA	NA	NA	6	NA
Mg_silicate	NA	1	NA	NA	3	65	NA	NA	NA	NA
Na_Al_Silicate	NA	NA	206	NA	2	28	NA	NA	53	NA
Na_K_Ca_S_Cl	11	54	NA	NA	1	NA	NA	NA	242	NA
NaCl	NA	112	NA	NA	NA	1	NA	NA	1	NA
Ni_Cr	11	32	NA	NA	NA	NA	NA	NA	NA	NA
Ni_phase	37	857	NA	NA	NA	1	NA	NA	1	12
P_phase	2	111	NA	NA	NA	NA	NA	NA	7	NA
Pb_phase	NA	NA	NA	NA	NA	1	NA	NA	NA	1
REE	6	14	2	NA	4	1	NA	NA	16	NA
S_Cl	219	196	NA	NA	NA	NA	NA	NA	NA	5
S_phase	529	1351	NA	NA	NA	NA	NA	NA	3	348
Silica	61	138	NA	NA	11	10639	NA	NA	34	NA
Silica_mix	3	12	11	NA	61	1271	NA	NA	44	NA
Sn_phase	NA	944	1	NA	2	11	NA	NA	3	NA
Ti_Fe_oxide	NA	NA	NA	NA	NA	NA	NA	NA	67	NA
Ti_Fe_oxide_mix	NA	NA	NA	NA	26	NA	11	NA	1	NA
Ti_oxide	NA	4	NA	NA	2	1	NA	NA	1674	NA
Ti_oxide_mix	NA	NA	NA	NA	899	NA	6	NA	213	NA
U_phase	NA	1	NA	NA	NA	NA	NA	NA	NA	NA
Zn_phase	1	7	3	NA	8	6	NA	NA	15	2
Zr_phase	2	11	9	NA	10	68	NA	NA	19	NA

A comparison between the particle concentrations in the historical and contemporary miners was conducted for each particle type (Figure 8). The median value of carbonaceous (C) and metal (M) particles particles in the contemporary group was statistically significantly less than the historical group (p=0.03 and 0.04 respectively). The number of carbonate particles was also lower, however this was not statistically significant (p=0.023) because a large number of samples had no carbonate particles present. The median values for mixed carbonaceous (MC), silicates (ASK, SLO, and S), and other (O) phases are similar with no statistically significant differences between the two miner groups. While the result for the aluminosilicate concentrations is similar to the *in situ* results, the silica concentrations reported for the *in situ* work were significantly higher in the contemporary group compared to the historical group (Table 7). This may reflect that the *in situ* work was conducted within the areas of PMF and the digestion results reflect areas both inside and outside the PMF areas.

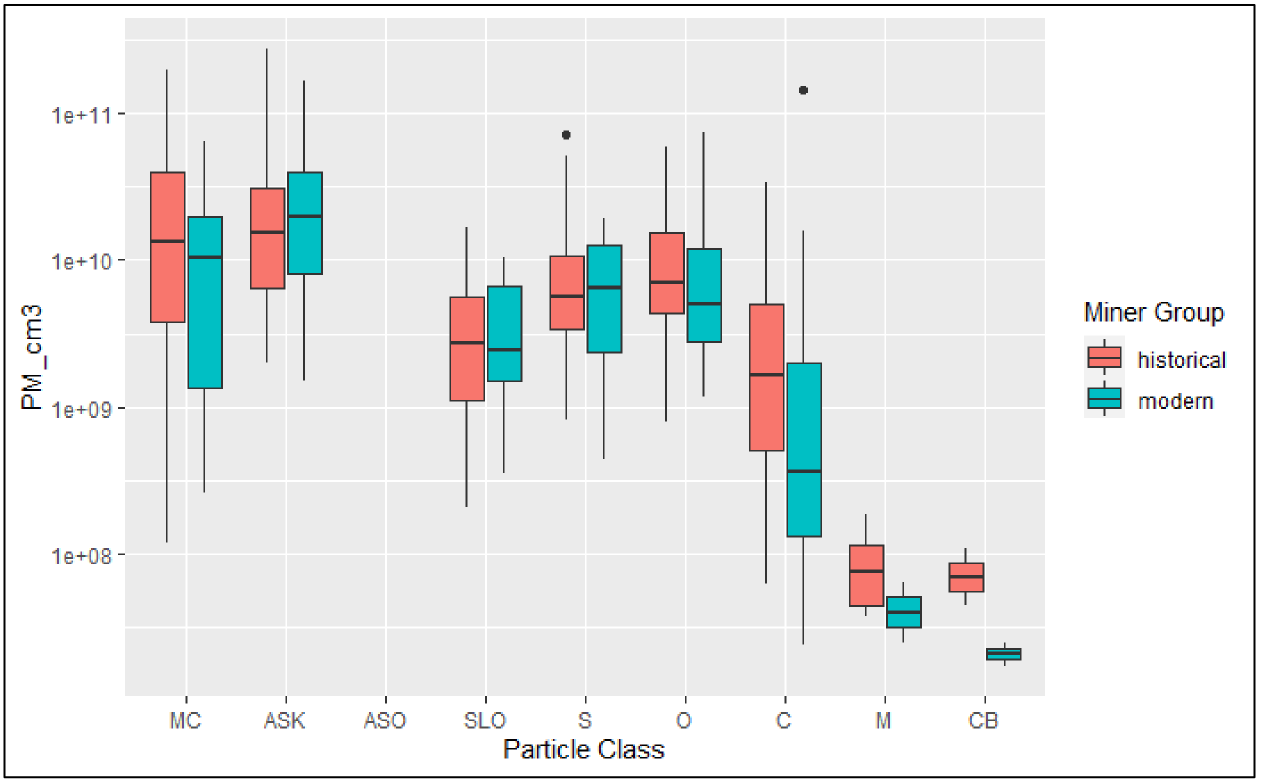


Figure 8. Particulate matter per cubic centimeter of tissue for mixed carbonaceous (MC), aluminosilicates (ASK), other aluminosilicates (ASO), other silicates (SLO), silica (S), other (O), carbonaceous particles (C), metals (M), and carbonate (CB) and in historical and contemporary (modern) miner groups. The data presented by dots above and below the box and whiskers represent outlying data points.

Results from the ICP-MS analysis were challenging due to the high concentration of elements of interest in the bleach reagent (Si, Ca, K, Mg, Sn, Sr), the ratio of the volume of bleach to volume of tissue, and loss of silicon from complexation with hydrofluoric acid. The final concentration of elements in several samples was less than zero after correcting for the concentration in the reagents and process blanks. Only the results from the particle fraction were evaluated as the bleach digestate results were near or below detection for all elements.

A comparison between historical and contemporary miners was carried out for the rock forming elements (Figure 9). As with the particulate concentrations in the tissue, the elemental concentrations are also similar between the historical and contemporary miners.

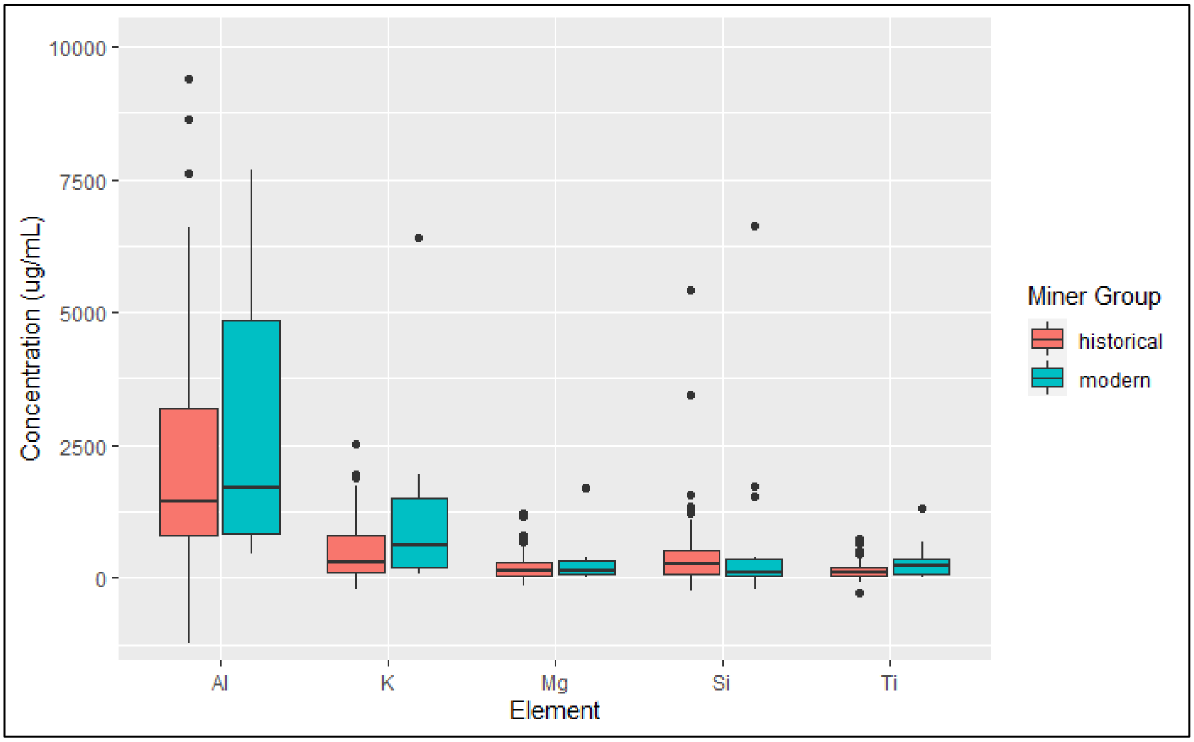


Figure 9. Concentration of rock forming elements in historical and contemporary (modern) miners. The data presented by dots above and below the box and whiskers represent outlying data points.

Task 2.3.2: Determine the size distributions of lung dust particles

The size distribution of the length (Figure 10), breadth (Figure 11), and area (Figure 12) for each particle class was similar for historical and contemporary miners. The carbonaceous (C) particles are slightly larger than the other particle types for both historic and contemporary miners.

The typical equant morphology of the particulate matter was shown in Figure 3. This morphology is supported by the data in Figure 13, which shows the 0.5 cumulative sum of the aspect ratio for all particle classes is ~1.25 and the 0.9 cumulative sum is ~2.

Please note that these data are considered provisional, are subject to change and have not been approved through the U.S. Geological Survey data release process. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

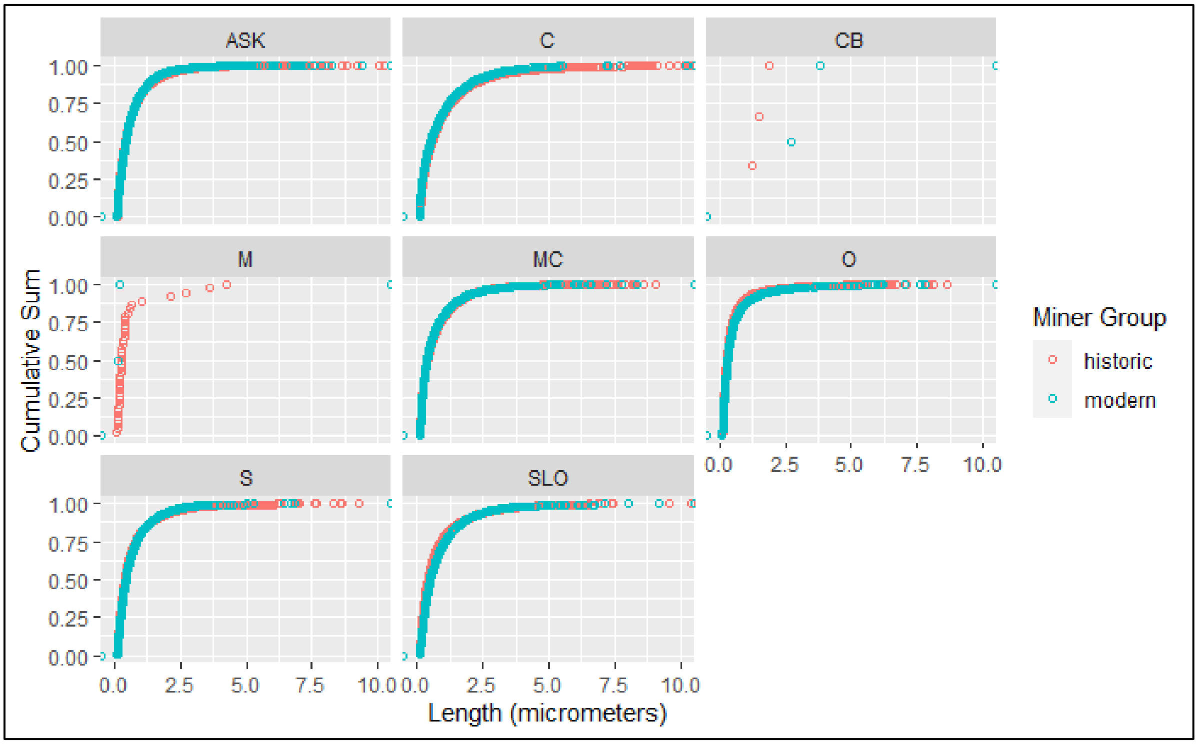


Figure 10. Length distribution for each particle type and historical and contemporary (modern) miners.

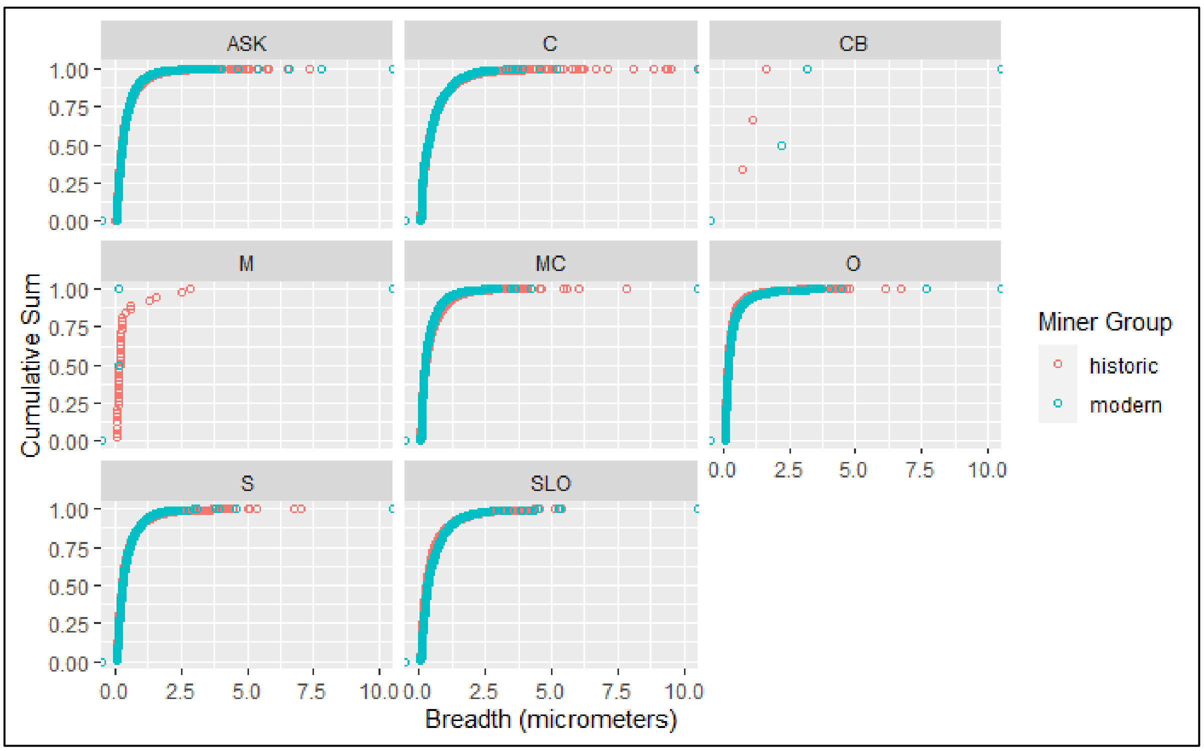


Figure 11. Breadth distribution for each particle type and historical and contemporary (modern) miners.

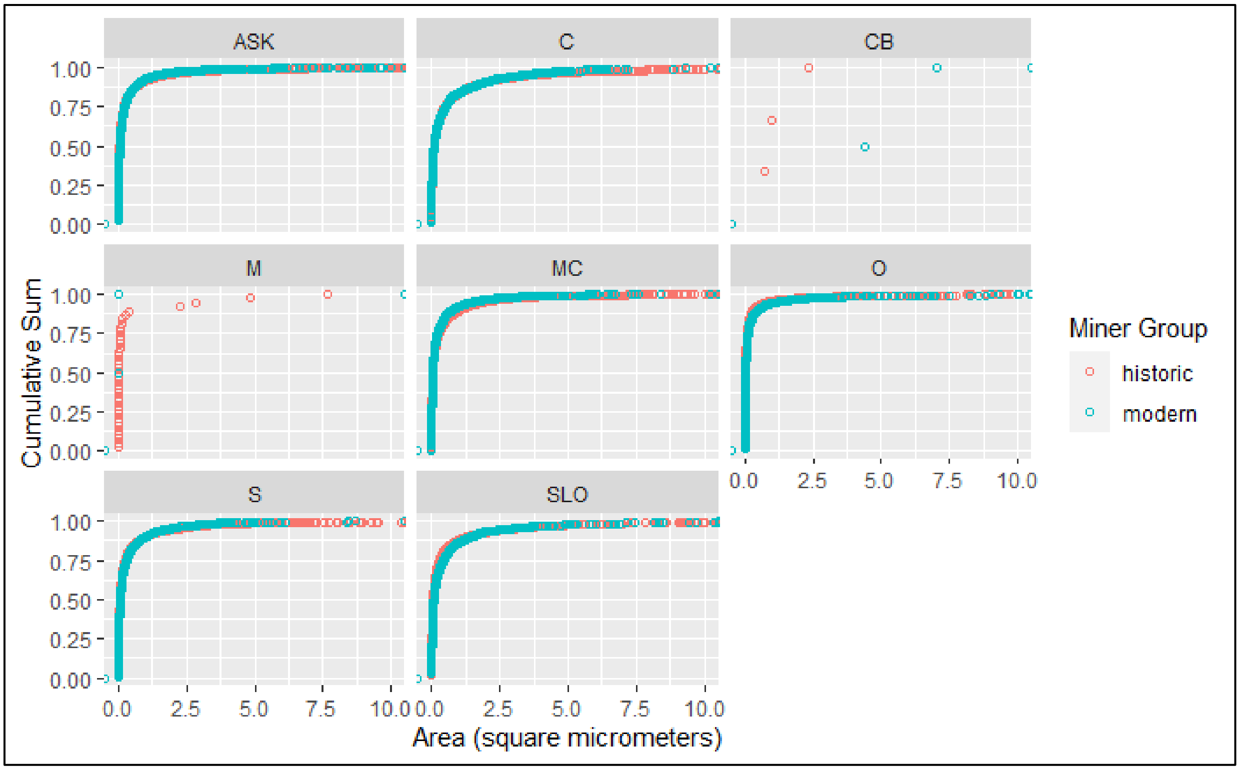


Figure 12. Area distribution for each particle type and historical and contemporary (modern) miners.

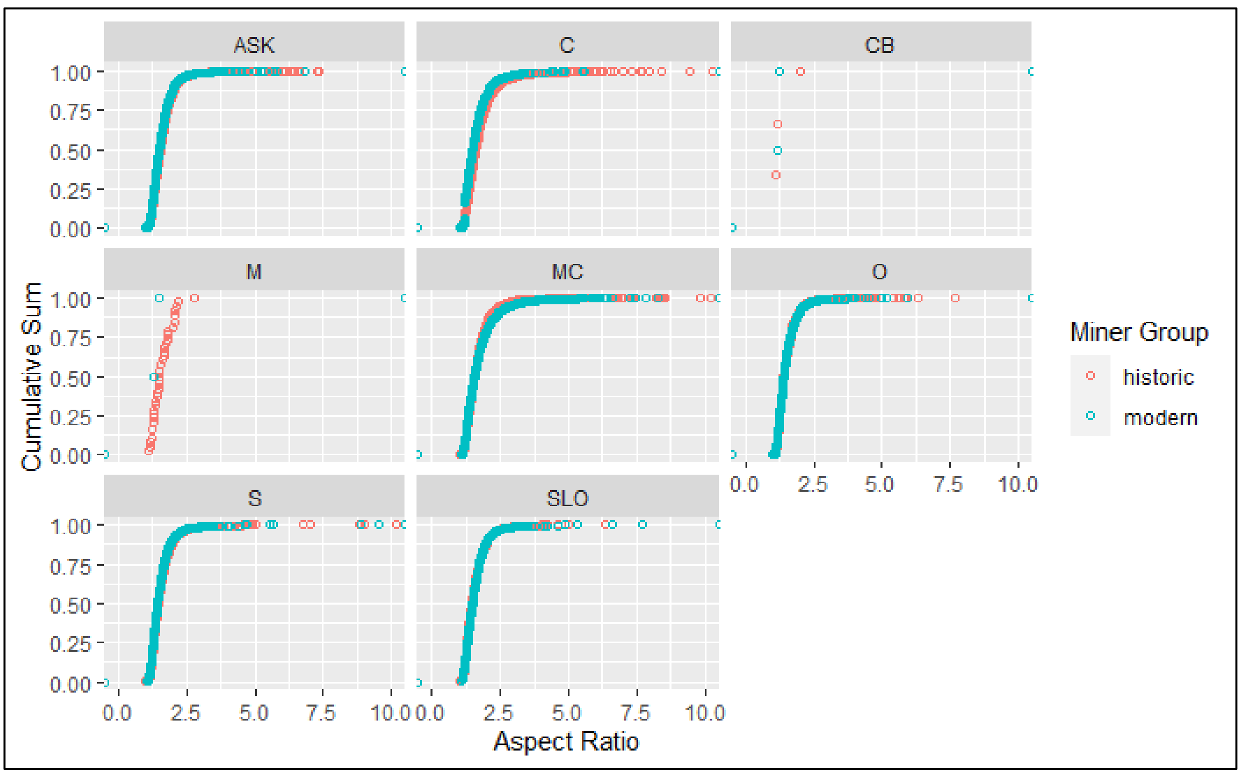


Figure 13. Aspect ratio of historical and contemporary (modern) miners broken out by particle class.

<u>Aim 3</u>: Characterize respirable dust particles from representative coal mine environments.

Objective 3.1: Collect dust samples in representative mine environments

Task 3.1.1: Dust sampling

The sampling campaign for this project was highly successful. We worked with a total of seven industry partners, including the three that expressed support during the project proposal phase, to sample Mines 10-24 (Table 3). We were also able to expand the inventory of samples available for the project work by leveraging those collected during separate efforts in Mines 1-9 and 25.

Objective 3.2: Characterization of mine dust

Task 3.2.1: Determine the mass concentration of metals and trace elements

Results of this analysis are tabulated in the Appendix (Section 11.1).

Task 3.2.2: Determine the size and mineralogy distribution of dust particles

Results of the SEM-EDS analysis are also tabulated in the Appendix (Section 11.1) and have been presented elsewhere[16,18]. Following is a summary of key findings:

- Across all mine regions, there appear to be three main sources of respirable dust: (1) the coal seam itself, to which relatively large (i.e., >400 nm) carbonaceous particles are attributed; (2) the rock strata that surrounds the coal seam, to which silica, silicates and other minerals are primarily attributed; and (3) the rock dust products that are applied in mines to mitigate explosibility hazards, to which carbonate particles are primarily attributed. Moreover, in mines operating diesel equipment, there can be relatively high number concentrations of diesel particulate matter (DPM), which are observed as relatively small (i.e., <400 nm) carbonaceous particles.

- Across all mine regions, the relative size distribution of particles can generally be summarized as diesel particulates < coal < minerals (Figure 14). Of the primary mineral classes analyzed here, silica appears to be somewhat finer than other minerals including silicates coal dust generally appears to be finer than mineral particles. With respect to specific sampling locations, dust nearby to cutting or drilling activities (i.e., near the production face or the roof bolter, or in the return airways) was found to be generally finer than dust in the intake or near the feeder breaker.

- Across all mine regions, dust in the production, return and roof bolter locations generally also had higher content (number %) of rock-strata sourced minerals (Figure 15).

- In central Appalachian mines, the silica and/or silicate content (number %) near the production face and/or in the return was also significantly higher than in mines outside of this region (Figure 16). This is consistent with the relatively large amount of rock being cut along with the coal in many of the central Appalachian mines (Table 3). On the other hand, carbonate content (i.e., attributed to rock dust application) was generally higher in the mines located outside of central Appalachia. These same trends were observed when the particle-level data was used to estimate constituent content on a mass % basis (Figure 17).

- Overall, the majority of particles reside in the submicron range. Comparing Figure 14 to Figure 17, the difference between respirable dust data reported on a number% versus mass % becomes very clear. Across all mineralogy classes considered by the SEM-EDX analysis, about 25-75% of particles have a projected area diameter (PAD) <1000nm; however, these particles are estimated to account for only about 2-12% of the dust mass.

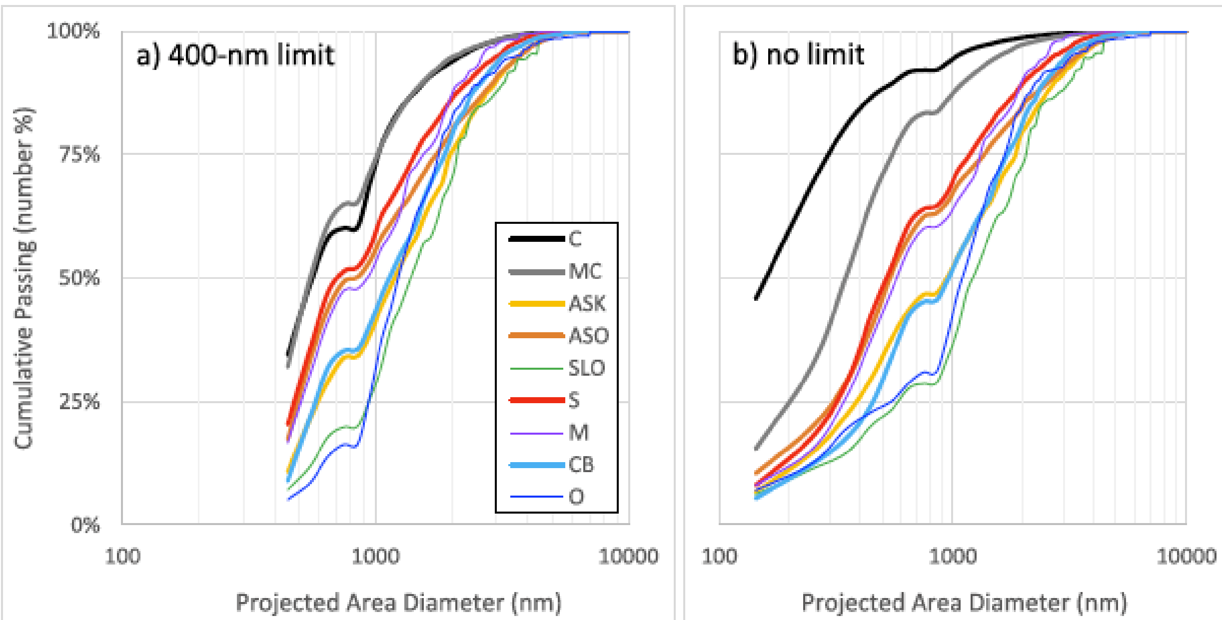


Figure 14. [Reproduced from Sarver et al., 2021.[16]] Average cumulative particle size distributions (number %) across all mines and sampling locations by mineralogy class (C=carbonaceous, MC=mixed carbonaceous, ASK=kaolinite-like aluminosilicates, ASO=other aluminosilicates, SLO=other silicates, S=silica, M=heavy minerals, CB=carbonates, O=other). Results are shown a) with and b) without a 400-nm threshold established to minimize the influence of diesel particulate matter (DPM) on the C class. A total of 171 samples were analyzed for this project; however, since the number of samples from each location varied by mine, sample results were first averaged by location in each mine and then by mine, yielding n=112. Data were computed using number percentage of particles in the 100-nm wide size bins, with points plotted at the bin mid-size.

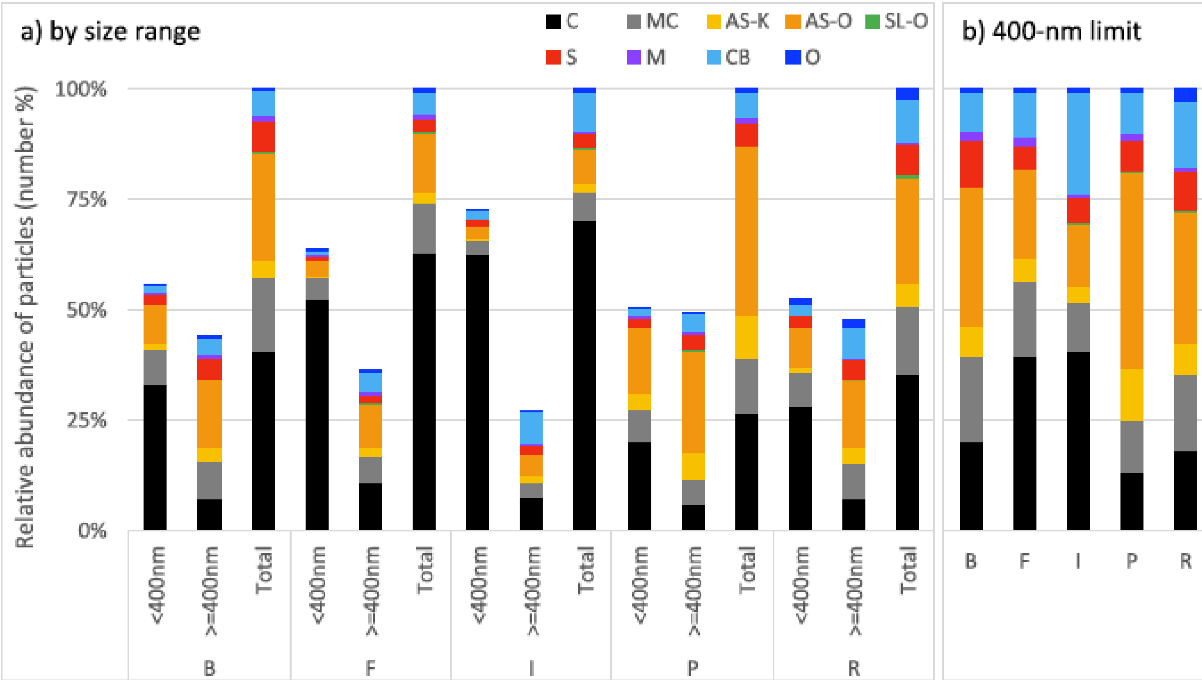


Figure 15. [Reproduced from Sarver et al., 2021.[16]] Average relative abundance of particles (number %) in each mineralogy class across all mines by sampling location. (C=carbonaceous, MC=mixed carbonaceous, ASK=kaolinite-like aluminosilicates, ASO=other aluminosilicates, SLO=other silicates, S=silica, M=heavy minerals, CB=carbonates, O=other; B=roof bolter, F=feeder breaker, I=intake, P=production, R=return.) In a), results are shown for the <400 nm and ≥400 nm size ranges, as well as for the total range covered by the SEM analysis. Since the number of samples from each location varied by mine, sample results were first averaged by location in each mine and then by mine, yielding n=22 for the B, F, and P locations and n=23 for the I and R locations. In b), results only for the ≥400 nm size range are normalized to compare dust constituents with minimal influence of diesel particulate matter (DPM). Data were computed using number percentage of particles in each mineralogy class.

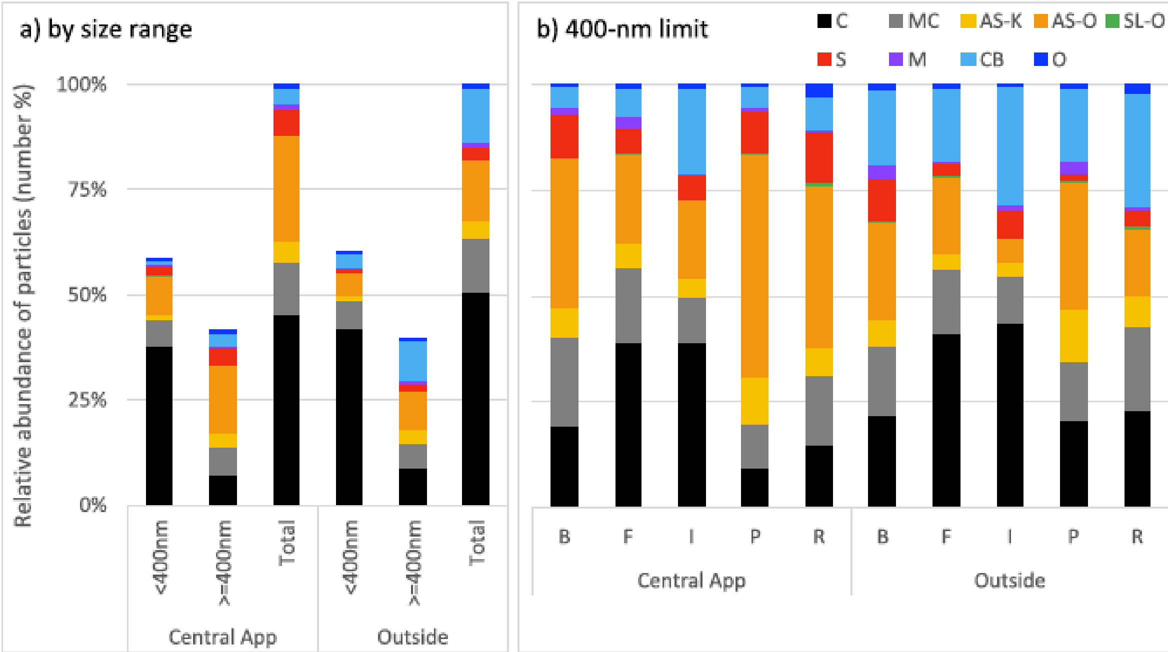


Figure 16. [Reproduced from Sarver et al., 2021.[16]] Average relative abundance of particles (number %) in each mineralogy class for mines in and outside of the central Appalachian region. (C=carbonaceous, MC=mixed carbonaceous, ASK=kaolinite-like aluminosilicates, ASO=other aluminosilicates, SLO=other silicates, S=silica, M=heavy minerals, CB=carbonates, O=other; B=roof bolter, F=feeder breaker, I=intake, P=production, R=return.) In a), results are averaged across all sampling locations for the <400 nm and ≥400 nm size ranges, as well as for the total range covered by the SEM analysis. Since the number of samples from each location varied by mine, sample results were first averaged by location in each mine and then across the region, yielding n=73 for central Appalachia and n=39 for outside central Appalachia. In b), results only for the ≥400 nm size range are normalized and separated by sampling location (n=15 for B, F, and I and n=14 for P and R locations in central Appalachia; n=7 for B and F, n=8 for I and P, and n=9 for R in mines outside central Appalachia). Data were computed using number percentage of particles in each mineralogy class.

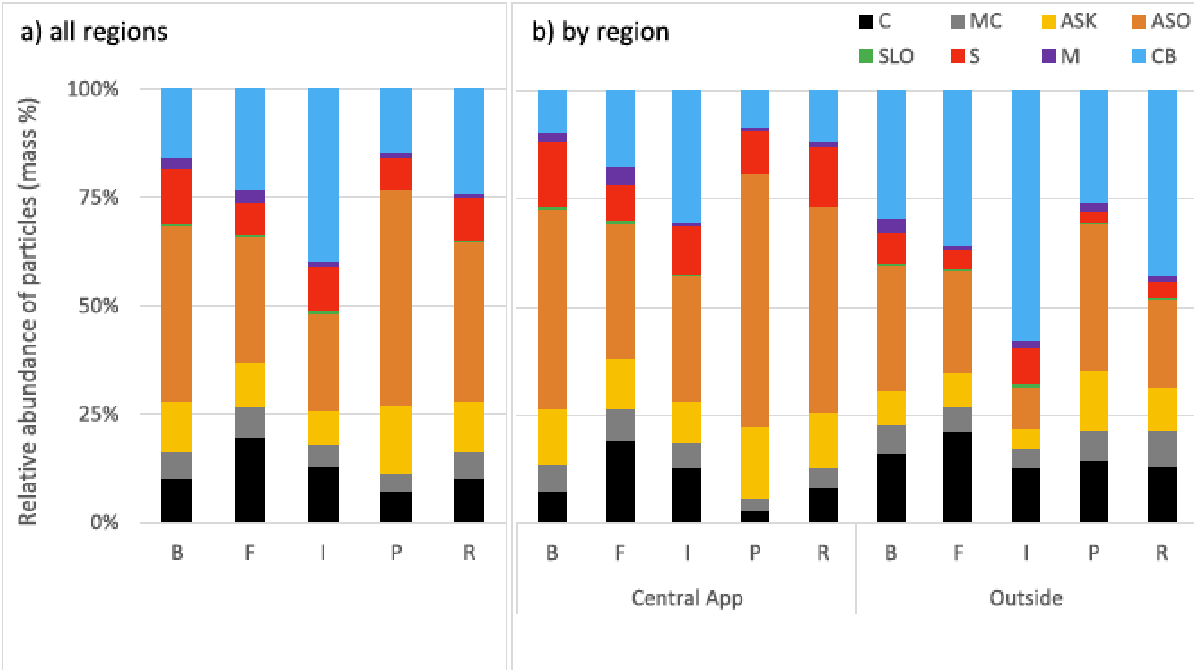


Figure 17. [Reproduced from Sarver et al., 2021.[16]] Average relative abundance of particles (mass %) in each mineralogy class by sampling location. (C=carbonaceous, MC=mixed carbonaceous, ASK=kaolinite-like aluminosilicates, ASO=other aluminosilicates, SLO=other silicates, S=silica, M=heavy minerals, CB=carbonates, O=other; B=roof bolter, F=feeder breaker, I=intake, P=production, R=return.) In a), results are shown across all mines (n values same as in Figure 15). In b), results are separated by mine region (n values same as in Figure 16). Data were computed using mass percentage of particles ≥400 nm in each mineralogy class.

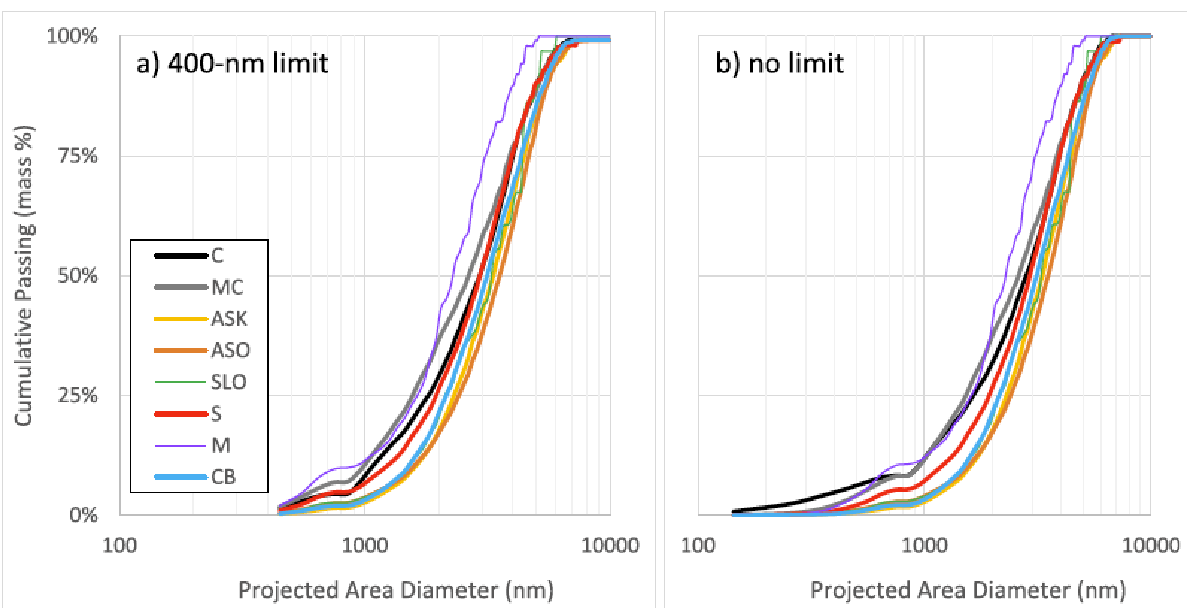


Figure 18. [Reproduced from Sarver et al., 2021.[16]] Average cumulative particle size distributions (mass %) across all mines and sampling locations by mineralogy class (C=carbonaceous, MC=mixed carbonaceous, ASK=kaolinite-like aluminosilicates, ASO=other aluminosilicates, SLO=other silicates, S=silica, M=heavy minerals, CB=carbonates, O=other.) Results are shown a) with and b) without the 400 nm threshold established to minimize the influence of diesel particulate matter (DPM) on the C class; n=112 for both plots. Data were computed using estimated mass percentage of particles in 100-nm wide size bins, with points plotted at the bin mid-size.

Aim 4: Analyze clinical, pathologic, mineralogic, and mine dust data from miners with RPP and PMF compared to historical referent cases from NCWAS to identify risk factors for severe disease and identify opportunities to design prevention strategies.

Objective 4.1: Statistical analysis of data from contemporary cases of RPP and PMF, historical referent cases, and current mine dust samples

Task 4.1.1: Integration of all study data
This study used individual and mine-level data to address the different aims outlined above. Where it was possible, we linked data for our epidemiologic analyses. Ultimately, we were unable to link mine- and individual-level data which precluded an investigation of mine-specific predictors of RPP/PMF. We compiled a database of all individual data pertaining to miners in the registry and NCWAS including demographic, occupational exposure history, clinical, histopathological, and mineralogic data.

Task 4.1.2: Complete case characterization of integrated data from contemporary and historical RPP and PMF cases

Task 4.1.3: Case-control study of risk factors for severe disease comparing contemporary RPP and PMF cases to NCWAS historical referent cases

The results of these two tasks are presented together to improve readability and interpretation of our findings.

Our final analyses of integrated pathology and mineralogy data included 85 miners (62 historical and 23 contemporary cases). Contemporary miners were significantly younger at the time their lung tissue was obtained (61 vs 65 years old, p=0.03) and had significantly fewer years of underground mining (30 vs 35 years, p=0.03) as well as a trend toward fewer total years mining (31 vs. 36, p=0.14). We observed no difference between groups for work in the central Appalachian states of Kentucky, Virginia, and West Virginia; race; or smoking status and total pack-years (Table 9).

Table 9. Comparison of demographic characteristics of historical and contemporary RPP/PMF cases used in final analyses of integrated pathology and mineralogy data (n = 85).

	Historical			**Contemporary**			
	n=62	**SD**	**Range**	**n=23**	**SD**	**Range**	**P value**
Age (Mean Years)	65.4	5.7	55 - 82	61.1	3.7	48 - 79	**0.03**
Birth Year (Mean Year)	1919	4.9	1910 – 1928	1942	11	1930 – 1961	**<0.0001**
White Race Yes n (%)	53 (87)			21 (91)			0.20
Smoker Yes; n (%)	50 (81)			17 (74)			0.55
Mean Pack-Years Smoking	20.1	19.0	0 – 96	18.6	18.4	0 – 60	0.76
[Work in] Central Appalachia* (Yes)	39 (63)			17 (74)			0.44
Mean Years of Coal Mining	35.8	10.0	10-50	31.4	2.2	10 – 42	0.14
Mean Years Worked Underground	34.9	8.9	3 – 50	30.2	8.7	10 – 42	**0.03**
Mean Years Worked at the Surface	2.6	10	0 – 45	0.8	1.3	0 – 3	0.44

*Central Appalachia refers to the states of Kentucky, Virginia, and West Virginia

Linked pathology and in situ *mineralogy findings*

Linkage of the pathology and *in situ* mineralogy data revealed that the concentration of silica particles was more than 50% greater when pathologic features associated with silica exposure were present in the sections analyzed, including MDAP, mature silicotic nodules and immature silicotic nodules ($p<0.05$, Table 10). One-way analysis of variance showed nearly double the percentage of silica particles in silica-type PMF compared to mixed- or coal-type PMF (29.6% vs. 16.9% and 16.0%, respectively, $p<0.01$, Table 11). Also, the concentration of silica particles was 70% higher in silica-type PMF compared to mixed or coal-type PMF (42.4 X 10^8 vs. 27.2 x 10^8 and 29.6 X 10^8 particles/cm^3, respectively, $p=0.28$, Table 11, however this was not statistically significant.

Other particles: The presence of MDAP or mature silicotic nodules was associated with significantly lower percentages, but not lower concentrations, of aluminum silicates compared to silica particles (Table 10). The percentage of aluminum silicates in miners with silica-type PMF was significantly reduced compared to miners with coal- and mixed-type PMF ($p<0.01$, Table 11). While the concentration of aluminum silicate particles was also reduced in silica-type PMF compared to coal- and mixed-type PMF, it was not statistically significant ($p=0.07$, Table 11). There were no other significant differences noted in the percentages or concentrations of other particles including titanium (Ti) or less commonly found metals.

Table 10. In Situ Lung Mineralogy-Pathology Correlations in Coal Miners with PMF:

Pathology Finding	**MDAP Absent Mean (SD)**	**MDAP Present Mean (SD)**	**P value**	**Sil Nod Absent Mean (SD)**	**Sil Nod Present Mean (SD)**	**P value**	**Immature Sil Nod Absent Mean (SD)**	**Immature Sil Nod Present Mean (SD)**	**P value***
Type of Particle	n=26	n=24		n=30	n=16		n=34	n=12	
Silica (Si)									
% of particles (SD)	**17.1 (7.3)**	**24.5 (12.4)**	**0.016**	**16.9 (8.0)**	**25.8 (11.6)**	**0.004**	17.8 (7.9)	26.0 (13.8)	0.071
Particle concentration (SD) †	**24.6 (20.3)**	**42.3 (3.3)**	**0.030**	**24.4 (22.4)**	**44.2 (31.6)**	**0.017**	**26.2 (23.8)**	**45.8 (32.4)**	**0.031**
Aluminum Silicates (SiAl and SiAlK)									
% of particles (SD)	**75.4 (7.9)**	**67.8 (11.9)**	**0.011**	**74.8 (8.4)**	**67.5 (12.4)**	**0.022**	74.2 (8.0)	66.7 (14.5)	0.11
Particle concentration (SD) †	116.8 (96.5)	112.0 (67.9)	0.84	110.6 (68.3)	105.7 (60.5)	0.81	108.0 (68.7)	112.0 (56.4)	0.85
Titanium (Ti)									
% of particles (SD)	5.70 (3.1)	6.16 (2.80)	0.58	6.2 (3.2)	5.8 (2.9)	0.69	6.1 (3.4)	5.89 (1.8)	0.76
Particle concentration (SD) †	9.3 (9.6)	10.2 (7.7)	0.72	9.4 (7.8)	9.7 (8.6)	0.89	9.3 (8.6)	10.1 (6.1)	0.75

* P < 0.05 in bold.

** P < Bonferroni correction for testing 18 comparisons value of 0.0028.

†Particle concentrations are particles X 10^8 per cm^3 of tissue.

Table 11. Lung Mineralogy-Pathology Correlations by PMF Type: Percentages and Concentrations by Particle Type

Pathology Finding	**Type of PMF**			
	Silica-type Mean (SD)	**Mixed-type Mean (SD)**	**Coal-type Mean (SD)**	**P value**
Type of Particle	n=16	n=13	n=21	
Silica (Si)				
% of particles	**29.6 (12.5)**	**16.9 (5.5)**	**16.0 (6.8)**	**<0.001****
Particle concentration†	42.4 (33.6)	27.2 (20.0)	29.6 (28.0)	0.28
Aluminum Silicates (SiAl and SiAlK)				
% of particles	62.3 (11.2)	76.8 (6.7)	75.8 (7.1)	**<0.001****
Particle concentration†	75.6 (39.7)	137.6 (118.5)	129.8 (74.5)	0.07
Titanium (Ti)				
% of particles	6.2 (2.9)	5.0 (2.2)	6.2 (3.4)	0.44
Particle concentration†	7.7 (5.6)	8.4 (8.7)	12.1 (10.2)	0.25

Linked pathology and digested mineralogy findings

Integrating the pathology and digested mineralogy data revealed that the total particles of silica (S), other silicates (SLO), and aluminosilicates (ASK) per cm^3 of tissue increase slightly as the PMF type moves from coal- to mixed- to silica-type. Conversely, the total amount of carbonaceous (C) particles decreases from coal to mixed-type to silica-type PMF (Figure 19). However, the statistical significance of these findings has not been evaluated.

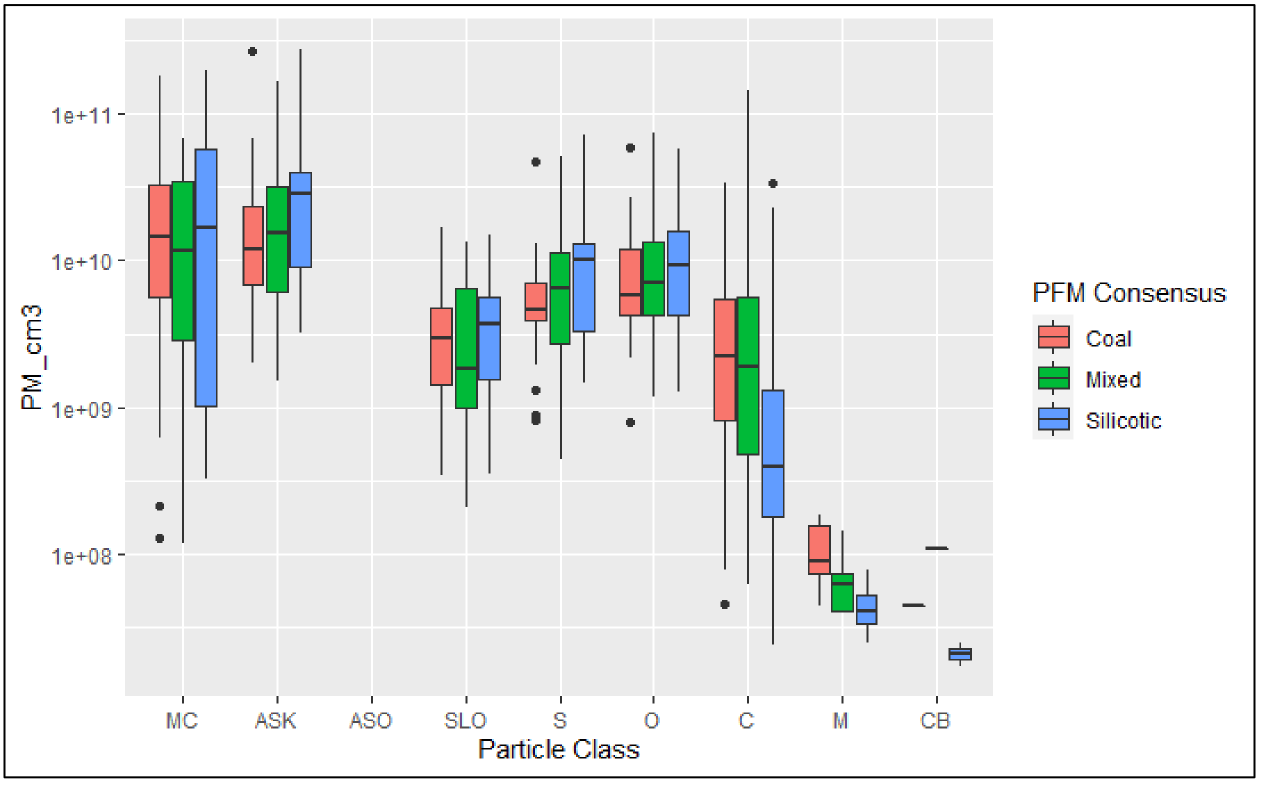


Figure 19. Particulate matter per cubic centimeter of tissue for mixed carbonaceous (MC), aluminosilicates (ASK), other aluminosilicates (ASO), other silicates (SLO), silica (S), other (O), carbonaceous particles (C), metals (M), and carbonate (CB) and in coal, mixed, and silica-type PMF samples.

ICP-MS results of the particulate fraction of the rock forming elements shows the median concentration of Al, K, Mg, and Ti is similar in all PMF types (Figure 20). The silicon median is higher in the silica-type PMF than coal- and mixed-type PMF (Figure 20). It was expected the silicon concentration should be greater than or equal to the aluminum concentration considering most of the particles identified included aluminosilicates and silica. As mentioned earlier, there was a loss of silica due to complexation with hydrofluoric acid.

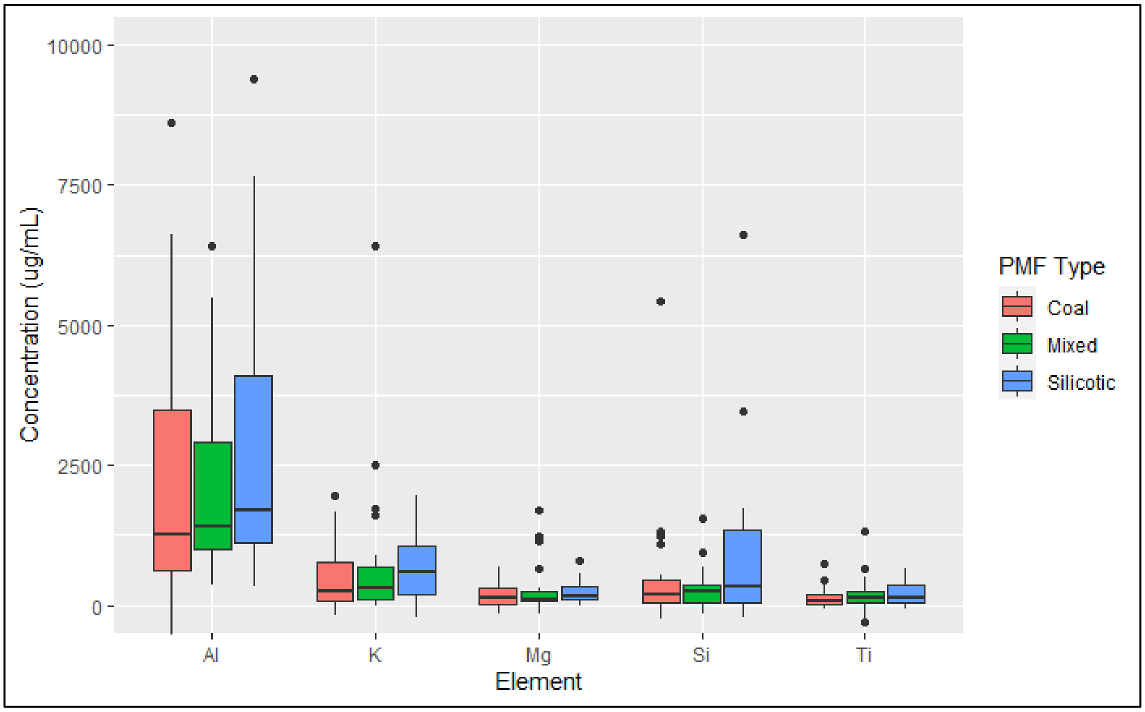


Figure 20. Concentration of rock forming elements by PMF type.

In addition to the rock forming elements, heavy metals (As, Cd, Cu, Mo, V) known to occur in coal deposits[19] were evaluated. The median values for As, Cd, and Mo are all near zero while the median values for Cu and V are higher (Figure 21). Cu is significantly higher in the silica-type PMF which may be from metal from grinding equipment. A positive V median value is likely due to the presence of both coal particles and aluminosilicates which are known to contain V[20].

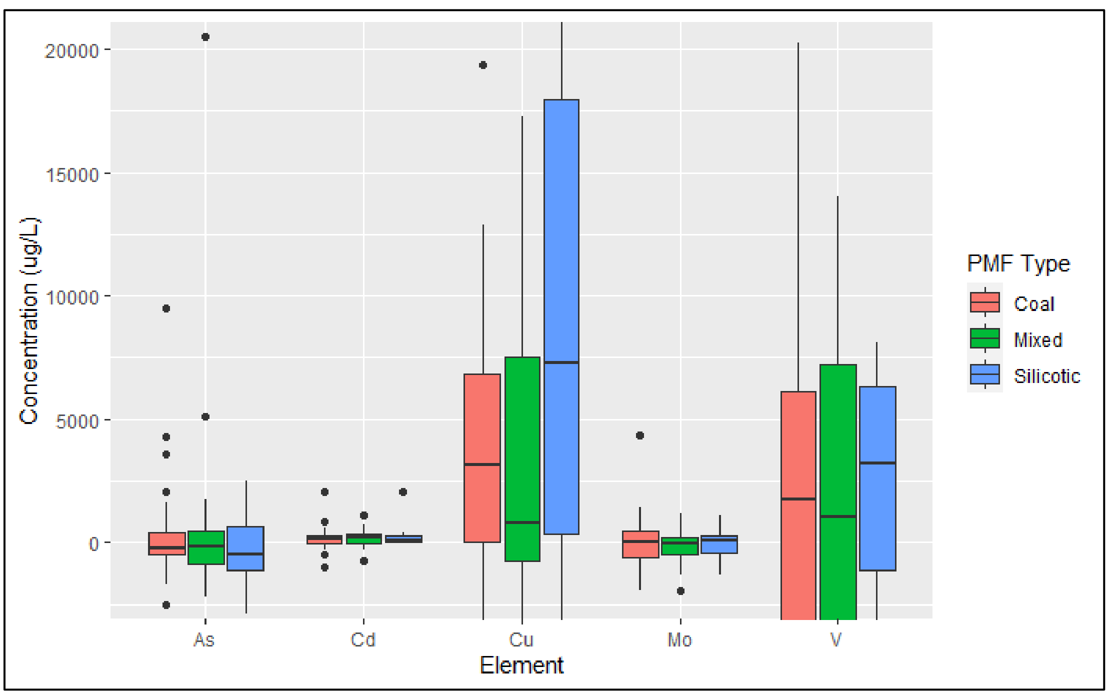


Figure 21. Concentration of heavy elements in the various PMF types.

Occupational risk factors for severe disease

Our analysis of demographic and occupational risk factors identified important differences between contemporary and historical RPP/PMF cases. All cases were male, with mean age 64.4 years at time of tissue collection. Comparing 60 historical and 24 contemporary cases, mean underground mining tenure was significantly shorter among contemporary compared to historical miners (p=0.03), with contemporary miners working on average 5.1 years less. Smoking histories were similar between groups (p=0.27), with 71% of contemporary miners reporting ever smoking compared to 82% of historic miners. Mean smoking pack-years did not differ between groups (22.6±18.2 contemporary vs. 23.4±18.8 historic, p=0.88). There were no differences between groups in states (mainly in Appalachia) where mining occurred. While not statistically significant, contemporary miners were more likely to work in high-risk DOs for most of their tenure (62% contemporary vs. 47% historic, p=0.39). In addition to high-dust exposure jobs such as roof bolters, continuous miner operators, cutting machine operators, and loaders, some underground coal miners with PMF reported longest mining tenure in non-DOs as electricians and foremen. Our findings indicate that contemporary miners are developing PMF after significantly shorter underground mining tenure than historic miners. This work was presented at the American Thoracic Society 2021 annual meeting.

Task 4.1.4: Identification of severe disease risk factors and recommendations for preventive strategies.

Our findings from our occupational exposure analysis indicate that contemporary miners are developing PMF after significantly shorter underground mining tenure than historic miners. While PMF risk is likely linked to high-dust exposure job duties, rates of PMF in contemporary miners are not explained by differences in the proportion of high-risk designated occupations (DO). Other factors such as increased silica concentrations in underground coal mines may be causally important. Current regulatory approaches monitoring total dust concentrations only without a specific permissible exposure limit for respirable crystalline silica in DOs may not be adequately protective.

Our data clearly shows an increased proportion of pathologic features consistent with substantial exposure to respirable crystalline silica in contemporary miners. These pathologic findings have been associated with acute and subacute silicosis.[21] They include the presence of foci of MDAP in association with PMF, a finding that has heretofore received little attention in the published literature.[22,23] We also developed a classification of PMF lesions into silica-, mixed-, and coal-type PMF that proved to be a useful method for characterizing lesions to better understand causal exposures. The increased proportion of mature and immature silicotic nodules seen in lung tissue adjacent to PMF lesions also points strongly toward a silica-driven etiology of disease among younger contemporary miners with significantly fewer years of mining tenure.

Our findings were further informed by results of *in situ* analysis of mineral particles in a subset of these miners. *In situ* findings also showed a significant increase in the percentage and concentration of silica particles in PMF lesions in lungs of contemporary coal miners compared to historical miners. There was a corresponding decrease in the percentage of aluminum silicate particles relative to silica particles in contemporary miners. This may reflect changes in geologic and/or mining conditions, including differences in silica content or dust generation from rock strata within or surrounding more contemporary coal mining seams. Of note, we found a significant correlation between the concentration and percentage of silica particles and the presence of lung pathologic lesions including PMF, MDAP, and both mature and immature silicotic nodules.

The mineralogy findings in these cases demonstrate some of the highest concentrations of silica particles reported with this *in situ* method.[24] For comparison, the total concentration of inorganic particles in the lungs of persons with no known dust exposures is in the range of 0.1 – 0.2 x 10^8 total particles/cm^3 tissue. The concentration of silica particles in PMF lesions reported in sandblasters was 1000 times higher — up to 146 x 10^8 silica particles/cm^3 tissue.[25,26] Thus, not only do our reported findings confirm the role of exceedingly high silica particles in the development of PMF, but they also provide evidence of the intense exposure to silica experienced by these coal miners, nearly one-third of the concentration seen in sandblasters with PMF.[25]

We divided our subjects based on birth year before and after 1930 in order to segregate miners likely to have worked mainly with historical mining methods from those who worked mainly with modern methods. Mechanized coal extraction devices such as continuous miner machines, longwall shears, and other advanced engineering technologies were introduced in the US in the 1950's, the age when a miner born in or after 1930 would have begun their career. These more efficient coal cutting devices,[7,8] along with improved and more cost-effective methods for separating silica-rich overburden rock from the coal being mined, may be driving increased exposure to respirable crystalline silica, and therefore account for the later surge in severe forms of CWP in contemporary miners.[27,28]

Based on the integrated pathologic and mineralogic findings in lung tissues from historical PMF and contemporary RPP/PMF cases, our study demonstrates that exposure to crystalline silica appears causal in the unexpected surge in severe disease in contemporary miners. Our findings underscore the importance of controlling workplace silica exposure in order to prevent the disabling and untreatable adverse health effects afflicting US coal miners.

6.0 Publication Record and Dissemination Efforts

We have made considerable efforts to disseminate our research findings to all relevant stakeholders, including federal agencies, the scientific community, and the general public. The results of these efforts are presented below.

6.1 Published and planned manuscripts

1. Pathology and mineralogy findings of contemporary versus historical miners (submitted and under revision with the Annals of the American Thoracic Society).
2. Occupational history differences in historical versus contemporary miners with PMF (in progress).
3. Characteristic pathologic features of PMF in the contemporary period (planned).
4. Historical trends in PMF type (in progress).
5. Go LHT, Cohen RA. Coal Workers' Pneumoconiosis and Other Mining-Related Lung Disease. *Clin Chest Med*. 2020;41(4):687-696. doi:10.1016/j.ccm.2020.08.002
6. Go LHT, Cohen RA. The Pneumoconioses. In V.Courtney Broaddus Joel Ernst Talmadge E King, Jr Stephen Lazarus Kathleen F. Sarmiento Lynn M. Schnapp Renee Stapleton Michael B. Gotway, eds. Murray & Nadel's Textbook of Respiratory Medicine. Elsevier. May 17, 2021. **ISBN:** 9780323655873

6.2 Peer reviewed presentations at national and international scientific conferences.

1. Cohen RA, Orandle M, Hubbs AF, et al. Pathologic type of progressive massive fibrosis in the National Coal Workers' Autopsy Study (NCWAS) 1971-1996. American Thoracic Society Conference, Dallas TX; May 2019. Poster presentation.
2. Go L, Lowers HA, Sanyal S, et al. Mineralogic analysis of lung tissue from a former coal miner with rapidly progressive pneumoconiosis and progressive massive fibrosis. American Thoracic Society Conference, Dallas TX; May 2019. Poster presentation.
3. Cohen RA, Sanyal S,Hubbs A, et al. Mineralogy of progressive massive fibrosis and rapidly progressive pneumoconiosis in US coal miners. Society of Toxicological Pathology Conference, Raleigh NC; June 2019. Oral presentation.
4. Almberg KS, Zell-Baran L, Abraham JL, et al. Intra- and inter-rater reliability of pathologic classification of type of progressive massive fibrosis among deceased US coal miners. American Thoracic Society Conference, Philadelphia PA; May 2020. Virtual poster presentation.
5. Go L, Abraham JL, Lowers H, et al. Mineralogic analysis of lung tissue from US coal miners demonstrates greater silica burden in modern cases of progressive massive fibrosis. American Thoracic Society Conference, Philadelphia PA; May 2020. Virtual poster presentation.

6. Sarver E, Keles C, Lowers H, et al. Mineralogic analysis of respirable dust from 24 underground coal mines in four geographic regions of the United States. American Thoracic Society Conference, Philadelphia PA; May 2020. Virtual poster presentation.
7. Zell-Baran L, Almberg KS, Go L, et al. Contemporary coal miners with progressive massive fibrosis have shorter mining tenures compared to their historic counterparts. American Thoracic Society Conference 2021. Virtual poster presentation.
8. Go L, Abraham JL, Almberg KS, et al. Increase in the proportion of silica-type progressive massive fibrosis suggested over the history of the National Coal Workers' Autopsy Study. American Thoracic Society Conference, 2021. Virtual poster presentation.
9. Cohen RA, Rose CS, Go LHT, et al. Pathologic findings of progressive massive fibrosis in contemporary coal miners show features of accelerated silicosis compared to historical cases. American Thoracic Society Conference, 2021. Virtual poster presentation.
10. Cohen RA, Rose CS, Go LHT, et al. Increased silica burden is associated with pathologic features of alveolar proteinosis, mature and immature silicotic nodules in US coal miners with progressive massive fibrosis (PMF). American Thoracic Society Conference, 2022. Poster presentation.

6.3 Invited Presentations at national and international meetings:

1. Cohen, RA. "Pathology and Mineralogy of Coal Mine Dust Lung Disease" University of Queensland Dust and Respiratory Health Forum. University of Queensland, Brisbane, Australia. November 12, 2018.

2. Cohen, RA. "Resurgence of Coal Workers' Pneumoconiosis in the US" The Sydney Partnership for Health, Education, Research, and Enterprise (SPHERE). Sydney Technical University, Sydney, Australia. November 16, 2018.

3. Cohen, RA. "Research to Study Rapidly Progressive Pneumoconiosis." Coal Miner Health and Safety Summit, Stone Mountain Health Services, St. Charles, Virginia. April 5, 2019.

4. Cohen, RA. "Testimony for House Committee Conference: Breathless and Betrayed: What is MSHA Doing to Protect Miners from the Resurgence of Black Lung Disease" Subcommittee on Workforce Protections, Committee on Education and Labor, U.S. House of Representatives. Washington, D.C. June 20, 2019.

5. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." University of Queensland Sustainable Minerals Institute. Virtual Presentation. December 3, 2020.

6. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." West Virginia Association of Black Lung Clinics. Virtual Presentation. June 3, 2020.

7. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." University of Kentucky Center for Appalachian Research in Environmental Sciences - Health Seminar. Virtual Presentation. July 15, 2020.

8. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." Icahn School of Medicine at Mount Sinai, Division of Occupational Medicine Grand Rounds. Virtual Presentation. March 19, 2021.

9. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." State University of New York, Stony Brook, Division of Pulmonary Medicine Grand Rounds. Virtual Presentation. August 23, 2021.

10. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." National Coalition of Black Lung and Respiratory Disease Clinics – National Conference. Virtual Presentation. September 22, 2021.

11. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." West Virginia University, Division of Pulmonary and Critical Care Medicine Grand Rounds. Virtual Presentation. October 1, 2021.

12. Cohen, RA. "The Role of Silica in Coal Mine Dust Lung disease." Sustainable Minerals Institute Respiratory Health Forum, University of Queensland, Virtual Presentation, November 4, 2021.

6.4 Dissemination of findings via the media

1. Cohen, RA. Quoted in "Paying for Black Lung: Renewal Deadline Looms for Overextended Federal Trust Fund." Ohio Valley ReSource. Published December 22, 2021. Accessed February 10, 2022. https://ohiovalleyresource.org/2021/12/22/paying-for-black-lung-renewal-deadline-looms-for-overextended-federal-trust-fund/

7.0 Conclusions and Impact Assessment

The US is in the midst of a resurgence of CWP, particularly in central Appalachia, despite regulatory data demonstrating a long-term decline in respirable dust levels. The identification of advanced pneumoconiosis in the form of RPP/PMF in recent decades raised the alarm, with urgent need to identify the underlying cause(s). Until the current project, the role of silica in the resurgence of disease was primarily inferred from chest imaging findings (i.e., 'r' opacities). We employed a multi-method approach, including a novel histopathologic characterization of PMF type; mineralogy using SEM/EDS *in situ* and of digested tissue; and characterization of coal mine dust samples to confirm the importance of silica in the pathogenesis of contemporary cases of RPP/PMF. The pathologic data of dust in the lungs of contemporary miners shows evidence of exposures that are many orders of magnitude above those predicted by contemporary dust exposure limits. Our findings highlight the importance of control of respirable crystalline silica (RCS) levels in the prevention of RPP/PMF.

There is no stated permissible exposure limit (PEL) for RCS for mines in the US, although there was a theoretical limit based on the formula for total respirable dust of 100 mcg/m^3 with the passage of the Federal Coal Mine Health and Safety Act of 1969. However, prior to 2016, dust sampling could be performed during mine operations in which production was as low as 50% of average, raising the probability that reported RCS levels were not representative of typical production conditions and the likelihood that miners were exposed to substantially more RCS. Operations involving mine construction or mine development, which likely produced significant concentrations of RCS, were also not subject to regulatory dust sampling. During the project period, US Department of Labor's Office of Inspector General recommended that MSHA adopt a legal exposure limit for silica in coal mines, which MSHA did not agree with at the time. Our work demonstrates the clear need to regulate RCS specifically, rather than the current indirect regulation.

Our findings also demonstrate that the majority of coal mine dust particles are quite small, in the submicron range. We've also demonstrated that silica particles were smaller than other minerals, and that there were higher concentrations of silica particles in central Appalachian mines compared to those outside of central Appalachia. This corresponds to the pathology findings described in our research. Recent technological advances have resulted in high powered machinery which may pulverize coal and rock generating greater quantities of smaller particles than previous conventional "drill and blast" techniques. Smaller particles, with greater surface areas are more likely to deposit in the most distal airways causing chronic lung diseases including RPP/PMF. It is likely that a greater number of respirable dust particles are now generated for a given mass concentration, and that pulmonary toxicity of coal mine dust is now greater than previously. Our results suggest that simple adherence to the established PEL for total respirable dust is may not be sufficient to prevent disease. Regulatory bodies should strongly consider improving dust control regulations to account for these findings with lower permissible total respirable dust levels and specific limits for RCS. The development of real-time RCS monitoring would contribute significantly to this effort.

Research impacts:

1. Preliminary data showing an increase in silica-type PMF and increased silica particulate in contemporary miners was presented to the Subcommittee on Workforce Protections, Committee on Education and Labor, U.S. House of Representatives in Washington, D.C. June 20, 2019 and had significant impact on the committee's deliberations. MSHA is currently working on a new standard for RCS in US mines.

2. The data from this project prompted our group to study the toxicology of various components of coal mine dust, work which is ongoing in collaboration with Northwestern University and Virginia Tech as part of the Alpha Foundation Grant AFC820-59.

3. Our work on the mineralogy using SEM *in situ* techniques prompted us to search for other technologies to evaluate mineral particulate in human tissues. We are currently engaged in a pilot project to evaluate the use of Keyence® digital microscopy coupled with automated software to identify, size, and quantify particles and compare this to conventional electron microscopy.

4. Our work classifying the pathology of PMF and rapidly progressive pneumoconiosis has prompted our group to promote the concept of PMF type, using the relative predominance of silica nodules within lesions. We have proposed the use of silica-, coal-, and mixed-types of PMF and will be publishing standard images of these types.

5. We have also demonstrated the importance of the pathologic finding of mineral dust alveolar proteinosis (MDAP) which we believe indicates significant exposure to RCS.

8.0 Recommendations for Future Work

We believe the following areas for future research are particularly important, as they may lead to specific interventions on the part of the mining industry that could prevent future development of disease or disease progression in miners:

1. Locate archival/historical samples of coal mine dust and compare particle characteristics to contemporary samples to determine if indeed there are differences in size and composition.

2. Evaluate the pathology of RPP defined by rapid progression of simple pneumoconiotic lesions without the presence of large opacities or complicated lesions. Of great interest would be disease characterized by irregular opacities that is rapidly progressive.

3. Evaluate the physiologic effect of high concentrations RCS in coal mine dust in the causation of obstructive lung diseases.

4. Study the pulmonary toxicology of fibrous silicates, nano-sized silica particles, surface free radicals, as well as the various types of silicates, the ratio of silica to silicates, and carbonaceous dusts.

In addition to the above, we believe the following areas would provide greater understanding of pneumoconiosis in miners, potentially informing interventions on the part of the mining industry:

5. Determine the significance of the inflammatory component of RPP which has not been described in classic PMF. The intense lymphocytic infiltration seen in RPP is a good indicator that we are dealing with an inflammatory component and/or auto-immune response triggered by silica or as yet an unknown pathogen.

6. Evaluate the role of lymphatic occlusion in the pathogenesis of RPP. Without clearance of the pathogen (silica) the disease persists. Our colleague Dr. Anne Hubbs at NIOSH noted lymphatic dilatation indicative of lymphatic obstruction when reviewing some NCWAS materials as part of case selection for this study.

7. Study the significance of mineral dust alveolar proteinosis as this may indicate surfactant deficiency. It is well known that the absence of effective surfactant causes the lungs to collapse and become scar tissue.

8. Study cases of infection associated with PMF. We identified a few cases on slides stained for TB and fungi, but this needs expanding to other potential pathogens using genetic techniques.

9. Study in greater detail the full variety of lesions that manifest as PMF radiologically or pathologically, for example rounded atelectasis, sub pleural silicotic lesions and pulmonary infarction. The pathologists have seen many of these variants in this study.

10. Explain the role of vasculitis that is a major feature of PMF. In this study we have seen dust laden macrophages invade the intima of blood vessels, obliterating the lumen and presumably entering the blood stream, causing systemic effects or thrombosis. The latter is accompanied by necrosis of cartilaginous airways, airway, and vascular obliteration (death) of lung tissue. Some of these features are considered diagnostic of malignancy.

11. Determine the role of infarction in the pathogenesis and progression of PMF, a feature important for interpretation of radiographs and/or interpreting the pathology.

12. Determine why some PMF lesions may behave like malignant neoplasms, invading blood vessels, airways, interlobular septa, the chest wall etc. These are not benign features, only missing is metastasis. There are other entities, e.g., sclerosing peritonitis, where so-called ‘benign lesions’ behave malignantly. In situ analyses on tissue specimens for genes or gene products indicative of cell proliferation might be informative.

9.0 References

1. Wade WA, Petsonk EL, Young B, Mogri I. Severe occupational pneumoconiosis among West Virginian coal miners: one hundred thirty-eight cases of progressive massive fibrosis compensated between 2000 and 2009. *Chest*. 2010;139:1458-1462. doi:10.1378/chest.10-1326

2. Antao VC dos S, Petsonk EL, Sokolow LZ, et al. Rapidly progressive coal workers' pneumoconiosis in the United States: geographic clustering and other factors. *Occup Environ Med*. 2005;62(10):670-674. doi:10.1136/oem.2004.019679

3. Blackley DJ, Halldin CN, Laney AS. Resurgence of a debilitating and entirely preventable respiratory disease among working coal miners. *Am J Respir Crit Care Med*. 2014;190(6):708-709.

4. Blackley DJ. Resurgence of Progressive Massive Fibrosis in Coal Miners — Eastern Kentucky, 2016. *MMWR Morb Mortal Wkly Rep*. 2016;65. doi:10.15585/mmwr.mm6549a1

5. Suarthana E, Laney AS, Storey E, Hale JM, Attfield MD. Coal workers' pneumoconiosis in the United States: regional differences 40 years after implementation of the 1969 Federal Coal Mine Health and Safety Act. *Occup Environ Med*. 2011;68(12):908-913. doi:10.1136/oem.2010.063594

6. Pollock DE, Potts JD, Joy GJ. Investigation into dust exposures and mining practices in mines in the southern Appalachian region. *Min Eng*. 2010;62(2):44-44.

7. Gardner HH. Mechanization in Alberta Coal Mines. *Can Min Metall Bull*. 1951;54:161-166.

8. Shields JJ, Magnuson MO, Haley WA, Dowd JJ. Mechanical Mining in Some Bituminous-Coal Mines; Progress Report 7. Methods of Mining With Continuous-Mining Machines. *Bur Mines Inf Circ*. 1954;7696:1-32.

9. Coal Workers' Autopsy Study. *JAMA*. 1971;217(10):1384-1384. doi:10.1001/jama.1971.03190100066017

10. Centers for Disease Control and Prevention. *Current Intelligence Bulletin 64: Coal Mine Dust Exposures and Associated Health Outcomes - a Review of Information Published since 1995*. DHHS (NIOSH) Publication No. 2011–172 Accessed October 28, 2012. http://www.cdc.gov/niosh/docs/2011-172/

11. Laney AS, Attfield MD. Coal workers' pneumoconiosis and progressive massive fibrosis are increasingly more prevalent among workers in small underground coal mines in the United States. *Occup Environ Med*. 2010;67:428-431. doi:10.1136/oem.2009.050757

12. Advanced Black Lung cases surge in Appalachia. *Things Consid*. Published online December 15, 2016. Accessed January 8, 2017. http://www.npr.org/2016/12/15/505577680/advanced-black-lung-cases-surge-in-appalachia

13. Black Lung returns to coal country: A deadly coal miners' disease is surging back despite rules created to control the disease. NPR.org. Accessed February 27, 2017. https://www.npr.org/series/156453033/black-lung-returns-to-coal-country

14. Almberg K s., Zell-Baran L, Abraham J l., et al. Intra- and Inter-Rater Reliability of Pathologic Classification of Type of Progressive Massive Fibrosis Among Deceased U.S. Coal Miners. In: *A105. SILICA, INORGANIC DUST, AND MINING*. American Thoracic Society International Conference Abstracts. American Thoracic Society; 2020:A2632-A2632. doi:10.1164/ajrccm-conference.2020.201.1_MeetingAbstracts.A2632

15. Method to characterize inorganic particulates in lung tissue biopsies using field emission scanning electron microscopy. Accessed February 4, 2022. https://pubs.er.usgs.gov/publication/70196777

16. Sarver E, Keleş Ç, Afrouz SG. Particle size and mineralogy distributions in respirable dust samples from 25 US underground coal mines. *Int J Coal Geol*. 2021;247:103851. doi:10.1016/j.coal.2021.103851

17. Sarver E, Keles C, Rezaee M. Characteristics of respirable dust in eight Appalachian coal mines: A dataset including particle size and mineralogy distributions, and metal and trace element mass concentrations. *Data Brief*. 2019;25:104032. doi:10.1016/j.dib.2019.104032

18. Sarver E, Keles C, Lowers H, et al. Mineralogic Analysis of Respirable Dust from 24 Underground Coal Mines in Four Geographic Regions of the United States. In: *A105. SILICA, INORGANIC DUST, AND MINING*. American Thoracic Society International Conference Abstracts. American Thoracic Society; 2020:A2635-A2635. doi:10.1164/ajrccm-conference.2020.201.1_MeetingAbstracts.A2635

19. Diehl SF, Goldhaber MB, Hatch JR. Modes of occurrence of mercury and other trace elements in coals from the warrior field, Black Warrior Basin, Northwestern Alabama. *Int J Coal Geol*. 2004;59(3-4):16. doi:10.1016/j.coal.2004.02.003

20. Peacor D. Authigenic Illite and Organic Matter: The Principal Hosts of Vanadium in the Mecca Quarry Shale at Velpen, Indiana. *Clays Clay Miner - CLAYS CLAY Min*. 2000;48:311-316. doi:10.1346/CCMN.2000.0480301

21. Craighead J, Kleinerman, J, Abraham JL, et al. Diseases associated with exposure to silica and non fibrous silicate minerals. Silicosis and Silicate Disease Committee. *Arch Pathol Lab Med*. 1988;112(7):673-720.

22. Honma K, Chiyotani K. Pulmonary alveolar proteinosis as a component of massive fibrosis in cases of chronic pneumoconiosis. An autopsy study of 79 cases. *Zentralbl Pathol*. 1991;137(5):414-417.

23. Cohen RA, Petsonk EL, Rose C, et al. Lung Pathology in U.S. Coal Workers with Rapidly Progressive Pneumoconiosis Implicates Silica and Silicates. *Am J Respir Crit Care Med*. 2016;193(6):673-680. doi:10.1164/rccm.201505-1014OC

24. Abraham JL, Hunt A, Burnett BR. Quantification of Non-Fibrous and Fibrous Particulates in Human Lungs: Twenty Year Update on Pneumoconiosis Database. *Ann Occup Hyg*. 2002;46(suppl 1):397-401. doi:10.1093/annhyg/46.suppl_1.397

25. Abraham JL, Wiesenfeld SL. Two cases of fatal PMF in an ongoing epidemic of accelerated silicosis in oilfield sandblasters: lung pathology and mineralogy. *Ann Occup Hyg*. 1997;41:440-447.

26. Abraham JL. *Analysis of Fibrous and Non-Fibrous Particles. Ch 18. in Rom W (Ed) Environmental and Occupational Medicine*. 4th ed. Wolters Kluwer Lippincott Williams & Wilkins; 2007.

27. Sarver E, Keleş Ç, Afrouz SG. Particle size and mineralogy distributions in respirable dust samples from 25 US underground coal mines. *Int J Coal Geol*. 2021;247:103851. doi:10.1016/j.coal.2021.103851

28. Johann-Essex V, Keles C, Rezaee M, Scaggs-Witte M, Sarver E. Respirable coal mine dust characteristics in samples collected in central and northern Appalachia. *Int J Coal Geol*. 2017;182:85-93. doi:10.1016/j.coal.2017.09.010

29. Paying for Black Lung: Renewal Deadline Looms for Overextended Federal Trust Fund. Ohio Valley ReSource. Published December 22, 2021. Accessed February 10, 2022. https://ohiovalleyresource.org/2021/12/22/paying-for-black-lung-renewal-deadline-looms-for-overextended-federal-trust-fund/

10.0 Abbreviations

CWP	Coal workers' pneumoconiosis
DO	Designated occupation
DPM	Diesel particulate matter
ICP-MS	Inductively coupled plasma mass spectrometry
MDAP	Mineral dust-related alveolar proteinosis
MSHA	Mine Safety and Health Administration
NCWAS	National Coal Workers' Autopsy Study
NIOSH	National Institute for Occupational Safety and Health
PAD	Projected area diameter
PAS-D	Periodic Acid Schiff-Diastase
PC	Polycarbonate
PEL	Permissible exposure limit
PMF	Progressive massive fibrosis
RCS	Respirable crystalline silica
RPP	Rapidly progressive pneumoconiosis
SEM-EDS	Scanning electron microscopy/energy dispersive x-ray spectroscopy
SLF	Simulated lung fluid
USGS	United States Geological Survey

11.0 Appendices

11.1 Mine dust characterization

Mine dust characterization including data description, data tables, and experimental design, materials, and methods.

Appendix 11.1 - Sarver et. al. methods.

Mine dust characterization

This document summarizes the characterization work completed on respirable coal mine dust samples under project Aim 3. Section 1 provides a brief description of the data collected, Section 2 presents the data in tabular form on a per sample basis, and Section 3 details the sample collection and analytical methods.

1. Data Description

This dataset includes 171 sets of respirable coal mine dust samples, which were collected in 25 US mines. Each set represents a unique sampling event (i.e., specific sampling location in a specific mine), and included at least two replicate samples. One sample from each set was analyzed by scanning electron microscope with energy dispersive X-ray (SEM-EDX) using two computer-controlled routines (i.e., one targeting submicron particles and the other targeting supramicron particles) to collect data on particle size and relative elemental ratios. From the elemental data, particle mineralogy was also inferred using published classification criteria [1]. SEM images and EDX data from the submicron routine were also reviewed manually to assess the presence of particles consistent with morphology and chemistry of diesel particulate matter (DPM) [1]. In addition to the SEM-EDX analysis, a sample from each set (except for those collected in Mines 9 and 25) was used for sequential digestions in simulated lung fluid and strong acid, and the digestates were analyzed by inductively coupled plasma-mass spectroscopy to estimate potentially bioaccessible and total acid-soluble concentrations of metals and trace elements.

Table 1 presents a summary of the particle analysis using the sub- and supramicron SEM-EDX routines for each sample, including the number of particles analyzed, total analyzed area, and resulting estimates of particle loading density and relative abundance (number %) of particles in each size range. Table 1 also indicates the assessment of DPM presence in each sample. Table 2 presents the relative abundance (number %) of sub- and supramicron particles in each of nine mineralogy classes per sample: carbonaceous (C); mixed carbonaceous (MC); aluminosilicates, kaolinite-like (ASK) or other (ASO); other silicates (SLO); silica (S); heavy minerals (M), which mostly included metal sulfides or oxides; carbonates (CB), or other (O). Tables 3a-i further divide the relative abundance of particles in each mineralogy class across a total of 14 size bins (based on projected area diameter (nm) covering the sub- and supramicron ranges.

Tables 4a-b present estimated mass concentrations (μg/g) for potentially bioaccessible and total acid-soluble elements for samples from Mines 10-24. (Data for Mines 1-8 were previously published [2]). Elements included in this analysis were Mg, Al, Si, K, V, Cr, Fe, Mn, Co, Ni, Cu, Zn, As, Se, Sr, Ag, Cd, Sn, Ba, Pb, and U.

2. Data Tables

Table 1 Summary of sub- and supramicron SEM-EDX analysis data per sample. Shown for each sample are the mine region (i.e., B=mid-central Appalachia, A=northern Appalachia, C=south-central Appalachia, D=western, and E=mid-western), mine number (i.e., 1-25), and sampling location (i.e., I=intake, R=return, P=production, B=bolter, F=feeder). Assessment of DPM presence is also indicated (DPM = particles observed that are consistent with DPM morphology and chemistry, no DPM = no such particles observed, maybe = particles observed that might be DPM).

Sample				DPM presence	# of particles analyzed		Analyzed area (μm^2)		Particle loading density (# particles/μm^2)		Abundance (number %)	
No.	Reg.	Mine	Loc.		Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra
1	B	1	F	no DPM	186	500	10940	121865	0.0170	0.0041	83	17
2	B	1	B	no DPM	277	500	2054	31343	0.1349	0.0160	91	9
3	B	1	I	no DPM	18	503	9397	1093950	0.0019	0.0005	84	16
4	B	1	R	no DPM	271	500	3431	32593	0.0790	0.0153	86	14
5	B	1	B	no DPM	16	236	11080	1795200	0.0014	0.0001	94	6
6	B	1	P	DPM	298	437	6311	178239	0.0472	0.0025	95	5
7	B	1	B	no DPM	259	500	3158	18019	0.0820	0.0277	79	21
8	B	2	I	maybe	107	508	11189	1697025	0.0096	0.0003	97	3
9	B	2	R	maybe	255	500	1266	21647	0.2014	0.0231	91	9
10	B	2	B	no DPM	258	500	1281	10732	0.2014	0.0466	84	16
11	B	2	P	no DPM	123	503	11080	361693	0.0111	0.0014	90	10
12	B	2	F	maybe	149	502	11220	305074	0.0133	0.0016	91	9
13	B	2	B	no DPM	254	500	1238	13659	0.2052	0.0366	87	13
14	B	3	I	maybe	25	503	10519	1093950	0.0024	0.0005	86	14
15	B	3	R	maybe	264	500	4111	26617	0.0642	0.0188	82	18
16	B	3	B	no DPM	24	500	10519	1122000	0.0023	0.0004	86	14
17	B	3	P	maybe	261	500	2781	23568	0.0938	0.0212	84	16
18	B	3	F	maybe	53	500	10940	1079925	0.0048	0.0005	92	8
19	B	4	I	maybe	59	504	10379	344192	0.0057	0.0015	81	19
20	B	4	R	no DPM	17	501	10379	1668975	0.0016	0.0003	86	14
21	B	4	B	maybe	279	500	3350	30184	0.0833	0.0166	86	14
22	B	4	F	no DPM	2	61	10799	2033625	0.0002	0.0000	89	11
23	A	5	R	no DPM	264	500	3828	28416	0.0690	0.0176	82	18
24	A	5	I	no DPM	22	503	9677	785400	0.0023	0.0006	79	21
25	A	5	I	no DPM	50	501	10659	226595	0.0047	0.0022	70	30
26	A	5	F	no DPM	86	505	10379	168483	0.0083	0.0030	75	25
27	A	5	P	no DPM	280	500	7409	47715	0.0378	0.0105	80	20
28	A	5	R	no DPM	267	500	4576	28721	0.0584	0.0174	79	21
29	A	5	B	no DPM	121	507	10238	80064	0.0118	0.0063	69	31
30	A	5	F	no DPM	136	502	10799	119700	0.0126	0.0042	78	22
31	A	5	F	no DPM	230	500	10260	30672	0.0224	0.0163	60	40
32	A	5	R	no DPM	251	500	1577	11311	0.1591	0.0442	80	20
33	A	6	I	DPM	266	504	8578	109547	0.0310	0.0046	88	12
34	A	6	R	no DPM	230	500	1527	11891	0.1506	0.0420	81	19
35	A	6	F	DPM	280	508	10098	95675	0.0277	0.0053	86	14
36	A	6	I	DPM	173	502	9911	127353	0.0175	0.0039	83	17
37	A	6	R	no DPM	217	500	3296	16952	0.0658	0.0295	73	27
38	A	6	P	no DPM	134	500	9933	85065	0.0135	0.0059	73	27
39	A	6	I	DPM	153	505	9785	92291	0.0156	0.0055	76	24
40	A	6	R	DPM	149	502	10238	160617	0.0146	0.0031	84	16
41	A	6	F	maybe	51	500	9818	288945	0.0052	0.0017	77	23
42	A	6	B	maybe	40	500	10379	1304325	0.0039	0.0004	92	8
43	A	6	I	DPM	44	501	9397	1949475	0.0047	0.0003	95	5
44	A	6	R	maybe	93	501	10519	1122000	0.0088	0.0004	96	4
45	A	6	I	maybe	34	467	9677	2187900	0.0035	0.0002	95	5
46	A	6	F	maybe	68	505	9818	241596	0.0069	0.0021	79	21
47	A	6	I	no DPM	34	495	10519	1711050	0.0032	0.0003	92	8
48	C	7	R	no DPM	277	503	10659	140067	0.0260	0.0036	89	11
49	C	7	I	no DPM	15	504	8555	1255451	0.0018	0.0004	83	17
50	C	7	R	no DPM	270	500	4972	49911	0.0543	0.0100	87	13
51	C	7	I	DPM	47	505	10379	1287312	0.0045	0.0004	93	7
52	C	7	F	no DPM	111	500	10519	40063	0.0106	0.0125	51	49

53	C	7	P	no DPM	87	502	10116	135463	0.0086	0.0037	73	27
54	C	7	P	DPM	268	500	6991	80186	0.0383	0.0062	88	12
55	C	7	F	no DPM	220	500	9843	65430	0.0224	0.0076	77	23
56	C	7	B	no DPM	268	500	3901	33751	0.0687	0.0148	85	15
57	C	7	R	no DPM	262	500	4307	52380	0.0608	0.0095	88	12
58	C	7	P	no DPM	263	505	7363	54088	0.0357	0.0093	81	19
59	C	7	B	no DPM	272	500	6492	43721	0.0419	0.0114	80	20
60	C	7	F	no DPM	245	500	5885	38050	0.0416	0.0131	79	21
61	C	7	P	no DPM	265	503	6171	72442	0.0429	0.0069	87	13
62	C	8	R	no DPM	252	500	2431	12623	0.1037	0.0396	75	25
63	C	8	P	maybe	263	500	4108	67350	0.0640	0.0074	92	8
64	C	8	I	no DPM	47	501	10238	203576	0.0046	0.0025	67	33
65	C	8	F	no DPM	82	500	10519	56710	0.0078	0.0088	50	50
66	C	8	I	maybe	74	504	10519	1105353	0.0070	0.0005	94	6
67	C	8	F	maybe	61	502	9537	296903	0.0064	0.0017	81	19
68	C	8	R	no DPM	251	500	7139	52472	0.0352	0.0095	82	18
69	C	8	R	no DPM	264	500	1298	21220	0.2034	0.0236	91	9
70	C	8	I	maybe	29	502	10519	151927	0.0028	0.0033	49	51
71	C	8	R	maybe	179	504	10659	127993	0.0168	0.0039	83	17
72	C	8	F	no DPM	223	500	10238	73387	0.0218	0.0068	79	21
73	C	8	P	no DPM	244	500	4944	23812	0.0493	0.0210	73	27
74	C	8	P	no DPM	242	500	1417	11007	0.1707	0.0454	82	18
75	C	8	B	no DPM	251	500	2586	21190	0.0970	0.0236	83	17
76	C	8	B	no DPM	270	500	1725	21525	0.1565	0.0232	89	11
77	C	9	I	DPM	291	512	9116	205293	0.0319	0.0025	94	6
78	C	9	F	no DPM	57	510	11080	419043	0.0051	0.0012	82	18
79	C	9	F	DPM	268	503	9714	82236	0.0276	0.0061	83	17
80	C	9	R	DPM	129	505	11220	126027	0.0115	0.0040	78	22
81	C	9	I	DPM	75	508	10940	588044	0.0069	0.0009	90	10
82	C	9	B	DPM	263	509	5891	30162	0.0446	0.0169	76	24
83	C	9	P	DPM	251	500	6469	26404	0.0388	0.0189	71	29
84	C	9	I	maybe	216	505	10519	308550	0.0205	0.0016	93	7
85	C	9	F	DPM	135	505	10940	193743	0.0123	0.0026	84	16
86	C	9	F	DPM	64	505	11080	265957	0.0058	0.0019	78	22
87	C	9	R	DPM	255	506	10519	45936	0.0242	0.0110	72	28
88	C	9	B	maybe	55	504	9818	230208	0.0056	0.0022	75	25
89	C	9	B	no DPM	103	503	9958	92169	0.0103	0.0055	67	33
90	C	9	P	DPM	279	504	3635	67617	0.0768	0.0075	92	8
91	C	11	R	maybe	254	506	2059	14751	0.1234	0.0343	81	19
92	C	11	P	no DPM	227	506	1023	16018	0.2219	0.0316	89	11
93	C	11	B	DPM	286	507	1159	20427	0.2469	0.0248	92	8
94	C	11	F	DPM	292	504	4657	149655	0.0627	0.0034	95	5
95	B	15	R	maybe	269	508	3619	27654	0.0743	0.0184	83	17
96	B	15	B	no DPM	176	504	10238	85107	0.0172	0.0059	77	23
97	B	15	I	no DPM	27	80	10799	336600	0.0025	0.0002	92	8
98	B	15	P	maybe	268	450	4769	30723	0.0562	0.0146	82	18
99	B	15	F	no DPM	113	504	10799	104049	0.0105	0.0048	71	29
100	C	12	I	DPM	299	504	1524	215391	0.1962	0.0023	99	1
101	C	12	P	DPM	258	504	1944	13728	0.1327	0.0367	81	19
102	C	12	B	DPM	296	502	3787	105633	0.0782	0.0048	95	5
103	C	12	R	DPM	242	508	1263	14058	0.1917	0.0361	87	13
104	C	14	F	DPM	269	501	2357	15906	0.1141	0.0315	81	19
105	C	14	P	no DPM	222	501	1217	13673	0.1825	0.0366	85	15
106	C	14	I	DPM	297	504	7433	163218	0.0400	0.0031	93	7
107	C	14	B	DPM	156	502	11080	208032	0.0141	0.0024	86	14
108	A	18	R	no DPM	267	512	3752	20922	0.0712	0.0245	78	22
109	A	18	P	no DPM	258	506	1615	10131	0.1598	0.0499	80	20
110	A	18	I	maybe	194	504	10238	452298	0.0189	0.0011	95	5
111	A	18	B	maybe	178	503	9397	65538	0.0189	0.0077	74	26
112	A	18	F	maybe	199	503	10342	155430	0.0192	0.0032	87	13
113	A	17	I	DPM	124	506	10379	181401	0.0119	0.0028	83	17
114	A	17	I	no DPM	104	505	10379	109989	0.0100	0.0046	72	28
115	A	17	P	no DPM	267	507	1531	11715	0.1744	0.0433	83	17

116	A	17	R	no DPM	234	503	2030	9009	0.1153	0.0558	72	28
117	A	17	B	DPM	285	511	3210	30096	0.0888	0.0170	86	14
118	A	17	R	DPM	266	509	5770	25047	0.0461	0.0203	73	27
119	A	16	P	DPM	275	504	1319	13563	0.2085	0.0372	87	13
120	A	16	R	DPM	282	509	2221	36333	0.1270	0.0140	91	9
121	A	16	B	DPM	291	507	5203	77814	0.0559	0.0065	91	9
122	A	16	F	DPM	288	507	1395	24222	0.2064	0.0209	92	8
123	D	19	P	DPM	283	510	668	13530	0.4238	0.0377	93	7
124	D	19	R	DPM	285	507	658	11154	0.4334	0.0455	92	8
125	D	19	I	DPM	292	503	5369	86691	0.0544	0.0058	91	9
126	D	19	F	no DPM	171	505	3445	18315	0.0496	0.0276	67	33
127	D	19	B	maybe	272	508	654	10989	0.4157	0.0462	92	8
128	D	19	R	maybe	276	506	804	11583	0.3432	0.0437	91	9
129	D	20	P	no DPM	268	503	912	10164	0.2939	0.0495	88	12
130	D	20	R	DPM	284	506	579	12606	0.4903	0.0401	94	6
131	D	20	F	DPM	297	509	880	93951	0.3376	0.0054	99	1
132	D	20	I	DPM	298	506	1953	272250	0.1526	0.0019	99	1
133	D	20	F	DPM	290	507	602	10428	0.4821	0.0486	92	8
134	D	20	B	DPM	273	505	613	10560	0.4457	0.0478	92	8
135	C	13	B	no DPM	237	504	1268	10890	0.1869	0.0463	83	17
136	C	13	R	no DPM	242	507	1352	10725	0.1790	0.0473	80	20
137	C	13	F	DPM	287	505	956	18084	0.3004	0.0279	92	8
138	C	13	I	DPM	147	505	9257	57849	0.0159	0.0087	68	32
139	C	13	R	DPM	264	504	4496	18711	0.0587	0.0269	72	28
140	C	13	I	DPM	271	504	2011	19338	0.1348	0.0261	85	15
141	C	10	F	DPM	213	506	6452	88671	0.0330	0.0057	84	16
142	C	10	R	DPM	280	515	6311	56298	0.0444	0.0091	80	20
143	C	10	B	DPM	266	506	1463	16038	0.1818	0.0316	81	19
144	C	10	P	no DPM	269	506	621	9240	0.4331	0.0548	84	16
145	C	10	F	DPM	278	505	3506	16401	0.0793	0.0308	71	29
146	C	10	B	DPM	265	508	890	9867	0.2977	0.0515	80	20
147	C	10	I	DPM	294	509	8836	387189	0.0333	0.0013	95	5
148	C	22	F	DPM	293	502	1632	122892	0.1795	0.0041	98	2
149	C	22	P	DPM	248	505	1906	12111	0.1301	0.0417	79	21
150	C	22	I	DPM	297	502	2092	236511	0.1420	0.0021	99	1
151	C	21	P	no DPM	257	502	1154	11583	0.2226	0.0433	86	14
152	C	21	F	DPM	271	500	5853	37290	0.0463	0.0134	80	20
153	C	21	I	DPM	261	509	3868	21846	0.0675	0.0233	78	22
154	C	21	B	DPM	265	508	7176	24783	0.0369	0.0205	69	31
155	C	21	R	no DPM	251	501	1694	11418	0.1481	0.0439	80	20
156	E	23	I	DPM	299	503	389	17391	0.7692	0.0289	97	3
157	E	23	I	DPM	295	505	437	31218	0.6757	0.0162	98	2
158	E	23	I	DPM	293	503	317	17655	0.9231	0.0285	97	3
159	E	23	R	DPM	272	510	715	10890	0.3803	0.0468	91	9
160	E	23	P	no DPM	257	508	2239	18381	0.1148	0.0276	84	16
161	E	24	R	DPM	296	507	317	15873	0.9344	0.0319	97	3
162	E	24	I	DPM	256	505	567	9900	0.4517	0.0510	91	9
163	E	24	F	DPM	300	504	382	34089	0.7847	0.0148	98	2
164	E	24	R	no DPM	252	506	573	9867	0.4396	0.0513	91	9
165	E	24	I	DPM	297	508	196	40656	1.5150	0.0125	99	1
166	E	24	I	DPM	299	504	247	47190	1.2122	0.0107	99	1
167	C	25	I	no DPM	311	500	5610	100052	0.0554	0.0050	92	8
168	C	25	F	no DPM	312	500	1706	12379	0.1829	0.0404	84	16
169	C	25	B	no DPM	300	500	1391	10535	0.2157	0.0475	84	16
170	C	25	R	no DPM	302	500	1712	24857	0.1764	0.0201	92	8
171	C	25	P	no DPM	320	500	899	14519	0.3561	0.0344	93	7

Table 2. Relative abundance of sub- and supramicron particles in each mineralogy class per sample. (Values sum to 100% for each sample.)

Sample No	Abundance (number %)																	
	C		MC		ASK		ASO		SLO		S		M		CB		O	
	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra	Sub	Supra
1	49	4	20	3	1	1	5	3	0	0	1	1	2	0	2	4	3	0
2	41	1	31	1	4	2	12	4	0	0	4	1	0	0	0	0	0	0
3	54	1	14	1	0	0	0	1	0	0	0	0	0	0	16	13	0	0
4	39	0	26	1	4	3	12	7	0	0	3	1	0	0	1	1	0	0
5	82	4	12	1	0	0	0	0	0	0	0	0	0	0	0	1	0	0
6	91	0	1	0	0	0	1	1	0	0	0	0	0	0	1	3	1	0
7	21	1	22	2	6	4	24	12	0	0	5	1	1	0	1	0	0	0
8	44	0	4	0	3	0	24	1	0	0	2	0	0	0	17	0	5	0
9	2	0	9	0	1	0	14	3	2	0	64	5	0	0	0	0	0	0
10	1	0	7	0	0	0	31	10	2	0	43	6	0	0	1	0	0	0
11	34	1	5	0	2	0	22	5	1	0	24	3	1	0	1	0	0	0
12	77	4	5	1	1	1	4	2	0	0	3	1	1	0	0	0	0	0
13	2	0	5	0	3	0	69	13	0	0	8	0	0	0	0	0	0	0
14	61	1	4	0	0	1	10	3	0	0	4	2	0	0	8	6	0	0
15	24	1	23	2	5	3	22	9	0	0	7	2	1	0	0	0	0	0
16	39	2	21	1	0	2	7	3	0	0	0	2	7	1	11	4	0	0
17	11	1	13	2	8	3	45	9	0	0	4	2	1	0	0	0	0	0
18	81	4	7	1	0	1	0	2	0	0	2	0	2	0	0	1	0	0
19	26	2	15	1	7	3	26	9	0	0	4	3	1	0	1	1	0	0
20	70	1	5	0	0	0	0	1	0	0	0	0	0	0	11	10	0	0
21	31	1	20	1	1	2	10	5	0	0	23	4	1	0	0	0	0	0
22	88	7	0	1	0	0	0	1	0	0	0	1	0	0	0	1	0	0
23	35	3	19	2	9	4	16	7	0	0	2	1	0	0	1	2	0	0
24	53	2	0	1	4	2	7	6	0	0	0	2	0	0	15	7	0	0
25	32	7	7	2	5	2	12	4	0	0	1	1	1	0	9	13	3	0
26	21	2	6	1	4	1	9	2	0	0	2	1	0	0	33	17	1	1
27	48	6	13	2	10	4	6	4	0	0	1	1	1	0	2	3	0	0
28	39	3	15	2	8	3	10	6	0	0	1	1	0	0	6	5	0	0
29	18	8	11	2	2	2	9	5	1	0	2	1	1	0	23	13	2	1
30	35	6	11	3	2	1	2	2	0	0	1	0	0	0	26	11	1	0
31	18	5	5	1	3	0	6	3	1	0	2	1	1	0	24	27	2	2
32	2	0	5	0	0	0	3	1	1	0	0	0	2	0	55	15	12	3
33	40	1	14	1	1	0	8	2	0	0	1	0	3	0	21	6	0	0
34	1	0	3	0	0	0	0	0	2	0	0	0	1	0	65	18	7	1
35	59	2	16	2	1	1	3	3	0	0	1	1	1	0	6	6	0	0
36	7	0	6	0	0	0	4	0	0	0	0	0	0	0	61	16	4	0
37	10	0	6	0	0	0	2	0	0	0	0	0	0	0	54	25	0	1
38	36	19	12	3	2	2	1	0	0	0	1	0	0	0	21	3	1	0
39	32	3	8	1	0	0	0	0	0	0	0	0	0	0	35	19	0	0
40	49	3	17	1	1	1	2	1	0	0	1	0	1	0	14	9	0	0
41	58	7	5	1	0	1	5	1	0	0	0	0	0	0	10	12	0	0
42	80	1	2	1	2	0	0	1	0	0	2	0	2	0	3	4	0	0
43	82	0	4	0	0	0	2	0	0	0	2	0	0	0	5	3	0	0
44	88	1	1	0	2	0	2	0	1	0	1	0	0	0	0	3	0	0
45	83	1	3	0	0	0	0	0	0	0	0	0	0	0	9	3	0	0
46	61	11	11	3	3	2	1	1	0	0	0	0	0	0	3	4	0	0
47	46	1	19	0	0	0	0	0	3	0	0	0	3	0	22	5	0	0
48	47	2	24	1	3	2	7	4	0	0	3	1	4	0	1	1	0	0
49	65	1	0	0	0	1	0	6	0	0	5	1	0	0	11	7	0	1
50	39	1	25	2	4	3	15	6	0	0	3	1	0	0	1	0	0	0
51	84	1	2	0	0	1	0	1	0	0	0	0	0	0	6	4	0	0
52	18	8	9	4	1	3	10	10	0	0	1	2	1	1	10	20	1	1
53	38	6	12	2	3	4	11	9	0	0	1	2	1	0	7	4	0	0
54	32	1	26	1	6	2	15	4	1	0	5	1	1	0	2	2	1	0
55	14	1	12	2	7	5	29	11	0	0	7	2	1	0	5	1	2	0
56	28	2	24	2	3	1	19	5	0	0	3	1	0	0	6	5	0	0
57	25	1	29	1	4	1	19	6	0	0	10	2	1	0	0	0	0	0
58	36	3	18	2	4	2	15	6	0	0	3	1	0	0	5	5	0	0
59	27	1	19	2	8	3	22	11	0	0	4	1	0	0	1	1	0	0
60	22	1	16	1	5	4	30	12	0	0	2	1	0	0	3	2	1	0
61	31	1	19	1	5	2	27	7	0	0	4	1	0	0	2	1	0	0
62	1	0	3	0	0	0	5	2	0	0	0	0	0	0	58	14	7	9

63	37	0	22	1	3	2	8	3	0	0	21	2	0	0	0	0	0	0
64	14	3	3	1	3	3	23	14	0	0	18	3	0	0	6	7	0	2
65	20	5	11	4	3	9	12	24	0	0	1	4	1	0	1	3	0	0
66	89	1	1	0	0	1	3	3	0	0	1	0	0	0	0	1	0	0
67	63	3	11	1	0	3	3	6	0	0	0	2	0	0	4	3	0	0
68	17	1	16	1	9	5	28	10	1	0	9	2	0	0	1	1	1	0
69	14	0	12	0	0	0	5	2	1	0	59	6	0	0	0	0	0	0
70	27	2	2	1	2	6	13	27	0	0	3	5	0	0	2	9	0	1
71	44	2	12	1	5	4	8	6	0	0	13	2	1	0	1	2	0	0
72	27	2	16	2	2	2	11	8	0	0	18	6	1	0	2	1	0	0
73	10	0	16	1	2	2	28	19	1	0	15	4	1	0	0	0	0	0
74	0	0	0	0	0	0	27	7	0	0	55	12	0	0	0	0	0	0
75	30	1	16	1	3	3	13	8	0	0	21	4	0	0	0	0	0	0
76	40	0	18	1	0	1	10	4	0	0	21	5	0	0	0	0	0	0
77	81	3	9	1	0	0	2	1	0	0	2	0	0	0	0	0	0	0
78	46	1	22	4	0	1	3	6	0	0	6	1	3	1	3	2	0	2
79	39	4	21	4	3	1	13	4	1	0	4	1	2	0	0	0	1	0
80	41	10	21	5	1	1	10	3	0	0	4	1	2	0	0	0	0	0
81	84	3	2	2	0	1	0	2	0	0	0	0	1	0	2	2	1	0
82	37	9	21	7	3	2	14	6	0	0	1	1	1	0	0	0	0	0
83	25	3	17	4	4	3	18	15	0	0	5	2	1	0	1	2	1	0
84	82	2	5	1	0	0	3	1	0	0	2	0	0	0	2	2	0	0
85	56	8	16	3	1	0	6	1	1	0	1	0	2	0	1	3	0	0
86	44	2	15	2	5	4	9	10	0	0	4	2	0	0	1	1	0	0
87	40	10	14	7	1	2	10	5	0	0	4	2	1	0	1	2	0	0
88	27	5	24	5	1	1	15	10	0	0	3	2	3	0	3	1	0	0
89	37	11	7	6	0	3	15	6	0	0	3	2	1	0	4	4	1	1
90	48	2	23	1	1	1	10	2	1	0	1	0	4	0	5	1	0	0
91	0	0	1	0	5	1	72	18	0	0	3	0	1	0	0	0	0	0
92	0	0	2	0	6	0	76	11	0	0	5	0	0	0	0	0	0	0
93	59	0	14	1	2	1	13	5	0	0	1	1	2	0	1	0	0	0
94	89	2	4	1	0	0	0	1	0	0	0	0	1	0	0	0	0	0
95	28	3	23	3	9	4	19	5	0	0	4	1	0	0	0	0	0	0
96	42	10	17	4	2	2	10	3	0	0	4	2	0	0	2	2	0	0
97	85	1	3	1	0	0	0	0	0	0	0	0	3	0	0	6	0	0
98	49	5	19	3	6	4	7	3	0	0	2	1	0	0	0	1	0	0
99	61	18	4	2	1	1	4	2	0	0	0	1	0	0	2	5	0	0
100	99	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	20	2	19	3	6	5	31	8	0	0	3	1	2	0	0	0	0	0
102	92	3	1	1	0	0	1	0	0	0	0	0	0	0	1	0	0	0
103	2	0	6	0	6	1	66	12	0	0	3	0	0	0	1	0	2	0
104	42	2	15	3	1	3	17	10	0	0	2	1	3	0	1	0	0	0
105	0	0	0	0	2	0	83	15	0	0	1	0	0	0	0	0	0	0
106	87	2	2	1	0	0	3	1	0	0	0	0	0	0	1	2	0	0
107	60	5	15	3	1	2	9	2	0	0	0	1	1	0	1	0	0	0
108	46	6	17	5	5	5	8	5	0	0	2	1	0	0	0	0	0	0
109	12	1	19	1	12	9	35	9	0	0	2	0	0	0	0	0	0	0
110	89	1	3	0	1	0	0	0	0	0	1	0	0	0	0	2	0	0
111	33	9	20	5	5	4	11	4	0	0	3	2	2	0	1	1	0	0
112	80	6	4	2	0	1	0	1	0	0	0	0	0	0	1	2	0	0
113	46	2	19	1	0	0	2	0	0	0	0	0	1	0	15	13	0	0
114	39	12	20	4	3	6	3	3	0	0	1	1	2	0	5	2	0	0
115	13	1	16	2	26	9	24	5	0	0	2	0	1	0	0	0	1	0
116	0	0	0	0	29	17	38	11	0	0	5	0	0	0	0	0	0	0
117	53	3	14	1	1	0	2	1	0	0	8	2	0	0	8	6	0	0
118	38	10	14	2	1	0	1	0	0	0	1	0	0	0	18	13	1	0
119	42	5	16	4	5	1	8	2	0	0	1	1	12	0	1	0	1	1
120	73	6	10	1	2	0	1	0	0	0	1	0	3	0	1	0	0	0
121	79	5	7	1	1	0	2	0	0	0	1	0	1	0	0	1	0	0
122	81	5	7	1	2	1	1	0	0	0	0	0	0	0	1	0	0	0
123	43	2	23	2	0	0	16	2	1	0	2	0	2	0	6	1	0	0
124	30	1	32	2	0	0	16	3	1	0	6	0	1	0	6	1	1	0
125	85	3	1	1	0	0	1	1	0	0	0	0	0	0	3	3	0	0
126	3	0	11	1	1	0	45	28	0	0	6	3	0	0	1	0	0	0
127	28	1	30	2	1	0	26	4	0	0	7	0	0	0	0	0	0	0
128	34	1	31	2	0	0	17	4	1	0	5	0	2	0	1	0	0	0
129	0	0	2	0	0	0	78	12	0	0	3	0	0	0	4	0	2	0
130	39	1	33	1	1	0	17	3	0	0	0	0	1	0	2	1	0	0

131	96	1	1	0	0	0	1	0	0	0	0	0	0	0	1	0	0	0
132	97	0	1	0	0	0	0	0	0	0	1	0	0	0	0	1	0	0
133	62	2	16	1	1	0	11	2	0	0	1	0	0	0	1	1	0	0
134	2	0	13	0	0	0	56	6	0	0	3	0	1	0	15	2	2	0
135	0	0	0	0	7	1	68	17	0	0	8	0	0	0	0	0	0	0
136	0	0	0	0	0	0	21	1	8	0	0	0	0	0	6	4	46	16
137	85	5	4	1	0	0	1	0	0	0	0	0	1	0	1	1	0	0
138	52	13	4	2	0	0	1	1	0	0	0	0	1	0	9	16	0	0
139	61	21	7	3	1	1	1	2	0	0	1	1	1	0	0	1	0	0
140	74	6	4	2	0	1	4	4	0	0	1	1	0	0	1	1	1	0
141	70	6	8	3	0	2	4	3	1	0	0	1	1	0	0	0	0	0
142	58	10	15	3	1	1	3	3	1	0	1	2	1	0	0	0	0	0
143	42	6	29	4	3	2	6	7	0	0	1	1	0	0	0	0	0	0
144	0	0	0	0	50	11	33	5	0	0	0	1	0	0	0	0	0	0
145	49	6	11	5	3	8	6	8	1	0	0	1	1	1	0	0	0	0
146	17	2	22	2	16	7	23	8	0	0	1	1	0	0	0	0	0	0
147	87	2	5	1	0	0	1	0	0	0	1	1	1	0	0	0	0	0
148	92	1	2	0	0	0	0	0	0	0	2	0	1	0	0	0	0	0
149	1	0	3	0	2	0	67	20	0	0	7	0	0	0	0	0	0	0
150	97	1	1	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0
151	1	0	0	0	3	0	77	14	0	0	4	0	0	0	0	0	0	0
152	31	5	25	5	2	2	14	5	0	0	3	1	4	0	1	1	0	0
153	46	4	18	5	1	3	10	7	0	0	3	1	0	0	1	1	0	0
154	32	6	20	6	1	3	8	9	0	0	3	2	2	0	2	4	0	1
155	0	0	1	0	2	1	71	19	0	0	6	0	0	0	0	0	0	0
156	92	2	3	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0
157	80	1	4	0	0	0	0	0	0	0	0	0	0	0	13	1	0	0
158	83	1	11	0	0	0	0	0	0	0	0	0	0	0	3	1	0	0
159	53	0	15	0	0	0	0	0	0	0	0	0	0	0	22	8	0	0
160	16	1	13	1	0	0	1	0	0	0	0	0	1	0	51	14	1	0
161	83	1	5	0	0	0	4	0	0	0	4	1	0	0	1	0	0	0
162	6	0	5	0	0	0	5	1	0	0	76	7	0	0	0	0	0	0
163	97	1	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	2	0	1	0	1	0	17	0	3	0	0	0	0	0	58	7	11	2
165	99	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	99	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	61	2	5	1	2	1	15	1	1	0	4	1	1	0	3	1	0	0
168	15	1	16	2	3	1	38	9	1	0	5	1	4	0	2	1	1	1
169	3	0	9	0	8	4	58	11	0	0	5	0	1	0	0	0	0	0
170	38	2	21	2	3	1	26	2	0	0	3	0	1	0	0	0	0	0
171	32	1	25	1	4	2	28	3	0	0	3	0	1	0	0	0	0	0

Table 3a Relative abundance of carbonaceous (C) particles by size bin (nm) for each sample. (Values sum to total abundance in C class as shown in Table 2 for each sample.)

Sample No	Relative abundance of C particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	≥5000
1	19	17	6	3	2	0	1	0	1	3	0	0	0	0
2	11	16	8	4	1	0	0	0	0	1	0	0	0	0
3	18	4	9	4	13	4	0	0	0	1	0	0	0	0
4	11	14	9	2	2	0	1	0	0	0	0	0	0	0
5	57	23	0	0	0	0	0	0	2	4	0	0	0	0
6	67	18	4	2	0	0	0	0	0	0	0	0	0	0
7	5	7	3	3	1	0	0	0	0	1	0	0	0	0
8	33	6	3	1	1	0	0	0	0	0	0	0	0	0
9	1	0	0	1	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
11	30	2	0	1	1	0	0	0	0	1	0	0	0	0
12	50	20	4	2	1	0	1	0	1	3	1	0	0	0
13	1	0	0	1	0	0	0	0	0	0	0	0	0	0
14	34	17	3	3	0	3	0	0	0	1	0	0	0	0
15	13	5	4	1	0	0	0	0	1	1	0	0	0	0
16	24	10	0	3	0	0	0	0	0	1	0	0	0	0
17	5	3	2	1	0	0	0	0	0	1	0	0	0	0
18	40	21	3	5	7	3	2	0	0	2	1	1	0	0
19	8	5	8	4	0	0	0	0	0	1	0	0	0	0
20	40	15	10	0	0	5	0	0	0	1	0	0	0	0
21	16	6	4	3	1	0	0	0	0	1	0	0	0	0
22	43	0	43	0	0	0	0	0	2	6	0	0	0	0
23	12	11	5	2	3	1	0	0	0	2	1	0	0	0
24	43	4	7	0	0	0	0	0	0	1	1	0	0	0
25	15	10	1	3	3	0	0	0	1	5	2	0	0	0
26	9	7	2	3	1	0	0	0	0	1	1	0	0	0
27	13	12	8	4	5	4	1	0	1	4	1	1	0	0
28	14	9	5	3	2	2	2	0	1	2	1	0	0	0
29	4	4	2	2	2	1	1	0	1	5	1	1	0	0
30	8	5	6	8	4	3	1	0	1	4	1	0	0	0
31	2	4	4	5	2	1	0	0	1	5	1	0	0	0
32	0	0	1	0	0	0	0	0	0	0	0	0	0	0
33	21	8	6	2	1	0	0	0	0	1	0	0	0	0
34	1	0	0	0	0	0	0	0	0	0	0	0	0	0
35	29	16	9	2	2	0	0	0	0	2	0	0	0	0
36	4	1	1	0	0	0	0	0	0	0	0	0	0	0
37	3	2	2	1	1	1	0	0	0	0	0	0	0	0
38	7	7	6	5	6	3	0	0	2	11	5	2	1	0
39	15	4	5	2	1	3	0	0	0	1	1	1	0	0
40	25	10	5	3	3	2	1	0	1	2	1	0	0	0
41	37	16	1	3	0	0	0	0	1	3	2	1	1	0
42	59	14	5	2	0	0	0	0	0	1	0	0	0	0
43	60	15	4	0	0	0	0	0	2	0	0	0	0	0
44	67	12	5	1	3	0	0	0	0	0	0	0	0	0
45	55	22	3	3	0	0	0	0	0	1	0	0	0	0
46	33	15	8	2	2	0	0	0	1	7	3	1	0	0
47	38	5	0	0	0	3	0	0	0	1	0	0	0	0
48	17	14	10	2	2	1	0	0	0	1	0	0	0	0
49	22	33	11	0	0	0	0	0	0	1	0	0	0	0
50	12	12	9	3	2	0	0	0	1	1	0	0	0	0
51	53	18	10	2	0	2	0	0	0	1	0	0	0	0
52	6	4	4	2	0	0	0	0	1	6	2	0	0	0
53	15	9	4	6	2	1	0	0	1	4	2	1	0	0
54	10	9	7	2	2	2	0	0	0	1	0	0	0	0
55	5	4	2	0	1	0	0	0	0	1	0	0	0	0
56	6	12	7	1	1	0	0	0	0	1	1	0	0	0

57	10	8	4	2	1	0	0	0	0	0	0	0	0	0
58	9	11	8	4	2	0	1	0	1	2	1	0	0	0
59	10	7	4	4	1	1	0	0	0	1	0	0	0	0
60	6	6	4	3	2	0	0	0	0	1	0	0	0	0
61	11	10	6	2	2	0	0	0	0	1	0	0	0	0
62	1	1	0	0	0	0	0	0	0	0	0	0	0	0
63	15	10	7	2	1	1	0	0	0	0	0	0	0	0
64	7	1	0	4	1	0	0	0	0	2	1	0	0	0
65	9	3	5	1	1	1	0	0	1	4	1	1	0	0
66	72	10	4	1	0	1	0	0	0	0	0	0	0	0
67	39	13	4	3	4	0	0	0	0	1	1	1	0	0
68	7	6	2	2	1	0	0	0	0	1	0	0	0	0
69	5	4	2	1	0	0	0	0	0	0	0	0	0	0
70	19	2	6	0	0	0	0	0	0	2	0	0	0	0
71	20	12	7	4	1	0	0	0	0	1	1	0	0	0
72	8	11	3	2	2	0	0	0	0	2	1	0	0	0
73	4	3	2	1	0	0	0	0	0	0	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	13	8	6	2	2	0	0	0	0	0	0	0	0	0
76	16	13	6	3	1	1	0	0	0	0	0	0	0	0
77	38	24	8	6	2	1	1	0	1	3	0	0	0	0
78	11	20	7	3	1	1	1	0	0	0	0	0	0	0
79	16	10	7	3	2	0	0	0	1	4	0	0	0	0
80	17	11	3	5	1	1	0	0	3	10	0	0	0	0
81	40	22	9	2	4	5	0	0	1	2	0	0	0	0
82	14	11	4	2	2	1	0	0	2	9	0	0	0	0
83	10	6	5	3	1	1	0	0	0	2	0	0	0	0
84	44	24	9	2	3	0	0	0	0	2	0	0	0	0
85	27	14	9	5	0	1	0	0	1	7	1	0	0	0
86	20	5	7	8	4	0	0	0	1	1	0	0	0	0
87	16	10	5	2	3	1	1	0	3	10	1	0	0	0
88	10	10	1	1	1	0	0	0	2	4	1	0	0	0
89	8	11	6	4	3	3	1	0	1	9	1	0	0	0
90	19	19	6	2	1	0	0	0	1	2	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0	0	0	0	0
93	31	17	7	2	2	0	0	0	0	0	0	0	0	0
94	56	19	7	7	0	1	0	0	0	1	0	0	0	0
95	11	8	6	1	0	1	0	0	1	3	0	0	0	0
96	14	11	9	1	1	1	1	0	2	8	1	0	0	0
97	57	20	3	3	0	0	0	0	0	1	1	0	0	0
98	20	13	8	4	1	0	0	0	2	5	0	0	0	0
99	25	13	4	5	6	3	2	1	2	11	4	2	1	0
100	73	16	7	2	1	0	0	0	0	1	0	0	0	0
101	9	5	3	2	1	0	0	0	1	2	0	0	0	0
102	59	20	8	2	1	1	0	0	1	3	1	0	0	0
103	0	1	1	0	0	0	0	0	0	0	0	0	0	0
104	22	11	5	3	0	0	0	0	1	1	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	59	18	6	3	0	0	1	0	0	2	0	0	0	0
107	31	20	4	2	0	1	0	0	1	5	0	0	0	0
108	14	14	6	6	3	2	0	0	2	5	0	0	0	0
109	4	4	3	1	0	0	0	0	0	1	0	0	0	0
110	51	22	11	3	1	0	0	0	1	1	0	0	0	0
111	9	12	4	4	1	0	0	0	2	7	1	1	0	0
112	41	21	12	4	0	1	0	0	1	5	1	0	0	0
113	27	14	3	1	1	0	0	0	1	2	0	0	0	0
114	15	12	7	3	1	0	1	0	2	9	2	0	0	0
115	5	3	4	0	0	0	0	0	0	1	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	33	12	3	2	1	1	0	0	1	2	0	0	0	0
118	11	7	7	4	3	2	1	0	2	8	2	0	0	0
119	18	10	6	4	3	1	0	0	1	3	1	0	0	0

120	34	15	11	6	3	3	1	0	1	5	1	0	0	0
121	48	16	6	4	2	1	0	0	1	4	1	0	0	0
122	43	22	8	5	2	1	0	0	1	4	1	0	0	0
123	22	9	5	4	2	0	0	0	0	1	0	0	0	0
124	13	9	5	2	0	0	1	0	0	1	0	0	0	0
125	60	16	5	2	1	1	0	0	0	2	0	0	0	0
126	1	1	0	0	0	1	0	0	0	0	0	0	0	0
127	9	13	4	1	1	0	0	0	0	1	0	0	0	0
128	14	8	6	3	1	1	0	0	0	1	0	0	0	0
129	0	0	0	0	0	0	0	0	0	0	0	0	0	0
130	18	11	6	2	1	1	0	0	0	1	0	0	0	0
131	56	24	11	3	2	0	0	0	0	1	0	0	0	0
132	63	21	10	2	1	0	0	0	0	0	0	0	0	0
133	39	15	5	3	1	0	0	0	0	2	0	0	0	0
134	1	1	0	0	0	0	0	0	0	0	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	0	0	0	0	0	0	0	0	0	0	0	0	0	0
137	48	23	8	4	1	1	0	0	1	3	1	0	0	0
138	29	15	4	1	0	0	0	0	2	11	1	0	0	0
139	26	9	11	5	4	3	2	0	2	14	4	2	0	0
140	41	13	6	6	4	3	1	0	1	4	1	1	0	0
141	0	37	16	11	4	1	1	0	0	1	4	0	0	0
142	0	25	16	10	4	1	1	0	0	3	7	0	0	0
143	0	13	12	6	4	4	2	0	0	2	4	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	27	12	5	3	2	0	0	0	1	4	1	0	0
146	0	10	2	3	1	0	0	0	0	1	1	0	0	0
147	0	54	20	9	3	2	0	0	0	1	1	0	0	0
148	55	25	8	3	1	0	0	0	0	1	0	0	0	0
149	0	0	0	0	0	0	0	0	0	0	0	0	0	0
150	57	25	10	3	1	2	0	0	0	1	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	15	7	3	3	1	0	0	0	1	4	0	0	0	0
153	21	12	5	4	2	1	0	0	2	4	0	0	0	0
154	12	9	6	2	1	0	0	0	2	5	0	0	0	0
155	0	0	0	0	0	0	0	0	0	0	0	0	0	0
156	61	21	7	2	1	1	0	0	0	2	0	0	0	0
157	54	15	4	4	2	0	1	0	0	1	0	0	0	0
158	50	19	7	3	2	1	0	0	0	1	0	0	0	0
159	37	10	4	1	1	0	0	0	0	0	0	0	0	0
160	9	3	1	1	0	1	0	0	1	1	0	0	0	0
161	56	16	7	2	1	1	0	0	0	1	0	0	0	0
162	2	1	1	1	0	0	0	0	0	0	0	0	0	0
163	60	25	7	2	1	1	0	0	0	1	0	0	0	0
164	0	0	0	0	0	0	0	0	0	0	0	0	0	0
165	49	25	12	7	5	1	0	0	0	0	0	0	0	0
166	54	27	10	5	3	0	0	0	0	0	0	0	0	0
167	43	10	4	1	0	0	1	0	1	2	0	0	0	0
168	6	5	1	2	0	0	0	0	0	1	0	0	0	0
169	2	1	0	0	0	0	0	0	0	0	0	0	0	0
170	16	11	7	1	1	1	0	0	1	1	0	0	0	0
171	15	10	4	2	1	0	0	0	0	1	0	0	0	0

Table 3b Relative abundance of mixed-carbonaceous (MC) particles by size bin (nm) for each sample. (Values sum to total abundance in C class as shown in Table 2 for each sample.)

Sample No	Relative abundance of MC particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	1	3	5	4	2	1	2	0	1	2	0	0	0	0
2	1	4	7	6	8	2	1	0	1	1	0	0	0	0
3	0	0	4	4	0	4	0	0	0	1	0	0	0	0
4	1	3	8	7	5	1	0	0	1	1	0	0	0	0
5	6	6	0	0	0	0	0	0	0	1	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	1	3	6	5	3	2	1	0	1	2	0	0	0	0
8	1	1	1	1	0	0	0	0	0	0	0	0	0	0
9	2	2	2	2	1	0	0	0	0	0	0	0	0	0
10	1	3	1	1	1	0	0	0	0	0	0	0	0	0
11	3	1	0	1	0	0	0	0	0	0	0	0	0	0
12	1	1	2	1	0	0	0	0	0	1	0	0	0	0
13	2	1	1	0	0	0	0	0	0	0	0	0	0	0
14	0	3	0	0	0	0	0	0	0	0	0	0	0	0
15	3	5	4	5	3	1	1	0	1	2	0	0	0	0
16	0	7	3	3	3	3	0	0	0	1	0	0	0	0
17	2	5	4	0	1	0	0	0	1	2	0	0	0	0
18	2	3	0	2	0	0	0	0	0	0	0	0	0	0
19	5	3	4	1	1	0	0	0	0	1	0	0	0	0
20	0	0	5	0	0	0	0	0	0	0	0	0	0	0
21	3	2	4	6	4	0	0	0	0	1	0	0	0	0
22	0	0	0	0	0	0	0	0	0	1	0	0	0	0
23	1	5	4	3	2	3	1	0	0	1	0	0	0	0
24	0	0	0	0	0	0	0	0	0	1	0	0	0	0
25	0	3	1	0	1	0	1	0	0	1	1	0	0	0
26	0	3	2	1	1	0	0	0	0	0	0	0	0	0
27	1	3	3	2	2	1	0	0	0	1	1	0	0	0
28	1	2	3	2	3	2	0	0	0	2	0	0	0	0
29	1	2	4	3	1	1	1	0	0	2	0	0	0	0
30	2	3	3	2	1	0	1	0	0	2	0	0	0	0
31	0	1	2	1	0	1	0	0	0	1	0	0	0	0
32	0	1	1	2	0	1	0	0	0	0	0	0	0	0
33	2	2	4	3	3	1	0	0	0	1	0	0	0	0
34	0	1	1	0	0	0	0	0	0	0	0	0	0	0
35	2	1	3	3	2	2	1	0	1	1	0	0	0	0
36	2	3	1	0	0	0	0	0	0	0	0	0	0	0
37	1	1	1	2	1	1	0	0	0	0	0	0	0	0
38	2	5	2	2	1	0	0	0	0	2	0	0	0	0
39	1	2	2	0	1	0	0	0	0	0	0	0	0	0
40	1	6	4	4	1	1	0	0	0	1	0	0	0	0
41	0	0	0	3	0	0	1	0	0	1	0	0	0	0
42	2	0	0	0	0	0	0	0	0	0	0	0	0	0
43	0	0	2	0	0	2	0	0	0	0	0	0	0	0
44	0	0	0	1	0	0	0	0	0	0	0	0	0	0
45	3	0	0	0	0	0	0	0	0	0	0	0	0	0
46	2	3	2	0	1	1	0	0	0	2	0	0	0	0
47	5	5	5	3	0	0	0	0	0	0	0	0	0	0
48	2	3	7	5	5	2	0	0	0	1	0	0	0	0
49	0	0	0	0	0	0	0	0	0	0	0	0	0	0
50	2	4	5	7	3	3	0	0	1	2	0	0	0	0
51	2	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	2	2	2	1	0	0	0	1	2	1	0	0	0
53	0	2	2	4	2	1	2	0	0	2	0	0	0	0
54	4	4	5	5	5	2	1	0	0	1	0	0	0	0
55	2	2	3	3	0	1	0	0	1	2	0	0	0	0
56	2	6	9	6	2	0	0	0	1	1	0	0	0	0

57	3	6	9	6	3	2	0	0	0	0	0	0	0	0
58	2	5	5	3	3	0	0	0	0	1	0	0	0	0
59	3	4	5	4	2	1	0	0	0	2	0	0	0	0
60	1	4	3	2	2	2	1	0	0	1	0	0	0	0
61	3	5	5	3	2	1	1	0	0	1	0	0	0	0
62	1	1	1	0	0	0	0	0	0	0	0	0	0	0
63	3	5	4	4	2	2	0	0	1	1	0	0	0	0
64	0	0	1	1	0	0	0	0	0	1	0	0	0	0
65	1	1	3	2	1	2	0	0	1	3	0	0	0	0
66	1	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	4	3	3	0	1	0	0	0	1	0	0	0	0
68	1	3	4	3	3	1	1	0	1	1	0	0	0	0
69	4	2	3	2	1	0	0	0	0	0	0	0	0	0
70	0	0	2	0	0	0	0	0	0	1	0	0	0	0
71	1	2	4	3	1	0	1	0	0	1	0	0	0	0
72	2	3	3	4	1	1	1	0	0	1	0	0	0	0
73	3	4	4	2	1	1	0	0	0	1	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	3	3	2	4	3	2	0	0	1	1	0	0	0	0
76	3	4	3	3	2	3	1	0	0	1	0	0	0	0
77	2	2	2	2	1	1	0	0	0	1	0	0	0	0
78	6	4	3	3	3	1	1	0	1	3	1	0	0	0
79	5	3	5	4	2	1	1	0	0	4	1	0	0	0
80	2	5	6	5	2	2	0	0	1	5	1	0	0	0
81	0	1	0	0	0	0	0	0	0	2	0	0	0	0
82	3	3	5	3	4	1	1	0	1	5	1	0	0	0
83	2	1	3	4	2	1	1	0	1	4	0	0	0	0
84	2	1	1	1	0	0	0	0	0	0	0	0	0	0
85	1	4	4	3	3	1	1	0	0	2	1	0	0	0
86	2	1	2	4	2	2	0	0	1	2	0	0	0	0
87	1	2	2	3	3	2	0	0	1	6	1	0	0	0
88	1	5	5	5	4	3	0	0	1	4	1	0	0	0
89	1	1	1	3	0	0	1	0	0	3	2	0	0	0
90	3	7	8	3	2	0	0	0	0	1	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	1	0	0	0	0	0	0	0	0	0	0	0
93	5	3	2	1	1	2	1	0	0	1	0	0	0	0
94	1	1	0	0	1	0	1	0	0	1	0	0	0	0
95	4	3	7	3	2	1	1	0	1	3	0	0	0	0
96	3	1	3	5	3	1	1	0	1	3	1	0	0	0
97	3	0	0	0	0	0	0	0	0	1	0	0	0	0
98	1	1	4	5	5	2	1	0	0	3	0	0	0	0
99	1	1	1	0	1	0	0	0	0	2	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	3	3	6	3	2	1	1	0	1	3	0	0	0	0
102	0	0	0	0	1	0	0	0	0	0	0	0	0	0
103	1	1	0	2	1	0	0	0	0	0	0	0	0	0
104	2	2	2	3	2	1	1	0	1	3	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	1	1	0	0	0	0	0	0	1	0	0	0	0
107	2	2	3	3	4	1	1	0	0	2	1	0	0	0
108	1	3	5	3	3	1	1	0	1	4	0	0	0	0
109	3	6	5	3	1	1	0	0	0	1	0	0	0	0
110	0	0	2	1	0	0	0	0	0	0	0	0	0	0
111	2	3	5	4	4	1	0	0	1	5	0	0	0	0
112	0	1	0	1	0	1	0	0	0	1	0	0	0	0
113	1	5	4	6	2	1	0	0	0	1	0	0	0	0
114	1	3	5	5	3	2	1	0	1	4	1	0	0	0
115	3	4	3	3	2	0	0	0	1	2	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	2	4	4	2	1	0	1	0	0	1	0	0	0	0
118	1	3	4	2	3	0	0	0	1	1	0	0	0	0
119	2	6	2	3	2	1	0	0	1	3	1	0	0	0

120	1	2	1	4	2	1	0	0	0	1	0	0	0	0
121	0	2	1	1	2	1	1	0	0	1	0	0	0	0
122	1	1	1	2	2	1	0	0	0	1	0	0	0	0
123	4	4	4	5	3	2	1	0	0	2	0	0	0	0
124	5	8	5	5	3	3	2	0	0	2	0	0	0	0
125	0	0	0	0	0	0	0	0	0	1	0	0	0	0
126	2	2	3	2	1	1	0	0	0	1	0	0	0	0
127	7	6	8	2	3	3	0	0	1	2	0	0	0	0
128	4	8	7	4	4	2	1	0	1	2	0	0	0	0
129	1	0	0	0	0	0	0	0	0	0	0	0	0	0
130	4	7	7	7	4	1	1	0	1	1	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	4	3	2	3	2	1	1	0	0	1	0	0	0	0
134	5	2	3	2	1	0	0	0	0	0	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	0	0	0	0	0	0	0	0	0	0	0	0	0	0
137	1	0	2	0	1	0	0	0	0	0	0	0	0	0
138	0	0	1	2	0	0	0	0	0	2	0	0	0	0
139	0	0	1	3	1	1	0	0	1	2	1	0	0	0
140	1	1	0	0	1	1	0	0	0	1	0	0	0	0
141	0	0	0	0	1	2	2	2	1	0	0	2	1	0
142	0	0	1	2	4	4	2	1	0	0	0	2	0	0
143	0	0	2	3	7	7	5	3	1	0	1	3	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	0	1	0	2	2	4	2	1	0	1	4	1	0
146	0	0	5	5	4	4	3	1	0	0	1	1	0	0
147	0	0	0	1	1	2	1	0	0	0	0	1	0	0
148	0	0	0	0	1	0	0	0	0	0	0	0	0	0
149	0	2	0	1	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	1	0	0	0	0	0	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	1	4	5	7	4	3	0	0	1	4	1	0	0	0
153	2	1	4	5	3	1	0	0	1	5	0	0	0	0
154	1	3	4	5	4	1	1	0	1	5	1	0	0	0
155	0	0	1	0	0	0	0	0	0	0	0	0	0	0
156	2	0	0	0	0	0	0	0	0	0	0	0	0	0
157	2	1	1	0	0	0	0	0	0	0	0	0	0	0
158	7	1	1	1	0	0	0	0	0	0	0	0	0	0
159	7	4	1	1	0	0	1	0	0	0	0	0	0	0
160	2	4	4	3	1	0	0	0	0	1	0	0	0	0
161	3	0	1	0	0	0	0	0	0	0	0	0	0	0
162	1	0	1	0	1	1	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	0	0	0	0	0	0	0	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	2	1	1	1	1	0	0	0	0	1	0	0	0	0
168	6	4	1	1	2	1	1	0	1	2	0	0	0	0
169	4	2	1	1	0	0	0	0	0	0	0	0	0	0
170	3	3	4	6	3	1	0	0	1	2	0	0	0	0
171	6	6	7	4	1	1	0	0	0	1	0	0	0	0

Table 3c Relative abundance of kaolinite-like aluminosilicates (ASK) particles by size bin (nm) for each sample. (Values sum to total abundance in ASK class as shown in Table 2 for each sample.)

Sample No	Relative abundance of ASK particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	1	0	0	0	0	0	0	0	1	0	0	0	0
2	0	0	1	0	1	1	0	0	0	1	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	1	0	1	1	0	1	0	0	1	2	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	0	0	1	1	1	1	0	0	1	3	1	0	0	0
8	0	1	1	0	0	0	1	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
11	0	1	0	1	1	0	0	0	0	0	0	0	0	0
12	0	0	0	1	0	0	0	0	0	1	0	0	0	0
13	1	1	1	0	0	0	0	0	0	0	0	0	0	0
14	0	0	0	0	0	0	0	0	0	1	0	0	0	0
15	1	1	1	0	1	1	0	0	1	2	1	0	0	0
16	0	0	0	0	0	0	0	0	0	1	1	0	0	0
17	1	1	2	1	1	1	0	0	0	2	1	0	0	0
18	0	0	0	0	0	0	0	0	0	1	0	0	0	0
19	1	0	0	3	0	1	1	0	0	2	1	0	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	0	0	0	0	0	0	0	0	0	1	0	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	2	0	0	2	1	2	1	0	1	3	1	0	0	0
24	0	0	0	4	0	0	0	0	0	0	1	0	0	0
25	1	1	0	0	1	0	0	0	0	1	1	0	0	0
26	0	2	1	1	1	0	0	0	0	0	0	0	0	0
27	1	2	1	2	1	1	1	0	1	2	1	0	0	0
28	1	2	1	1	2	1	0	0	0	3	0	0	0	0
29	1	1	0	0	0	1	0	0	0	1	0	0	0	0
30	0	1	1	1	0	0	1	0	0	0	0	0	0	0
31	0	1	0	1	1	0	0	0	0	0	0	0	0	0
32	0	0	0	0	0	0	0	0	0	0	0	0	0	0
33	0	0	0	0	0	0	0	0	0	0	0	0	0	0
34	0	0	0	0	0	0	0	0	0	0	0	0	0	0
35	0	0	1	0	0	0	0	0	0	0	0	0	0	0
36	0	0	0	0	0	0	0	0	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	0	0	0	0
38	1	1	0	0	1	0	0	0	0	1	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	0	1	0	0	0	0	0	0	1	0	0	0	0
41	0	0	0	0	0	0	0	0	0	0	0	0	0	0
42	0	0	0	0	2	0	0	0	0	0	0	0	0	0
43	0	0	0	0	0	0	0	0	0	0	0	0	0	0
44	0	0	0	0	0	1	1	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	2	0	0	0	1	0	0	1	0	0	0	0
47	0	0	0	0	0	0	0	0	0	0	0	0	0	0
48	0	0	0	1	0	1	0	0	0	2	0	0	0	0
49	0	0	0	0	0	0	0	0	0	1	0	0	0	0
50	0	1	0	0	2	0	0	0	0	2	0	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	0	0	0	0	0	0	0	0	2	1	1	0	0
53	0	0	1	1	1	0	0	0	0	2	1	0	0	0
54	0	1	1	1	1	1	0	0	0	1	0	0	0	0
55	0	2	1	1	2	1	0	0	0	3	1	1	0	0
56	0	1	1	1	0	0	0	0	0	1	0	0	0	0

57	1	0	1	1	1	1	0	0	0	1	0	0	0	0
58	1	0	0	1	1	1	0	0	0	1	0	0	0	0
59	1	0	2	1	1	2	1	0	0	2	1	0	0	0
60	1	0	1	1	1	1	0	0	1	2	1	0	0	0
61	0	1	1	1	1	1	0	0	0	1	0	0	0	0
62	0	0	0	0	0	0	0	0	0	0	0	0	0	0
63	0	0	1	1	1	0	0	0	0	1	0	0	0	0
64	0	0	1	1	0	0	0	0	0	1	0	1	0	0
65	1	0	0	1	1	0	1	0	0	4	3	1	0	0
66	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	0	0	0	0	0	0	0	0	1	1	0	0	0
68	1	1	2	1	1	1	1	0	1	3	1	0	0	0
69	0	0	0	0	0	0	0	0	0	0	0	0	0	0
70	0	0	2	0	0	0	0	0	0	3	1	1	0	0
71	0	1	0	0	2	0	0	0	0	2	1	0	0	0
72	0	0	0	0	1	1	0	0	0	1	1	0	0	0
73	0	0	1	1	0	0	0	0	0	1	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	1	1	1	0	0	0	0	0	0	2	0	0	0	0
76	0	0	0	0	0	0	0	0	0	1	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	0	0	0	0	0	0	0	0	0	0	0	0	0	0
79	0	0	1	1	1	0	0	0	0	0	1	0	0	0
80	0	0	0	1	0	0	0	0	0	1	0	0	0	0
81	0	0	0	0	0	0	0	0	0	1	0	0	0	0
82	0	0	0	0	1	1	0	0	0	1	0	0	0	0
83	0	1	1	0	1	1	0	0	0	2	1	0	0	0
84	0	0	0	0	0	0	0	0	0	0	0	0	0	0
85	0	0	0	0	0	1	0	0	0	0	0	0	0	0
86	0	1	1	0	1	0	1	0	0	2	1	0	0	0
87	0	0	0	0	1	0	0	0	0	1	1	0	0	0
88	0	0	0	1	0	0	0	0	0	0	1	0	0	0
89	0	0	0	0	0	0	0	0	0	1	1	1	0	0
90	1	0	0	0	0	0	0	0	0	0	0	0	0	0
91	0	1	3	1	0	0	0	0	0	1	0	0	0	0
92	1	1	2	0	0	0	1	0	0	0	0	0	0	0
93	1	0	0	0	0	0	0	0	0	1	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	2	1	1	1	1	1	1	0	0	2	1	0	0	0
96	0	0	0	1	0	0	0	0	0	1	1	0	0	0
97	0	0	0	0	0	0	0	0	0	0	0	0	0	0
98	1	1	1	1	1	0	1	0	0	2	2	0	0	0
99	0	0	0	0	1	0	0	0	0	0	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	1	1	1	2	2	1	0	0	1	4	1	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	1	1	2	1	1	0	0	0	1	1	0	0	0	0
104	0	0	0	0	0	0	0	0	0	2	1	0	0	0
105	0	0	0	0	1	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	0	0	0	0	0
107	0	0	1	0	0	0	0	0	0	1	1	0	0	0
108	0	1	1	1	0	1	1	0	0	3	1	0	0	0
109	2	2	2	2	2	1	0	0	1	7	1	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	1	0	0	1	2	1	0	0	0	2	2	1	0	0
112	0	0	0	0	0	0	0	0	0	0	1	0	0	0
113	0	0	0	0	0	0	0	0	0	0	0	0	0	0
114	0	0	0	1	2	0	0	0	0	3	2	1	0	0
115	2	3	4	5	7	3	1	0	1	7	1	0	0	0
116	3	6	5	5	3	3	1	0	3	14	2	1	0	0
117	0	0	0	0	0	0	1	0	0	0	0	0	0	0
118	0	0	0	0	0	0	0	0	0	0	0	0	0	0
119	1	0	0	1	1	1	0	0	0	1	0	0	0	0

120	0	1	0	1	0	1	0	0	0	0	0	0	0	0
121	0	0	0	0	0	1	0	0	0	0	0	0	0	0
122	1	0	0	0	0	0	1	0	0	0	0	0	0	0
123	0	0	0	0	0	0	0	0	0	0	0	0	0	0
124	0	0	0	0	0	0	0	0	0	0	0	0	0	0
125	0	0	0	0	0	0	0	0	0	0	0	0	0	0
126	0	0	0	0	0	0	0	0	0	0	0	0	0	0
127	0	0	0	0	0	0	0	0	0	0	0	0	0	0
128	0	0	0	0	0	0	0	0	0	0	0	0	0	0
129	0	0	0	0	0	0	0	0	0	0	0	0	0	0
130	0	1	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	0	0	0	0	0	0	0	0	0	0
134	0	0	0	0	0	0	0	0	0	0	0	0	0	0
135	1	1	2	1	1	1	0	0	0	1	0	0	0	0
136	0	0	0	0	0	0	0	0	0	0	0	0	0	0
137	0	0	0	0	0	0	0	0	0	0	0	0	0	0
138	0	0	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	1	0	0	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	0	0	0	0	0	0	0	0
141	0	0	0	0	0	0	0	0	0	0	0	0	1	1
142	0	0	0	0	0	0	0	0	0	0	0	0	0	1
143	0	0	0	0	1	0	0	1	1	0	0	0	1	1
144	0	0	0	8	14	12	10	4	3	2	0	1	6	1
145	0	0	0	0	0	0	0	2	1	0	0	0	4	3
146	0	0	0	3	2	2	3	4	2	1	0	1	4	1
147	0	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	0	1	0	0	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	0	1	1	0	0	0	0	0	0	0	0	0	0	0
152	0	0	0	1	0	1	0	0	0	1	1	0	0	0
153	0	0	0	0	0	0	0	0	0	2	1	0	0	0
154	0	0	0	0	1	0	0	0	0	2	1	0	0	0
155	0	0	1	0	0	0	0	0	0	1	0	0	0	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	0	0	0	0	0	0	0	0	0	0	0	0	0
161	0	0	0	0	0	0	0	0	0	0	0	0	0	0
162	0	0	0	0	0	0	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	0	0	0	0	0	0	0	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	1	1	0	0	0	0	0	0	0	0	0	0	0	0
168	1	0	1	1	0	0	1	0	0	1	0	0	0	0
169	3	1	1	1	1	1	0	0	1	3	1	0	0	0
170	1	1	0	0	0	1	0	0	0	1	0	0	0	0
171	1	0	1	1	1	1	0	0	0	1	0	0	0	0

Table 3d Relative abundance of other aluminosilicates (ASO) particles by size bin (nm) for each sample. (Values sum to total abundance in ASO class as shown in Table 2 for each sample.)

Sample No	Relative abundance of ASO particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	1	0	0	1	1	1	0	0	2	1	0	0	0
2	0	2	1	1	4	2	1	0	0	3	1	0	0	0
3	0	0	0	0	0	0	0	0	0	1	0	0	0	0
4	1	2	1	3	2	1	2	0	1	6	1	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	1	2	3	5	3	5	3	0	2	10	2	1	0	0
8	5	5	4	5	5	1	1	0	0	1	0	0	0	0
9	2	1	2	3	2	1	0	0	1	3	0	0	0	0
10	5	9	4	3	4	3	1	0	2	8	2	1	0	0
11	4	7	3	3	4	1	0	0	0	2	1	1	0	0
12	1	1	0	1	2	1	0	0	0	2	1	0	0	0
13	12	15	11	14	7	6	3	0	2	9	2	1	0	0
14	3	0	0	3	3	0	0	0	0	2	1	0	0	0
15	2	3	3	5	3	2	2	0	1	7	1	0	0	0
16	0	0	0	0	3	0	3	0	0	2	1	0	0	0
17	8	5	8	9	9	3	3	0	1	6	2	1	0	0
18	0	0	0	0	0	0	0	0	0	1	0	0	0	0
19	5	4	5	4	3	1	3	0	1	4	2	1	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	0	1	1	2	3	1	1	0	1	3	1	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	3	2	2	1	5	2	2	0	1	5	1	0	0	0
24	0	0	4	0	0	0	0	4	0	2	1	1	1	1
25	1	5	1	1	1	1	0	0	0	2	1	1	0	0
26	4	1	1	1	1	0	1	0	0	1	1	0	0	0
27	1	1	1	1	2	0	0	0	0	2	1	0	0	0
28	1	2	1	2	1	1	1	0	1	4	2	0	0	0
29	2	3	1	1	1	1	1	0	0	2	1	1	1	1
30	0	0	1	0	1	1	0	0	0	1	0	1	0	0
31	1	1	1	2	1	0	0	0	0	1	1	0	0	0
32	1	1	0	1	0	0	1	0	0	1	0	0	0	0
33	2	0	2	1	1	1	0	0	0	2	0	0	0	0
34	0	0	0	0	0	0	0	0	0	0	0	0	0	0
35	1	0	0	1	0	1	0	0	0	2	0	0	0	0
36	0	0	2	1	0	0	0	0	0	0	0	0	0	0
37	0	0	0	1	1	0	0	0	0	0	0	0	0	0
38	0	0	0	1	1	0	0	0	0	0	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	0	0	1	0	1	0	0	0	0	0	0	0	0
41	0	0	0	1	3	0	0	0	0	1	0	0	0	0
42	0	0	0	0	0	0	0	0	0	0	0	0	0	0
43	0	2	0	0	0	0	0	0	0	0	0	0	0	0
44	0	0	1	1	0	0	0	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	0	0	0	0	0	1	0	1	0	0	0	0
47	0	0	0	0	0	0	0	0	0	0	0	0	0	0
48	1	1	1	1	1	2	0	0	0	3	1	0	0	0
49	0	0	0	0	0	0	0	0	0	3	2	1	0	0
50	1	1	1	2	3	4	2	0	1	4	1	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	1	1	2	2	2	0	0	1	4	4	2	1	0
53	2	2	2	1	3	0	1	0	1	5	2	1	1	0
54	2	2	2	3	2	3	1	0	1	2	1	1	0	0
55	3	6	4	8	2	4	1	0	1	6	3	1	1	1
56	1	2	4	4	3	3	1	0	1	3	1	0	0	0

57	2	2	2	4	3	4	1	0	1	5	1	0	0	0
58	0	2	3	4	4	1	0	0	1	4	1	0	0	0
59	3	2	4	5	3	2	1	0	1	7	2	1	0	0
60	1	7	5	6	2	6	2	0	1	7	2	1	1	0
61	3	4	5	5	6	4	1	0	1	4	2	1	0	0
62	0	1	1	0	2	0	1	0	0	1	0	0	0	0
63	0	0	1	2	2	1	0	0	0	2	0	0	0	0
64	1	3	7	4	0	7	0	0	1	6	4	2	2	1
65	2	2	1	2	2	2	1	0	0	13	5	3	2	2
66	0	0	1	0	1	0	0	0	0	1	1	1	0	0
67	0	0	0	1	0	0	1	0	0	3	2	1	1	0
68	3	3	8	5	3	3	2	0	1	6	2	1	0	0
69	0	1	1	1	1	0	0	0	0	2	0	0	0	0
70	3	0	3	0	5	0	0	0	2	12	7	5	2	1
71	0	1	1	1	0	3	0	0	0	3	2	1	1	0
72	1	2	1	2	2	2	1	0	1	6	1	0	0	0
73	4	4	4	4	5	3	1	1	2	12	3	2	1	0
74	4	4	6	4	3	3	2	0	1	6	1	0	0	0
75	2	1	2	1	3	1	1	0	1	6	1	0	0	0
76	0	1	2	2	2	2	0	0	1	3	0	0	0	0
77	0	0	0	1	0	0	0	0	0	0	0	0	0	0
78	3	0	0	0	0	0	0	0	0	5	1	0	0	0
79	1	2	2	3	2	2	1	0	0	2	2	1	0	0
80	0	1	1	4	2	0	2	0	0	2	1	0	0	0
81	0	0	0	0	0	0	0	0	0	1	0	0	0	0
82	1	1	2	2	2	3	2	0	0	3	2	1	0	0
83	1	2	4	2	3	5	1	0	1	8	4	2	0	1
84	0	0	0	1	0	0	0	0	0	0	0	0	0	0
85	0	0	2	2	0	1	0	0	0	0	0	0	0	0
86	0	1	1	0	5	0	0	1	1	7	2	1	0	0
87	1	1	1	1	1	3	1	1	0	2	2	1	0	0
88	1	0	3	7	3	0	1	0	0	3	3	2	1	1
89	3	3	1	3	3	1	2	0	0	1	2	2	1	0
90	3	1	3	1	1	0	0	0	0	1	0	0	0	0
91	11	13	14	12	9	7	2	1	2	13	4	1	0	0
92	13	12	13	15	11	7	3	0	1	8	2	1	0	0
93	3	3	3	3	2	0	0	0	0	3	1	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	3	3	1	4	3	3	1	0	0	3	1	1	0	0
96	3	2	2	0	2	2	0	0	0	1	1	1	0	0
97	0	0	0	0	0	0	0	0	0	0	0	0	0	0
98	1	1	1	1	2	0	1	0	0	2	1	0	0	0
99	1	1	0	1	1	1	0	0	0	1	1	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	3	4	3	6	5	6	2	0	1	6	2	0	0	0
102	1	0	0	0	0	0	0	0	0	0	0	0	0	0
103	7	15	14	10	9	6	2	0	2	9	2	1	0	0
104	1	2	2	3	3	3	1	0	1	6	3	1	0	0
105	11	15	16	16	12	9	3	0	2	11	3	1	0	0
106	0	1	1	0	0	0	0	0	0	0	0	0	0	0
107	1	1	2	2	2	1	0	0	0	0	1	1	0	0
108	1	0	0	2	2	2	1	0	0	3	1	0	0	0
109	2	6	5	6	7	4	2	0	2	7	2	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	1	2	0	0	2	4	2	0	0	2	1	0	0	0
112	0	0	0	0	0	0	0	0	0	0	0	0	0	0
113	0	0	0	1	1	0	0	0	0	0	0	0	0	0
114	0	0	0	0	1	2	0	0	0	2	1	0	0	0
115	2	3	4	3	5	5	2	0	1	4	1	0	0	0
116	5	5	7	6	7	5	1	0	2	8	2	1	0	0
117	0	0	0	0	0	1	0	0	0	0	0	0	0	0
118	0	0	0	0	1	0	0	0	0	0	0	0	0	0
119	2	1	2	2	0	2	0	0	0	1	0	0	0	0

120	0	0	0	0	0	0	0	0	0	0	0	0	0	0
121	0	0	0	1	0	1	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	3	2	3	3	3	2	0	0	0	1	0	0	0	0
124	1	3	2	3	2	2	1	0	0	2	0	0	0	0
125	0	0	1	0	0	0	0	0	0	0	0	0	0	0
126	8	9	6	8	7	3	2	0	2	18	6	2	1	0
127	3	5	4	5	5	3	1	0	0	3	1	0	0	0
128	3	3	5	2	1	2	0	0	1	4	0	0	0	0
129	9	15	13	15	12	8	2	1	2	9	2	0	0	0
130	3	4	3	2	3	2	1	0	1	2	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	2	3	0	2	2	1	1	0	0	2	1	0	0	0
134	10	13	9	9	8	5	1	0	1	5	1	0	0	0
135	8	9	13	13	12	7	3	0	2	13	3	1	0	0
136	2	4	2	4	5	3	1	0	0	0	0	0	0	0
137	1	0	0	0	0	0	0	0	0	0	0	0	0	0
138	0	0	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	0	0	1	0	0	0	1	1	0	0	0
140	1	0	1	0	0	1	0	0	0	2	1	1	0	0
141	0	0	0	0	1	1	0	0	1	1	0	0	0	2
142	0	0	0	0	0	1	0	1	1	1	0	0	0	1
143	0	0	0	0	1	0	2	1	1	2	2	0	0	2
144	0	0	0	0	5	10	7	6	4	2	0	0	1	2
145	0	0	0	0	1	1	1	2	1	1	1	0	1	6
146	0	0	0	0	3	3	5	6	5	3	1	0	1	4
147	0	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	9	11	13	9	9	8	3	1	3	15	3	1	1	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	13	13	8	14	14	9	4	0	2	10	3	1	0	0
152	2	1	2	4	2	3	1	0	0	2	2	1	0	0
153	1	1	1	1	1	3	2	0	0	5	1	1	0	0
154	1	2	0	1	1	1	0	0	1	4	3	1	0	0
155	10	13	12	12	10	8	3	0	2	14	4	1	1	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	1	0	0	0	0	0	0	0	0	0	0	0	0
161	2	1	0	0	0	0	0	0	0	0	0	0	0	0
162	1	2	0	1	0	0	0	0	0	1	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	2	2	4	2	4	1	1	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	4	3	3	2	2	1	1	0	0	0	0	0	0	0
168	7	7	5	5	6	4	3	0	1	6	2	1	0	0
169	14	12	8	7	8	5	2	0	1	8	2	1	0	0
170	4	3	3	5	5	3	1	0	0	2	0	0	0	0
171	7	5	3	3	4	3	1	0	1	2	0	0	0	0

Table 3e Relative abundance of other silicate (SLO) particles by size bin (nm) for each sample. (Values sum to total abundance in SLO class as shown in Table 2 for each sample.)

Sample No	Relative abundance of SLO particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	0	0	0	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	0	0	0	0	0	0	0	0	0	0	0	0	0	0
8	0	0	0	0	0	0	0	0	0	0	0	0	0	0
9	0	1	0	0	0	0	0	0	0	0	0	0	0	0
10	1	1	0	0	0	0	0	0	0	0	0	0	0	0
11	0	0	0	1	0	0	0	0	0	0	0	0	0	0
12	0	0	0	0	0	0	0	0	0	0	0	0	0	0
13	0	0	0	0	0	0	0	0	0	0	0	0	0	0
14	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15	0	0	0	0	0	0	0	0	0	0	0	0	0	0
16	0	0	0	0	0	0	0	0	0	0	0	0	0	0
17	0	0	0	0	0	0	0	0	0	0	0	0	0	0
18	0	0	0	0	0	0	0	0	0	0	0	0	0	0
19	0	0	0	0	0	0	0	0	0	0	0	0	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	0	0	0	0	0	0	0	0	0	0	0	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	0	0	0	0	0	0	0	0	0	0	0	0	0	0
24	0	0	0	0	0	0	0	0	0	0	0	0	0	0
25	0	0	0	0	0	0	0	0	0	0	0	0	0	0
26	0	0	0	0	0	0	0	0	0	0	0	0	0	0
27	0	0	0	0	0	0	0	0	0	0	0	0	0	0
28	0	0	0	0	0	0	0	0	0	0	0	0	0	0
29	0	0	0	0	0	1	0	0	0	0	0	0	0	0
30	0	0	0	0	0	0	0	0	0	0	0	0	0	0
31	0	1	0	0	0	0	0	0	0	0	0	0	0	0
32	0	0	0	0	0	0	0	0	0	0	0	0	0	0
33	0	0	0	0	0	0	0	0	0	0	0	0	0	0
34	0	0	0	0	1	0	0	0	0	0	0	0	0	0
35	0	0	0	0	0	0	0	0	0	0	0	0	0	0
36	0	0	0	0	0	0	0	0	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	0	0	0	0
38	0	0	0	0	0	0	0	0	0	0	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	0	0	0	0	0	0	0	0	0	0	0	0	0
41	0	0	0	0	0	0	0	0	0	0	0	0	0	0
42	0	0	0	0	0	0	0	0	0	0	0	0	0	0
43	0	0	0	0	0	0	0	0	0	0	0	0	0	0
44	1	0	0	0	0	0	0	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	0	0	0	0	0	0	0	0	0	0	0	0
47	0	0	0	3	0	0	0	0	0	0	0	0	0	0
48	0	0	0	0	0	0	0	0	0	0	0	0	0	0
49	0	0	0	0	0	0	0	0	0	0	0	0	0	0
50	0	0	0	0	0	0	0	0	0	0	0	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	0	0	0	0	0	0	0	0	0	0	0	0	0
53	0	0	0	0	0	0	0	0	0	0	0	0	0	0
54	0	0	0	0	0	0	0	0	0	0	0	0	0	0
55	0	0	0	0	0	0	0	0	0	0	0	0	0	0
56	0	0	0	0	0	0	0	0	0	0	0	0	0	0

57	0	0	0	0	0	0	0	0	0	0	0	0	0	0
58	0	0	0	0	0	0	0	0	0	0	0	0	0	0
59	0	0	0	0	0	0	0	0	0	0	0	0	0	0
60	0	0	0	0	0	0	0	0	0	0	0	0	0	0
61	0	0	0	0	0	0	0	0	0	0	0	0	0	0
62	0	0	0	0	0	0	0	0	0	0	0	0	0	0
63	0	0	0	0	0	0	0	0	0	0	0	0	0	0
64	0	0	0	0	0	0	0	0	0	0	0	0	0	0
65	0	0	0	0	0	0	0	0	0	0	0	0	0	0
66	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	0	0	0	0	0	0	0	0	0	0	0	0	0
68	0	0	0	0	0	0	0	0	0	0	0	0	0	0
69	0	0	0	0	0	0	0	0	0	0	0	0	0	0
70	0	0	0	0	0	0	0	0	0	0	0	0	0	0
71	0	0	0	0	0	0	0	0	0	0	0	0	0	0
72	0	0	0	0	0	0	0	0	0	0	0	0	0	0
73	0	0	0	0	0	0	0	0	0	0	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	0	0	0	0	0	0	0	0	0	0	0	0	0	0
76	0	0	0	0	0	0	0	0	0	0	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	0	0	0	0	0	0	0	0	0	0	0	0	0	0
79	0	0	0	0	0	0	0	0	0	0	0	0	0	0
80	0	0	0	0	0	0	0	0	0	0	0	0	0	0
81	0	0	0	0	0	0	0	0	0	0	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0	0	0	0	0
83	0	0	0	0	0	0	0	0	0	0	0	0	0	0
84	0	0	0	0	0	0	0	0	0	0	0	0	0	0
85	0	1	0	0	0	0	0	0	0	0	0	0	0	0
86	0	0	0	0	0	0	0	0	0	0	0	0	0	0
87	0	0	0	0	0	0	0	0	0	0	0	0	0	0
88	0	0	0	0	0	0	0	0	0	0	0	0	0	0
89	0	0	0	0	0	0	0	0	0	0	0	0	0	0
90	0	0	0	0	0	0	0	0	0	0	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0	0	0	0	0	0
96	0	0	0	0	0	0	0	0	0	0	0	0	0	0
97	0	0	0	0	0	0	0	0	0	0	0	0	0	0
98	0	0	0	0	0	0	0	0	0	0	0	0	0	0
99	0	0	0	0	0	0	0	0	0	0	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	0	0	0	0	0	0	0	0	0	0	0	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	0	0	0	0	0	0	0	0	0	0	0	0	0	0
104	0	0	0	0	0	0	0	0	0	0	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	0	0	0	0	0
107	0	0	0	0	0	0	0	0	0	0	0	0	0	0
108	0	0	0	0	0	0	0	0	0	0	0	0	0	0
109	0	0	0	0	0	0	0	0	0	0	0	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	0	0	0	0	0	0	0	0	0	0	0	0	0	0
112	0	0	0	0	0	0	0	0	0	0	0	0	0	0
113	0	0	0	0	0	0	0	0	0	0	0	0	0	0
114	0	0	0	0	0	0	0	0	0	0	0	0	0	0
115	0	0	0	0	0	0	0	0	0	0	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	0	0	0	0	0	0	0	0	0	0	0	0	0	0
118	0	0	0	0	0	0	0	0	0	0	0	0	0	0
119	0	0	0	0	0	0	0	0	0	0	0	0	0	0

120	0	0	0	0	0	0	0	0	0	0	0	0	0	0
121	0	0	0	0	0	0	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	0	0	0	0	0	0	0	0	0	0	0	0	0	0
124	0	1	0	0	0	0	0	0	0	0	0	0	0	0
125	0	0	0	0	0	0	0	0	0	0	0	0	0	0
126	0	0	0	0	0	0	0	0	0	0	0	0	0	0
127	0	0	0	0	0	0	0	0	0	0	0	0	0	0
128	0	0	0	0	0	0	0	0	0	0	0	0	0	0
129	0	0	0	0	0	0	0	0	0	0	0	0	0	0
130	0	0	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	0	0	0	0	0	0	0	0	0	0
134	0	0	0	0	0	0	0	0	0	0	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	2	1	1	3	1	1	0	0	0	0	0	0	0	0
137	0	0	0	0	0	0	0	0	0	0	0	0	0	0
138	0	0	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	0	0	0	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	0	0	0	0	0	0	0	0
141	0	0	0	0	0	0	0	0	0	0	0	0	0	0
142	0	0	0	0	0	0	0	0	0	0	0	0	0	0
143	0	0	0	0	0	0	0	0	0	0	0	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	1	0	0	0	0	0	0	0	0	0	0	0	0	0
146	0	0	0	0	0	0	0	0	0	0	0	0	0	0
147	0	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	0	0	0	0	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	0	0	0	0	0	0	0	0	0	0	0	0	0	0
153	0	0	0	0	0	0	0	0	0	0	0	0	0	0
154	0	0	0	0	0	0	0	0	0	0	0	0	0	0
155	0	0	0	0	0	0	0	0	0	0	0	0	0	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	0	0	0	0	0	0	0	0	0	0	0	0	0
161	0	0	0	0	0	0	0	0	0	0	0	0	0	0
162	0	0	0	0	0	0	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	1	0	0	0	0	0	0	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	0	0	0	0	0	0	0	0	0	0	0	0	0	0
168	0	0	0	0	0	0	0	0	0	0	0	0	0	0
169	0	0	0	0	0	0	0	0	0	0	0	0	0	0
170	0	0	0	0	0	0	0	0	0	0	0	0	0	0
171	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Table 3f Relative abundance of silica (S) particles by size bin (nm) for each sample. (Values sum to total abundance in S class as shown in Table 2 for each sample.)

Sample No	Relative abundance of S particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	0	0	1	0	0	0	0	0	1	0	0	0	0
2	1	0	1	1	0	1	0	0	0	1	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	0	0	0	1	1	1	0	0	0	1	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	0	1	2	1	1	1	0	0	0	1	0	0	0	0
8	0	0	1	1	0	0	0	0	0	0	0	0	0	0
9	9	12	11	11	9	8	4	0	1	5	1	0	0	0
10	11	7	9	8	4	3	1	0	1	5	1	0	0	0
11	4	4	3	4	6	3	0	0	0	1	1	0	0	0
12	0	0	1	1	0	1	0	0	0	0	0	0	0	0
13	1	1	2	2	1	0	1	0	0	0	0	0	0	0
14	0	3	0	0	0	0	0	0	0	1	1	0	0	0
15	1	2	1	1	0	0	1	0	0	2	0	0	0	0
16	0	0	0	0	0	0	0	0	0	1	0	0	0	0
17	1	1	1	1	0	0	0	0	0	1	0	0	0	0
18	0	0	2	0	0	0	0	0	0	0	0	0	0	0
19	0	1	1	0	1	0	0	0	0	2	1	0	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	2	2	4	6	3	4	1	0	1	3	1	0	0	0
22	0	0	0	0	0	0	0	0	0	1	0	0	0	0
23	0	0	0	0	1	0	1	0	0	0	0	0	0	0
24	0	0	0	0	0	0	0	0	0	1	0	0	0	0
25	0	0	0	1	0	0	0	0	0	0	0	0	0	0
26	1	1	0	0	0	0	0	0	0	0	0	0	0	0
27	0	0	0	0	0	0	0	0	0	1	0	0	0	0
28	0	0	0	0	0	0	0	0	0	1	0	0	0	0
29	1	0	0	1	0	0	0	0	0	0	0	0	0	0
30	0	1	0	0	0	1	0	0	0	0	0	0	0	0
31	0	0	1	1	0	0	0	0	0	1	0	0	0	0
32	0	0	0	0	0	0	0	0	0	0	0	0	0	0
33	0	0	0	1	0	0	0	0	0	0	0	0	0	0
34	0	0	0	0	0	0	0	0	0	0	0	0	0	0
35	0	0	0	0	0	0	0	0	0	0	0	0	0	0
36	0	0	0	0	0	0	0	0	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	0	0	0	0
38	0	1	0	0	0	0	0	0	0	0	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	0	0	1	0	0	0	0	0	0	0	0	0	0
41	0	0	0	0	0	0	0	0	0	0	0	0	0	0
42	0	0	0	0	0	2	0	0	0	0	0	0	0	0
43	2	0	0	0	0	0	0	0	0	0	0	0	0	0
44	0	0	1	0	0	0	0	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	0	0	0	0	0	0	0	0	0	0	0	0
47	0	0	0	0	0	0	0	0	0	0	0	0	0	0
48	0	1	1	0	1	0	0	0	0	1	0	0	0	0
49	0	0	0	5	0	0	0	0	0	1	0	0	0	0
50	0	0	1	1	1	0	0	0	0	1	0	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	0	0	1	0	0	0	0	0	1	0	0	0	0
53	0	0	0	0	0	0	1	0	0	1	1	0	0	0
54	1	1	2	0	1	1	0	0	0	1	0	0	0	0
55	1	2	1	2	1	0	0	0	0	2	0	0	0	0
56	0	0	1	1	0	0	1	0	0	0	0	0	0	0

57	1	1	1	4	2	0	0	0	0	2	0	0	0	0
58	0	1	1	0	0	0	0	0	0	1	0	0	0	0
59	0	0	1	1	1	1	0	0	0	1	0	0	0	0
60	0	0	0	0	0	1	0	0	0	0	0	0	0	0
61	0	0	1	2	0	0	0	0	0	1	0	0	0	0
62	0	0	0	0	0	0	0	0	0	0	0	0	0	0
63	1	1	5	5	3	4	1	0	1	2	0	0	0	0
64	1	6	1	3	1	3	3	0	0	2	1	0	0	0
65	0	0	1	0	0	0	0	0	0	2	1	1	0	0
66	0	0	0	0	0	1	0	0	0	0	0	0	0	0
67	0	0	0	0	0	0	0	0	0	1	1	0	0	0
68	1	2	3	1	0	2	0	0	0	1	1	0	0	0
69	4	9	11	14	10	6	3	0	1	5	1	0	0	0
70	0	3	0	0	0	0	0	0	0	3	1	0	0	0
71	0	2	0	4	3	2	1	0	0	2	0	0	0	0
72	1	3	3	3	5	2	0	0	1	5	1	0	0	0
73	2	2	3	3	3	1	1	0	0	3	1	0	0	0
74	9	8	6	12	9	6	3	0	1	9	2	1	0	0
75	2	3	4	3	3	3	3	0	1	3	0	0	0	0
76	2	4	4	4	5	2	1	0	1	4	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	1	1	0	1	1	0	0	0	0	0	0	0	0	0
79	0	1	0	1	1	1	1	0	0	1	0	0	0	0
80	0	0	1	1	1	1	1	0	0	1	0	0	0	0
81	0	0	0	0	0	0	0	0	0	0	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0	0	0	0	0
83	1	0	1	0	1	1	0	0	0	1	0	0	0	0
84	0	0	0	1	1	0	0	0	0	0	0	0	0	0
85	0	0	0	0	1	0	0	0	0	0	0	0	0	0
86	0	2	1	0	0	0	0	0	0	2	0	0	0	0
87	0	0	0	1	1	0	1	0	0	1	0	0	0	0
88	0	1	0	0	0	1	0	0	0	1	1	0	0	0
89	0	0	0	1	1	1	1	0	0	1	0	0	0	0
90	0	0	0	0	0	0	0	0	0	0	0	0	0	0
91	0	0	0	1	1	0	0	0	0	0	0	0	0	0
92	0	0	2	1	1	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	0	0	1	1	0	1	1	0	0	1	0	0	0	0
96	0	0	1	0	0	1	0	0	0	1	0	0	0	0
97	0	0	0	0	0	0	0	0	0	0	0	0	0	0
98	0	0	0	0	1	0	0	0	0	1	0	0	0	0
99	0	0	0	0	0	0	0	0	0	0	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	0	0	0	0	2	1	0	0	0	1	0	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	0	1	1	1	1	0	0	0	0	0	0	0	0	0
104	0	0	0	0	0	0	0	0	0	1	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	0	0	0	0	0
107	0	0	0	0	0	0	0	0	0	1	0	0	0	0
108	0	0	0	1	0	1	0	0	0	1	0	0	0	0
109	0	1	0	0	0	1	0	0	0	0	0	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	0	0	0	0	1	1	0	0	0	1	0	0	0	0
112	0	0	0	0	0	0	0	0	0	0	0	0	0	0
113	0	0	0	0	0	0	0	0	0	0	0	0	0	0
114	0	0	0	0	0	0	0	1	0	0	0	0	0	0
115	0	0	1	0	1	0	0	0	0	0	0	0	0	0
116	0	1	1	1	1	1	1	0	0	0	0	0	0	0
117	0	1	0	2	1	1	0	0	0	2	0	0	0	0
118	0	0	0	0	0	0	0	0	0	0	0	0	0	0
119	0	0	0	1	0	0	0	0	0	0	0	0	0	0

120	0	0	1	1	0	0	0	0	0	0	0	0	0	0
121	0	0	0	0	0	0	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	0	0	1	0	0	0	0	0	0	0	0	0	0	0
124	1	2	0	1	1	1	1	0	0	0	0	0	0	0
125	0	0	0	0	0	0	0	0	0	0	0	0	0	0
126	0	1	2	1	1	2	0	0	0	2	0	0	0	0
127	1	1	1	2	1	0	0	0	0	0	0	0	0	0
128	0	1	1	1	1	1	0	0	0	0	0	0	0	0
129	1	1	0	1	0	0	0	0	0	0	0	0	0	0
130	0	0	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	1	0	0	0	0	0	0	0	0	0
134	0	0	0	1	1	0	0	0	0	0	0	0	0	0
135	2	1	2	1	1	1	0	0	0	0	0	0	0	0
136	0	0	0	0	0	0	0	0	0	0	0	0	0	0
137	0	0	0	0	0	0	0	0	0	0	0	0	0	0
138	0	0	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	0	0	0	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	0	0	0	0	0	0	0	0
141	0	0	0	0	0	0	0	0	0	0	1	0	0	0
142	0	0	0	0	0	0	0	1	0	1	1	1	0	0
143	0	0	0	0	0	0	0	0	1	0	0	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	0	0	0	0	0	0	0	0	0	0	0	0	0
146	0	0	0	0	0	0	0	1	0	0	0	0	0	0
147	0	0	0	0	0	0	0	0	0	1	0	0	0	0
148	1	0	0	0	0	0	0	0	0	0	0	0	0	0
149	1	2	2	1	1	1	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	1	0	0	1	1	1	0	0	0	0	0	0	0	0
152	0	0	1	0	0	1	1	0	0	1	0	0	0	0
153	0	0	0	1	1	1	0	0	0	1	0	0	0	0
154	0	0	1	0	0	0	0	0	0	1	0	0	0	0
155	1	1	0	0	1	2	1	0	0	0	0	0	0	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	0	0	0	0	0	0	0	0	0	0	0	0	0
161	2	0	1	0	1	0	0	0	0	0	0	0	0	0
162	16	21	13	8	10	4	2	0	1	6	1	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	0	0	0	0	0	0	0	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	2	1	1	0	0	0	0	0	0	0	0	0	0	0
168	2	1	1	0	1	0	0	0	0	0	0	0	0	0
169	1	1	1	1	1	1	0	0	0	0	0	0	0	0
170	0	1	1	1	0	0	0	0	0	0	0	0	0	0
171	0	1	0	1	1	0	0	0	0	0	0	0	0	0

Table 3g Relative abundance of other heavy mineral (M) particles by size bin (nm) for each sample. (Values sum to total abundance in M class as shown in Table 2 for each sample.)

Sample No	Relative abundance of M particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	0	0	0	0	1	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	0	0	0	0	0	0	0	0	0	0	0	0	0	0
8	0	0	0	0	0	0	0	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
11	1	0	0	0	0	0	0	0	0	0	0	0	0	0
12	0	0	0	1	0	0	0	0	0	0	0	0	0	0
13	0	0	0	0	0	0	0	0	0	0	0	0	0	0
14	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15	0	0	0	0	0	0	0	0	0	0	0	0	0	0
16	0	3	0	0	0	3	0	0	0	0	0	0	0	0
17	0	0	0	0	0	0	0	0	0	0	0	0	0	0
18	2	0	0	0	0	0	0	0	0	0	0	0	0	0
19	0	0	0	0	1	0	0	0	0	0	0	0	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	0	0	0	0	0	0	0	0	0	0	0	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	0	0	0	0	0	0	0	0	0	0	0	0	0	0
24	0	0	0	0	0	0	0	0	0	0	0	0	0	0
25	0	0	0	1	0	0	0	0	0	0	0	0	0	0
26	0	0	0	0	0	0	0	0	0	0	0	0	0	0
27	0	0	0	0	0	0	0	0	0	0	0	0	0	0
28	0	0	0	0	0	0	0	0	0	0	0	0	0	0
29	0	0	1	0	1	0	0	0	0	0	0	0	0	0
30	0	0	0	0	0	0	0	0	0	0	0	0	0	0
31	0	0	0	0	0	0	0	0	0	0	0	0	0	0
32	0	0	1	1	1	0	0	0	0	0	0	0	0	0
33	0	0	1	1	1	0	0	0	0	0	0	0	0	0
34	0	0	0	0	1	0	0	0	0	0	0	0	0	0
35	0	0	0	0	1	0	0	0	0	0	0	0	0	0
36	0	0	0	0	0	0	0	0	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	0	0	0	0
38	0	0	0	0	0	0	0	0	0	0	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	1	0	0	1	0	0	0	0	0	0	0	0	0
41	0	0	0	0	0	0	0	0	0	0	0	0	0	0
42	0	0	0	0	2	0	0	0	0	0	0	0	0	0
43	0	0	0	0	0	0	0	0	0	0	0	0	0	0
44	0	0	0	0	0	0	0	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	0	0	0	0	0	0	0	0	0	0	0	0
47	0	0	0	3	0	0	0	0	0	0	0	0	0	0
48	0	2	1	0	1	0	0	0	0	0	0	0	0	0
49	0	0	0	0	0	0	0	0	0	0	0	0	0	0
50	0	0	0	0	0	0	0	0	0	0	0	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	0	0	0	0	0	1	0	0	1	0	0	0	0
53	0	0	0	0	0	1	0	0	0	0	0	0	0	0
54	0	0	0	0	0	0	0	0	0	0	0	0	0	0
55	0	0	0	0	0	0	0	0	0	0	0	0	0	0
56	0	0	0	0	0	0	0	0	0	0	0	0	0	0

57	0	0	0	0	0	0	0	0	0	0	0	0	0	0
58	0	0	0	0	0	0	0	0	0	0	0	0	0	0
59	0	0	0	0	0	0	0	0	0	0	0	0	0	0
60	0	0	0	0	0	0	0	0	0	0	0	0	0	0
61	0	0	0	0	0	0	0	0	0	0	0	0	0	0
62	0	0	0	0	0	0	0	0	0	0	0	0	0	0
63	0	0	0	0	0	0	0	0	0	0	0	0	0	0
64	0	0	0	0	0	0	0	0	0	0	0	0	0	0
65	0	0	0	1	0	0	0	0	0	0	0	0	0	0
66	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	0	0	0	0	0	0	0	0	0	0	0	0	0
68	0	0	0	0	0	0	0	0	0	0	0	0	0	0
69	0	0	0	0	0	0	0	0	0	0	0	0	0	0
70	0	0	0	0	0	0	0	0	0	0	0	0	0	0
71	0	0	0	0	0	0	0	0	0	0	0	0	0	0
72	0	0	0	0	0	0	0	0	0	0	0	0	0	0
73	0	0	0	0	0	0	0	0	0	0	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	0	0	0	0	0	0	0	0	0	0	0	0	0	0
76	0	0	0	0	0	0	0	0	0	0	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	0	0	0	1	0	1	0	0	0	1	0	0	0	0
79	0	0	0	0	1	1	0	0	0	0	0	0	0	0
80	0	1	0	1	0	0	0	0	0	0	0	0	0	0
81	0	1	0	0	0	0	0	0	0	0	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0	0	0	0	0
83	0	0	0	0	0	0	0	0	0	0	0	0	0	0
84	0	0	0	0	0	0	0	0	0	0	0	0	0	0
85	0	1	1	1	0	0	0	0	0	0	0	0	0	0
86	0	0	0	0	0	0	0	0	0	0	0	0	0	0
87	0	0	0	0	1	0	0	0	0	0	0	0	0	0
88	0	0	0	0	1	1	0	0	0	0	0	0	0	0
89	0	0	0	1	0	0	0	0	0	0	0	0	0	0
90	0	0	1	1	0	1	0	0	0	0	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0	0	0	0	0
93	1	0	1	1	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	1	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0	0	0	0	0	0
96	0	0	0	0	0	0	0	0	0	0	0	0	0	0
97	0	0	3	0	0	0	0	0	0	0	0	0	0	0
98	0	0	0	0	0	0	0	0	0	0	0	0	0	0
99	0	0	0	0	0	0	0	0	0	0	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	0	0	1	0	1	0	0	0	0	0	0	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	0	0	0	0	0	0	0	0	0	0	0	0	0	0
104	1	0	1	1	1	0	0	0	0	0	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	0	0	0	0	0
107	1	0	0	0	0	0	0	0	0	0	0	0	0	0
108	0	0	0	0	0	0	0	0	0	0	0	0	0	0
109	0	0	0	0	0	0	0	0	0	0	0	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	0	0	0	0	1	0	0	0	0	0	0	0	0	0
112	0	0	0	0	0	0	0	0	0	0	0	0	0	0
113	0	1	0	0	0	0	0	0	0	0	0	0	0	0
114	0	0	0	1	0	1	0	0	0	0	0	0	0	0
115	0	0	0	0	0	1	0	0	0	0	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	0	0	0	0	0	0	0	0	0	0	0	0	0	0
118	0	0	0	0	0	0	0	0	0	0	0	0	0	0
119	2	1	2	4	0	2	0	0	0	0	0	0	0	0

120	0	0	2	0	0	1	0	0	0	0	0	0	0	0
121	0	0	0	0	0	0	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	0	0	0	1	0	0	0	0	0	0	0	0	0	0
124	1	0	0	0	0	0	0	0	0	0	0	0	0	0
125	0	0	0	0	0	0	0	0	0	0	0	0	0	0
126	0	0	0	0	0	0	0	0	0	0	0	0	0	0
127	0	0	0	0	0	0	0	0	0	0	0	0	0	0
128	0	0	0	0	0	0	1	0	0	0	0	0	0	0
129	0	0	0	0	0	0	0	0	0	0	0	0	0	0
130	0	0	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	0	0	0	0	0	0	0	0	0	0
134	0	0	0	0	0	0	0	0	0	0	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	0	0	0	0	0	0	0	0	0	0	0	0	0	0
137	0	0	1	0	0	0	0	0	0	0	0	0	0	0
138	0	1	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	0	0	1	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	0	0	0	0	0	0	0	0
141	0	0	0	0	0	0	0	0	0	0	0	0	0	0
142	0	0	0	0	0	0	0	0	0	0	0	0	0	0
143	0	0	0	0	0	0	0	0	0	0	0	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	0	0	0	0	0	0	0	0	0	1	0	0	0
146	0	0	0	0	0	0	0	0	0	0	0	0	0	0
147	1	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	0	0	0	0	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	0	1	1	2	1	0	0	0	0	0	0	0	0	0
153	0	0	0	0	0	0	0	0	0	0	0	0	0	0
154	0	0	0	0	1	0	0	0	0	0	0	0	0	0
155	0	0	0	0	0	0	0	0	0	0	0	0	0	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	0	0	1	0	0	0	0	0	0	0	0	0	0
161	0	0	0	0	0	0	0	0	0	0	0	0	0	0
162	0	0	0	0	0	0	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	0	0	0	0	0	0	0	0	0	0	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	0	0	0	0	0	0	0	0	0	0	0	0	0	0
168	0	1	0	0	1	1	0	0	0	0	0	0	0	0
169	0	0	0	0	0	0	0	0	0	0	0	0	0	0
170	0	0	0	0	0	1	0	0	0	0	0	0	0	0
171	0	0	0	1	0	0	0	0	0	0	0	0	0	0

Table 3h Relative abundance of carbonate (CB) particles by size bin (nm) for each sample. (Values sum to total abundance in CB class as shown in Table 2 for each sample.)

Sample No	Relative abundance of CB particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	0	0	0	1	1	0	0	0	3	1	0	0	0
2	0	0	0	0	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	4	9	0	0	3	12	1	0	0	0
4	0	0	0	0	0	1	0	0	0	1	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	2	1	0	0	0
7	0	0	0	0	1	0	0	0	0	0	0	0	0	0
8	3	4	3	4	3	2	0	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
11	1	0	0	0	0	0	0	0	0	0	0	0	0	0
12	0	0	0	0	0	0	0	0	0	0	0	0	0	0
13	0	0	0	0	0	0	0	0	0	0	0	0	0	0
14	0	0	0	3	0	3	0	0	1	6	0	0	0	0
15	0	0	0	0	0	0	0	0	0	0	0	0	0	0
16	3	3	0	0	3	0	0	0	1	3	0	0	0	0
17	0	0	0	0	0	0	0	0	0	0	0	0	0	0
18	0	0	0	0	0	0	0	0	0	0	0	0	0	0
19	0	0	0	0	1	0	0	0	0	1	0	0	0	0
20	0	0	0	0	5	5	0	0	1	9	1	0	0	0
21	0	0	0	0	0	0	0	0	0	0	0	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	0	0	0	0	0	0	0	0	0	1	0	0	0	0
24	0	0	4	4	0	4	4	0	1	4	2	1	0	0
25	0	0	1	4	0	3	0	0	0	8	4	1	0	0
26	2	3	4	7	8	4	3	1	1	10	5	2	1	0
27	0	0	0	1	0	1	0	0	0	2	1	0	0	0
28	0	1	1	1	2	1	0	0	0	3	1	1	0	0
29	2	2	4	3	5	3	3	1	1	9	2	1	0	0
30	2	3	3	4	7	5	1	1	1	8	2	0	0	0
31	2	4	4	5	2	3	1	0	2	14	8	3	2	0
32	7	10	7	12	9	4	4	0	2	12	2	1	0	0
33	3	3	5	4	4	2	0	0	0	3	1	1	0	0
34	6	10	8	15	12	8	3	0	3	14	3	1	0	0
35	1	0	1	1	1	1	0	0	1	4	1	0	0	0
36	6	14	11	8	9	9	2	0	1	6	4	3	2	1
37	1	4	9	11	11	11	4	1	4	20	4	1	0	0
38	3	3	4	5	4	1	1	0	0	1	1	0	0	0
39	3	5	4	7	9	4	1	0	2	8	5	3	2	2
40	1	1	1	2	6	2	0	0	1	7	2	0	0	0
41	1	3	0	0	3	0	0	1	1	5	3	2	2	1
42	0	2	0	0	0	0	0	0	1	3	1	0	0	0
43	0	4	0	0	0	0	0	0	0	2	1	0	0	0
44	0	0	0	0	0	0	0	0	0	2	1	0	0	0
45	0	0	0	6	0	3	0	0	0	2	1	0	0	0
46	0	0	0	0	1	1	0	0	0	3	1	0	0	0
47	0	0	5	5	5	3	3	0	0	3	1	1	0	0
48	0	0	0	0	0	1	0	0	0	1	0	0	0	0
49	0	0	11	0	0	0	0	0	1	3	2	1	0	0
50	0	0	0	0	0	0	0	0	0	0	0	0	0	0
51	2	0	0	0	4	0	0	0	0	3	1	0	0	0
52	0	0	1	2	2	2	0	0	1	14	5	1	0	0
53	0	1	0	2	1	1	2	0	0	2	1	0	0	0
54	0	1	0	1	0	0	0	0	0	1	1	0	0	0
55	1	1	1	1	0	0	0	0	0	1	0	0	0	0
56	1	0	1	2	1	1	1	0	1	3	1	0	0	0

57	0	0	0	0	0	0	0	0	0	0	0	0	0	0
58	1	1	1	0	1	0	0	0	0	4	1	0	0	0
59	0	0	0	0	0	0	0	0	0	1	1	0	0	0
60	0	1	1	0	1	0	0	0	0	1	0	0	0	0
61	0	0	0	1	1	0	0	0	0	1	0	0	0	0
62	3	8	10	14	11	6	3	0	3	11	2	0	0	0
63	0	0	0	0	0	0	0	0	0	0	0	0	0	0
64	0	3	1	0	1	0	0	0	0	2	2	2	1	0
65	0	0	0	1	0	1	0	0	0	2	0	0	0	0
66	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	0	0	3	0	0	0	1	0	2	1	1	0	0
68	0	0	0	0	0	0	0	0	0	0	0	0	0	0
69	0	0	0	0	0	0	0	0	0	0	0	0	0	0
70	2	0	0	0	0	0	0	0	0	4	2	2	1	0
71	0	0	0	0	0	0	0	0	0	1	1	0	0	0
72	0	0	0	0	0	0	0	0	0	1	0	0	0	0
73	0	0	0	0	0	0	0	0	0	0	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	0	0	0	0	0	0	0	0	0	0	0	0	0	0
76	0	0	0	0	0	0	0	0	0	0	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	0	0	0	0	1	0	0	1	0	1	0	0	0	0
79	0	0	0	0	0	0	0	0	0	0	0	0	0	0
80	0	0	0	0	0	0	0	0	0	0	0	0	0	0
81	0	0	1	1	0	0	0	0	0	1	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0	0	0	0	0
83	0	0	0	1	0	0	0	0	0	1	0	0	0	0
84	0	0	0	0	0	1	0	0	0	1	1	0	0	0
85	0	0	0	0	1	1	0	0	0	2	1	0	0	0
86	0	0	0	0	1	0	0	0	0	1	0	0	0	0
87	0	0	0	0	0	1	0	0	0	1	0	0	0	0
88	0	0	1	0	1	0	0	0	0	1	0	0	0	0
89	1	1	1	1	1	1	0	0	0	3	1	0	0	0
90	0	0	1	1	1	1	0	0	0	1	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0	0	0	0	0	0
96	1	0	0	0	0	0	0	0	0	1	1	0	0	0
97	0	0	0	0	0	0	0	0	0	4	2	0	0	0
98	0	0	0	0	0	0	0	0	0	0	0	0	0	0
99	0	1	0	0	1	0	0	0	0	3	2	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	0	0	0	0	0	0	0	0	0	0	0	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	0	0	0	0	0	0	0	0	0	0	0	0	0	0
104	0	0	0	0	0	1	0	0	0	0	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	1	1	0	0	0
107	0	0	0	0	0	1	1	0	0	0	0	0	0	0
108	0	0	0	0	0	0	0	0	0	0	0	0	0	0
109	0	0	0	0	0	0	0	0	0	0	0	0	0	0
110	0	0	0	0	0	0	0	0	0	1	1	0	0	0
111	0	0	0	0	0	0	0	0	0	1	1	0	0	0
112	0	0	0	0	0	0	0	0	0	1	1	0	0	0
113	0	2	3	1	2	2	4	0	0	8	4	1	0	0
114	0	0	0	1	1	1	1	0	0	1	1	0	0	0
115	0	0	0	0	0	0	0	0	0	0	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	2	0	0	1	2	1	2	0	0	4	1	0	0	0
118	1	1	2	3	4	5	1	0	1	9	2	2	1	0
119	0	0	0	0	0	1	0	0	0	0	0	0	0	0

120	0	0	0	0	0	1	0	0	0	0	0	0	0	0
121	0	0	0	0	0	0	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	0	1	1	2	1	1	1	0	0	1	0	0	0	0
124	1	1	1	1	1	0	0	0	0	1	0	0	0	0
125	1	0	0	1	1	0	0	0	0	1	1	0	0	0
126	0	0	0	0	0	0	0	0	0	0	0	0	0	0
127	0	0	0	0	0	0	0	0	0	0	0	0	0	0
128	0	0	0	0	0	0	0	0	0	0	0	0	0	0
129	0	1	1	0	1	0	1	0	0	0	0	0	0	0
130	1	1	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	0	0	0	0	0	1	0	0	0	0
134	2	4	3	3	1	1	1	0	0	1	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	1	2	0	1	1	0	0	0	1	3	1	0	0	0
137	0	0	0	0	0	0	0	0	0	1	0	0	0	0
138	0	0	1	2	3	1	2	0	1	7	6	1	1	1
139	0	0	0	0	0	0	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	0	0	0	1	1	0	0	0
141	0	0	0	0	0	0	0	0	0	0	0	0	0	0
142	0	0	0	0	0	0	0	0	0	0	0	0	0	0
143	0	0	0	0	0	0	0	0	0	0	0	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	0	0	0	0	0	0	0	0	0	0	0	0	0
146	0	0	0	0	0	0	0	0	0	0	0	0	0	0
147	0	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	0	0	0	0	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	0	1	0	0	0	0	0	0	0	0	0	0	0	0
153	0	0	0	0	0	0	0	0	0	1	0	0	0	0
154	0	0	0	0	1	0	0	0	0	3	1	0	0	0
155	0	0	0	0	0	0	0	0	0	0	0	0	0	0
156	0	0	0	1	0	0	0	0	0	0	0	0	0	0
157	5	1	2	2	2	1	1	0	0	1	0	0	0	0
158	0	1	1	1	0	0	0	0	0	1	0	0	0	0
159	5	2	3	2	5	3	1	0	1	6	1	1	0	0
160	4	7	8	12	10	6	2	0	2	10	3	1	0	0
161	0	1	0	0	0	0	0	0	0	0	0	0	0	0
162	0	0	0	0	0	0	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	5	7	12	11	12	7	2	0	1	6	0	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	1	1	1	0	0	0	0	0	0	1	0	0	0	0
168	1	0	0	0	0	0	0	0	0	0	0	0	0	0
169	0	0	0	0	0	0	0	0	0	0	0	0	0	0
170	0	0	0	0	0	0	0	0	0	0	0	0	0	0
171	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Table 3i Relative abundance of other (O) particles by size bin (nm) for each sample. (Values sum to total abundance in O class as shown in Table 2 for each sample.)

Sample No	Relative abundance of O particles (number %)													
	Sub									Supra				
	[87, 200)	[200, 300)	[300, 400)	[400, 500)	[500, 600)	[600, 700)	[700, 800)	[800, 900)	[900, 1000)	[1000, 2000)	[2000, 3000)	[3000, 4000)	[4000, 5000)	>=5000
1	0	0	1	0	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0	0	0	0
7	0	0	0	0	0	0	0	0	0	0	0	0	0	0
8	0	2	1	0	1	1	0	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
11	0	0	0	0	0	0	0	0	0	0	0	0	0	0
12	0	0	0	0	0	0	0	0	0	0	0	0	0	0
13	0	0	0	0	0	0	0	0	0	0	0	0	0	0
14	0	0	0	0	0	0	0	0	0	0	0	0	0	0
15	0	0	0	0	0	0	0	0	0	0	0	0	0	0
16	0	0	0	0	0	0	0	0	0	0	0	0	0	0
17	0	0	0	0	0	0	0	0	0	0	0	0	0	0
18	0	0	0	0	0	0	0	0	0	0	0	0	0	0
19	0	0	0	0	0	0	0	0	0	0	0	0	0	0
20	0	0	0	0	0	0	0	0	0	0	0	0	0	0
21	0	0	0	0	0	0	0	0	0	0	0	0	0	0
22	0	0	0	0	0	0	0	0	0	0	0	0	0	0
23	0	0	0	0	0	0	0	0	0	0	0	0	0	0
24	0	0	0	0	0	0	0	0	0	0	0	0	0	0
25	0	0	0	3	0	0	0	0	0	0	0	0	0	0
26	0	0	0	0	1	0	0	0	0	0	0	0	0	0
27	0	0	0	0	0	0	0	0	0	0	0	0	0	0
28	0	0	0	0	0	0	0	0	0	0	0	0	0	0
29	0	0	0	0	0	0	1	1	0	0	0	0	0	0
30	0	0	0	0	1	0	0	0	0	0	0	0	0	0
31	0	0	1	1	0	1	0	0	0	0	1	1	0	0
32	0	2	2	2	4	1	1	1	0	0	2	1	0	0
33	0	0	0	0	0	0	0	0	0	0	0	0	0	0
34	0	0	3	2	1	1	0	0	0	0	0	0	0	0
35	0	0	0	0	0	0	0	0	0	0	0	0	0	0
36	0	0	0	2	0	0	1	0	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	1	0	0	0
38	0	0	0	0	0	0	0	1	0	0	0	0	0	0
39	0	0	0	0	0	0	0	0	0	0	0	0	0	0
40	0	0	0	0	0	0	0	0	0	0	0	0	0	0
41	0	0	0	0	0	0	0	0	0	0	0	0	0	0
42	0	0	0	0	0	0	0	0	0	0	0	0	0	0
43	0	0	0	0	0	0	0	0	0	0	0	0	0	0
44	0	0	0	0	0	0	0	0	0	0	0	0	0	0
45	0	0	0	0	0	0	0	0	0	0	0	0	0	0
46	0	0	0	0	0	0	0	0	0	0	0	0	0	0
47	0	0	0	0	0	0	0	0	0	0	0	0	0	0
48	0	0	0	0	0	0	0	0	0	0	0	0	0	0
49	0	0	0	0	0	0	0	0	0	0	0	0	0	0
50	0	0	0	0	0	0	0	0	0	0	0	0	0	0
51	0	0	0	0	0	0	0	0	0	0	0	0	0	0
52	0	0	0	0	0	0	0	0	0	0	0	0	0	0
53	0	0	0	0	0	0	0	0	0	0	0	0	0	0
54	0	0	1	0	0	0	0	0	0	0	0	0	0	0
55	0	1	1	0	0	0	0	0	0	0	0	0	0	0
56	0	0	0	0	0	0	0	0	0	0	0	0	0	0

57	0	0	0	0	0	0	0	0	0	0	0	0	0	0
58	0	0	0	0	0	0	0	0	0	0	0	0	0	0
59	0	0	0	0	0	0	0	0	0	0	0	0	0	0
60	0	0	0	0	0	0	0	0	0	0	0	0	0	0
61	0	0	0	0	0	0	0	0	0	0	0	0	0	0
62	0	1	1	2	1	1	1	0	0	1	6	2	0	0
63	0	0	0	0	0	0	0	0	0	0	0	0	0	0
64	0	0	0	0	0	0	0	0	0	0	1	0	0	0
65	0	0	0	0	0	0	0	0	0	0	0	0	0	0
66	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	0	0	0	0	0	0	0	0	0	0	0	0	0	0
68	0	0	0	0	0	0	0	0	0	0	0	0	0	0
69	0	0	0	0	0	0	0	0	0	0	0	0	0	0
70	0	0	0	0	0	0	0	0	0	0	0	1	0	0
71	0	0	0	0	0	0	0	0	0	0	0	0	0	0
72	0	0	0	0	0	0	0	0	0	0	0	0	0	0
73	0	0	0	0	0	0	0	0	0	0	0	0	0	0
74	0	0	0	0	0	0	0	0	0	0	0	0	0	0
75	0	0	0	0	0	0	0	0	0	0	0	0	0	0
76	0	0	0	0	0	0	0	0	0	0	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0	0	0	0	0
78	0	0	0	0	0	0	0	0	0	0	1	0	0	0
79	0	0	0	0	0	0	0	0	0	0	0	0	0	0
80	0	0	0	0	0	0	0	0	0	0	0	0	0	0
81	0	1	0	0	0	0	0	0	0	0	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0	0	0	0	0
83	0	0	0	0	0	0	0	0	0	0	0	0	0	0
84	0	0	0	0	0	0	0	0	0	0	0	0	0	0
85	0	0	0	0	0	0	0	0	0	0	0	0	0	0
86	0	0	0	0	0	0	0	0	0	0	0	0	0	0
87	0	0	0	0	0	0	0	0	0	0	0	0	0	0
88	0	0	0	0	0	0	0	0	0	0	0	0	0	0
89	0	0	0	0	0	0	1	0	0	0	1	0	0	0
90	0	0	0	0	0	0	0	0	0	0	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0	0	0	0	0	0
96	0	0	0	0	0	0	0	0	0	0	0	0	0	0
97	0	0	0	0	0	0	0	0	0	0	0	0	0	0
98	0	0	0	0	0	0	0	0	0	0	0	0	0	0
99	0	0	0	0	0	0	0	0	0	0	0	0	0	0
100	0	0	0	0	0	0	0	0	0	0	0	0	0	0
101	0	0	0	0	0	0	0	0	0	0	0	0	0	0
102	0	0	0	0	0	0	0	0	0	0	0	0	0	0
103	0	0	1	1	0	0	0	0	0	0	0	0	0	0
104	0	0	0	0	0	0	0	0	0	0	0	0	0	0
105	0	0	0	0	0	0	0	0	0	0	0	0	0	0
106	0	0	0	0	0	0	0	0	0	0	0	0	0	0
107	0	0	0	0	0	0	0	0	0	0	0	0	0	0
108	0	0	0	0	0	0	0	0	0	0	0	0	0	0
109	0	0	0	0	0	0	0	0	0	0	0	0	0	0
110	0	0	0	0	0	0	0	0	0	0	0	0	0	0
111	0	0	0	0	0	0	0	0	0	0	0	0	0	0
112	0	0	0	0	0	0	0	0	0	0	0	0	0	0
113	0	0	0	0	0	0	0	0	0	0	0	0	0	0
114	0	0	0	0	0	0	0	0	0	0	0	0	0	0
115	0	0	1	0	0	0	0	0	0	0	0	0	0	0
116	0	0	0	0	0	0	0	0	0	0	0	0	0	0
117	0	0	0	0	0	0	0	0	0	0	0	0	0	0
118	0	0	0	0	0	0	1	0	0	0	0	0	0	0
119	0	0	0	1	0	0	0	0	0	0	1	0	0	0

120	0	0	0	0	0	0	0	0	0	0	0	0	0	0
121	0	0	0	0	0	0	0	0	0	0	0	0	0	0
122	0	0	0	0	0	0	0	0	0	0	0	0	0	0
123	0	0	0	0	0	0	0	0	0	0	0	0	0	0
124	0	0	0	0	1	0	0	0	0	0	0	0	0	0
125	0	0	0	0	0	0	0	0	0	0	0	0	0	0
126	0	0	0	0	0	0	0	0	0	0	0	0	0	0
127	0	0	0	0	0	0	0	0	0	0	0	0	0	0
128	0	0	0	0	0	0	0	0	0	0	0	0	0	0
129	0	0	0	0	0	0	0	0	0	0	0	0	0	0
130	0	0	0	0	0	0	0	0	0	0	0	0	0	0
131	0	0	0	0	0	0	0	0	0	0	0	0	0	0
132	0	0	0	0	0	0	0	0	0	0	0	0	0	0
133	0	0	0	0	0	0	0	0	0	0	0	0	0	0
134	0	0	1	0	1	0	0	0	0	0	0	0	0	0
135	0	0	0	0	0	0	0	0	0	0	0	0	0	0
136	0	6	8	9	8	8	6	0	0	2	11	2	1	0
137	0	0	0	0	0	0	0	0	0	0	0	0	0	0
138	0	0	0	0	0	0	0	0	0	0	0	0	0	0
139	0	0	0	0	0	0	0	0	0	0	0	0	0	0
140	0	0	0	0	0	0	1	0	0	0	0	0	0	0
141	0	0	0	0	0	0	0	0	0	0	0	0	0	0
142	0	0	0	0	0	0	0	0	0	0	0	0	0	0
143	0	0	0	0	0	0	0	0	0	0	0	0	0	0
144	0	0	0	0	0	0	0	0	0	0	0	0	0	0
145	0	0	0	0	0	0	0	0	0	0	0	0	0	0
146	0	0	0	0	0	0	0	0	0	0	0	0	0	0
147	0	0	0	0	0	0	0	0	0	0	0	0	0	0
148	0	0	0	0	0	0	0	0	0	0	0	0	0	0
149	0	0	0	0	0	0	0	0	0	0	0	0	0	0
150	0	0	0	0	0	0	0	0	0	0	0	0	0	0
151	0	0	0	0	0	0	0	0	0	0	0	0	0	0
152	0	0	0	0	0	0	0	0	0	0	0	0	0	0
153	0	0	0	0	0	0	0	0	0	0	0	0	0	0
154	0	0	0	0	0	0	0	0	0	0	0	0	0	0
155	0	0	0	0	0	0	0	0	0	0	0	0	0	0
156	0	0	0	0	0	0	0	0	0	0	0	0	0	0
157	0	0	0	0	0	0	0	0	0	0	0	0	0	0
158	0	0	0	0	0	0	0	0	0	0	0	0	0	0
159	0	0	0	0	0	0	0	0	0	0	0	0	0	0
160	0	0	1	0	1	0	0	0	0	0	0	0	0	0
161	0	0	0	0	0	0	0	0	0	0	0	0	0	0
162	0	0	0	0	0	0	0	0	0	0	0	0	0	0
163	0	0	0	0	0	0	0	0	0	0	0	0	0	0
164	0	0	1	2	2	2	3	1	0	0	1	0	0	0
165	0	0	0	0	0	0	0	0	0	0	0	0	0	0
166	0	0	0	0	0	0	0	0	0	0	0	0	0	0
167	0	0	0	0	0	0	0	0	0	0	0	0	0	0
168	0	0	0	0	0	0	0	0	0	0	0	0	0	0
169	0	0	0	0	0	0	0	0	0	0	0	0	0	0
170	0	0	0	0	0	0	0	0	0	0	0	0	0	0
171	0	0	0	0	0	0	0	0	0	0	0	0	0	0

Table 4a. Mass concentrations of potentially bioaccessible (SLF) and total acid-soluble (total) Mg, Al, Si, K, V, Cr, Fe, Mn, Co, Ni estimated for samples collected in Mines 10-24. (Sample numbers here are the same as the paired samples used for SEM-EDX analysis, and thus can be cross-referenced with the sample numbers shown in Tables 1-3.) Elemental concentrations are reported on dry mass basis (i.e., µg per g of respirable dust). Sample mass values indicate dust mass recovered from the sample filter. Mass concentrations below the method reporting level (MRL) are shown in red.

Sample No	Mass (µg)	Mass concentration (µg/g)																			
		Mg		Al		Si		K		V		Cr		Fe		Mn		Co		Ni	
		SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total
91	163	2418	8576	1489	51395	0	13436	0	16332	47	294	12	88	2113	31189	256	529	3	12	84	205
92	395	5076	5483	8918	20504	922	5711	1240	4641	21	123	0	30	18666	24127	523	541	10	11	80	80
93	202	4025	5056	6178	27939	0	5818	1264	7975	0	327	0	179	11655	25427	515	839	6	12	15	29
94	69	960	960	0	19902	0	2991	0	2997	0	398	0	126	0	23549	54	54	1	6	212	261
95	673	1741	2516	6466	22132	0	1320	824	5172	0	58	559	600	17283	21601	316	336	6	9	95	105
96	248	2251	2251	6222	14210	0	4374	256	2206	0	90	24	77	9169	12758	295	295	7	9	86	135
97	28	183	183	10422	10643	0	0	0	0	0	349	152	152	1995	19924	193	193	0	0	232	232
98	102	1371	2369	6876	23377	0	0	954	4904	14	162	0	1016	1720	31947	148	249	4	8	32	335
99	28	2141	2141	5113	29543	0	3986	0	7909	37	515	0	219	0	26366	0	0	9	17	109	645
100	37	2670	2670	1800	5238	0	5380	0	331	0	267	0	0	0	5607	74	74	0	0	123	123
101	390	3966	5181	7204	14114	1899	1899	788	3540	0	43	0	67	17397	24964	523	598	8	10	20	71
102	31	6968	6968	0	12548	0	0	0	2167	0	0	8	262	0	47196	14	79	0	3	63	154
103	851	2223	8254	2439	59142	0	1668	702	17860	0	79	16	105	4444	33864	185	645	2	12	0	32
104	249	2701	9329	1176	73787	0	364	1422	24734	0	112	359	4636	2654	62581	311	1140	2	68	2	3037
105	5009	5460	7688	11643	32764	7592	7844	1657	8824	0	29	0	27	38169	50230	885	1077	13	17	18	33
106	27	22247	22247	0	8421	0	49820	0	0	0	691	0	161	0	28017	0	0	0	0	87	87
107	88	7445	7445	2951	20421	0	15147	453	6782	4	120	0	553	5611	24905	342	412	5	19	45	603
108	256	3963	4237	9491	18198	5360	6469	2394	4758	0	39	0	75	8344	12238	131	146	6	8	13	42
109	457	2860	2874	11054	18555	7861	9176	1778	3692	0	43	3762	3789	37105	38672	516	516	12	12	84	84
110	17	1400	8424	0	23605	0	0	0	12774	68	258	0	1594	0	60686	33	443	13	13	258	830
111	184	1360	3054	3009	64837	842	842	1741	14892	0	150	892	953	6204	35622	180	332	4	13	13	28
112	36	560	5453	0	9793	0	35152	0	9909	0	392	0	760	0	17400	6	341	5	15	79	754
113	41	6784	6784	0	5893	0	38511	1735	2762	132	346	1312	1920	16340	16340	300	300	3	3	285	285

114	138	1564	1564	4966	14214	2273	5468	1555	3704	15	84	5	149	2266	4715	120	120	6	8	10	10
115	413	1662	1662	6634	18616	5925	6481	1150	3526	0	25	12	34	10670	14956	229	229	10	11	29	36
116	464	1698	2449	8329	25184	6670	8686	1406	5151	0	43	3	65	11344	17449	245	289	11	13	18	33
117	82	2158	7344	0	15191	0	47	0	8071	0	141	0	294	47	23663	4	327	6	15	45	286
118	749	810	1338	0	5403	0	732	0	1228	0	11	0	32	0	3367	8	31	60	62	0	13
119	279	1178	1178	1091	4779	0	0	229	626	0	4	926	934	21435	30579	992	1129	7	9	30	41
120	68	706	706	0	7788	0	0	0	0	0	0	0	145	0	4943	52	121	7	10	27	27
121	35	1070	1070	0	10971	0	10286	0	0	0	1273	0	0	1364	1490	640	1010	8	9	0	0
122	28	2097	2097	0	13801	0	0	0	0	0	0	0	0	321	7554	176	176	4	4	45	45
123	801	886	1186	1803	6007	0	331	0	1195	0	6	74	103	4334	9541	110	145	2	4	12	30
124	193	2335	3263	3153	8156	7091	7407	2036	4092	0	0	6	91	5601	11957	245	330	6	10	39	97
125	40	74	4992	0	3280	0	0	0	3844	0	1605	7	524	0	33603	63	338	2	6	115	335
126	2977	1070	1309	6645	10966	740	740	0	903	0	19	0	29	10334	12497	95	103	4	5	0	6
127	175	1347	1347	1298	39073	0	0	210	8150	0	300	0	0	0	5015	64	64	3	8	0	0
128	106	3117	8485	1617	6398	0	0	0	3878	0	475	0	945	560	24510	60	395	0	7	18	517
129	876	3045	3992	6741	19388	2927	2927	1759	5727	0	88	386	466	12456	17753	110	124	9	11	30	41
130	164	2600	2600	3565	12394	0	0	0	672	0	186	37	37	3030	3030	53	53	1	2	5	5
131	515	2547	4522	2732	34117	1848	1848	1905	11642	0	163	0	143	535	20106	25	48	2	9	0	27
132	69	4036	4036	331	2513	0	6570	0	0	0	276	630	680	5251	48478	151	193	0	4	33	33
133	118	2006	2006	3503	11220	0	4202	0	1067	0	228	0	53	2657	4873	57	57	4	6	104	104
134	157	9563	9563	4209	14797	0	4103	1215	3026	0	223	0	603	4768	6629	172	203	5	19	22	218
135	1334	2200	5185	944	42257	0	0	501	13529	0	42	0	0	789	25162	147	505	1	7	0	0
136	11873	63295	72331	3137	6059	970	970	0	1133	0	4	0	7	10935	12702	251	279	4	5	0	1
137	86	16049	17420	42	4453	0	0	0	2153	0	138	0	152	130	17123	73	138	0	1	26	76
138	76	80176	80176	3972	7938	0	0	5438	5438	0	377	0	457	0	0	1959	1959	0	7	13	13
139	633	3373	3895	1668	12280	0	0	0	3304	0	49	3445	3445	32843	37169	453	472	8	11	64	64
140	276	10145	10145	1998	5256	0	0	0	160	0	14	0	0	3923	3923	72	72	2	4	243	243
141	64	3404	3404	5073	29657	0	8836	0	2690	0	111	0	863	0	2884	99	99	2	7	0	0
142	53	2498	9846	5776	15751	0	0	0	6604	0	6	0	463	5044	35104	140	661	7	60	0	200

143	43	2412	6487	1516	5827	0	0	0	4183	0	27	0	625	1700	34935	117	397	1	4	0	107
144	1618	1817	1817	13403	20041	13479	13479	2465	3569	0	6	0	0	15608	16366	276	276	9	9	0	0
145	89	1116	1116	5277	32750	294	294	0	2240	0	76	0	18	0	31691	70	70	2	6	219	219
146	208	1820	1820	7258	44510	6502	6502	1296	6906	0	49	0	38	9007	17659	210	210	4	8	44	44
147	43	1062	1062	1770	14756	0	7319	0	2959	0	13	1800	1944	24782	33755	421	421	3	19	24	24
148	58	2143	2281	585	6212	12371	25052	0	1685	0	0	0	303	5124	24964	158	228	2	5	48	141
149	1305	2265	4269	4069	33578	6027	6027	2875	13454	0	24	226	226	13896	30760	554	823	4	8	0	0
150	1893	2033	3747	5747	37761	5286	5286	2272	12714	0	35	20	20	10470	20496	191	240	3	7	0	0
151	41	66	8161	0	4678	0	48545	0	9897	0	465	0	473	185	28335	71	434	0	3	0	355
152	69	446	446	1472	36627	0	6196	0	10666	0	149	0	142	514	7425	3	3	0	3	0	0
153	55	0	0	1780	37014	0	0	0	8639	0	164	0	312	0	104462	0	108	1	8	4	65
154	107	1152	1152	373	37930	0	14863	0	0	0	305	0	0	0	0	73	73	0	0	40	40
155	4052	3073	4238	9555	20014	3490	3790	762	4570	0	13	0	16	22998	27919	246	295	8	9	0	3
156	145	4632	6627	0	8154	0	1208	0	660	0	19	0	0	0	4708	52	140	0	0	13	13
157	96	9927	21355	0	2837	0	80	0	5545	0	54	0	373	0	23650	125	718	0	0	9	198
158	201	11474	19841	0	1962	0	2222	0	4350	0	37	0	217	1287	17887	250	661	0	0	32	224
159	969	13020	18968	37	2245	0	1052	0	937	0	0	0	20	1871	5825	352	552	0	0	0	6
160	159	18289	28172	0	5064	0	2972	308	4228	0	17	1298	1351	9522	24985	238	910	1	1	36	49
161	105	3443	9125	577	14028	0	0	1174	6791	0	22	0	49	0	20024	24	293	2	2	0	208
162	1561	1170	1863	3456	6586	5307	5588	1737	2971	0	3	0	26	3529	5805	82	116	6	7	0	9
163	79	3475	15144	0	4889	0	0	1658	7614	0	83	0	151	0	27787	18	589	0	0	15	40
164	5068	715	1063	507	2125	0	134	0	488	0	2	0	11	1460	2492	141	174	1	1	0	1
165	77	3931	12894	2	5292	0	3189	4259	9375	16	16	0	1720	1357	31728	87	599	1	1	0	558
166	41	11663	13889	0	8437	0	0	3170	3170	108	238	0	4127	0	31147	0	92	0	0	0	0

Table 4b. Mass concentrations of potentially bioaccessible (SLF) and total acid-soluble (total) Cu, Zn, As, Se, Sr, Ag, Cd, Sn, Ba, Pb, U estimated for samples collected in Mines 10-24. (Sample numbers here are the same as the paired samples used for SEM-EDX analysis, and thus can be cross-referenced with the sample numbers shown in Tables 1-3.) Elemental concentrations are reported on dry mass basis (i.e., µg per g of respirable dust). Sample mass values indicate dust mass recovered from the sample filter. Mass concentrations below the method reporting level (MRL) are shown in red.

Sample No	Mass (µg)	Mass concentration (µg/g)																					
		Cu		Zn		As		Se		Sr		Ag		Cd		Sn		Ba		Pb		U	
		SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total	SLF	total
91	163	0	19	127	2841	56	720	0	0	76	171	0	1	0	0	1	220	615	2246	1	22	0	2.3
92	395	0	0	7	264	21	29	0	0	85	90	0	0	0	0	0	0	621	806	10	10	0	0.4
93	202	0	0	123	5524	0	0	0	0	89	97	0	0	1	2	7	24	177	796	10	11	0	1.3
94	69	0	0	101	3051	43	43	0	0	83	83	0	2	1	1	0	83	1073	1788	0	1	0	0.1
95	673	0	9	0	383	6	93	0	0	50	70	0	1	0	0	0	15	612	837	14	19	0	0.7
96	248	216	216	219	219	8	108	0	0	68	76	0	0	0	0	0	0	604	665	21	21	0	0.9
97	28	110	146	902	902	20	1114	0	0	80	80	0	2	0	0	0	66	3736	3736	39	39	0	2.1
98	102	0	2	997	1797	17	954	0	0	87	130	0	2	0	0	0	0	1294	1592	13	13	0	1
99	28	0	0	0	1786	52	1069	0	36	0	205	0	9	0	1	0	0	0	5089	0	6	0	1.6
100	37	0	832	36	1607	13	13	0	20	93	93	0	1	2	2	64	64	3231	3754	8	8	0	0
101	390	0	72	43	83	0	388	0	0	68	100	0	0	0	0	3	3	613	686	8	12	0	0.4
102	31	0	0	1325	7487	0	2120	0	0	42	123	0	10	0	1	0	0	500	1779	0	0	0	0.6
103	851	0	23	219	358	0	175	0	3	61	143	0	1	0	0	0	0	471	869	0	12	0	1.9
104	249	0	109	0	232	1	55	0	0	63	147	0	2	0	1	0	0	508	1007	0	17	0	2
105	5009	0	7	0	73	0	35	0	0	69	95	0	1	0	0	0	0	569	734	11	16	0	0.7
106	27	0	0	0	0	7	7	0	38	30	30	0	18	0	1	0	0	0	2471	0	0	0	1.6
107	88	0	0	441	625	6	193	0	13	99	122	0	2	0	0	0	0	1365	1917	0	0	0	0.6
108	256	0	0	0	439	0	221	0	0	70	84	0	0	0	0	0	0	543	658	10	10	0	0.4
109	457	80	80	0	131	0	27	0	2	53	59	0	0	0	0	0	0	554	617	12	12	0	0.2
110	17	0	23	945	945	118	939	0	0	218	462	0	5	9	9	127	147	4594	4967	0	0	0	3.3
111	184	0	0	0	231	6	6	0	0	74	110	0	1	0	1	4	4	540	777	0	13	0	1.7
112	36	0	49	10156	10459	33	3037	0	41	54	251	0	3	0	1	0	115	496	1336	0	0	0	0.3

113	41	456	456	182	550	84	84	0	42	164	164	0	4	0	0	0	0	1133	2312	0	44	0	0
114	138	0	0	16	306	10	143	0	13	105	130	0	2	0	0	0	0	1123	1222	2	2	0	0
115	413	53	207	0	187	2	73	0	1	84	134	0	1	0	0	0	0	1014	1052	11	21	0	0.4
116	464	0	0	0	132	1	1	0	1	74	166	0	1	0	0	0	7	884	982	10	14	0	0.9
117	82	677	827	0	1322	9	1972	0	14	77	334	0	1	0	1	0	0	0	831	0	33	0	2
118	749	0	0	0	16	0	145	0	2	170	224	0	2	0	0	0	0	445	474	0	10	0	0.4
119	279	13	13	1	28	6	156	0	0	55	56	0	1	0	0	0	51	569	586	68	84	0	0.1
120	68	0	0	0	0	0	0	0	0	38	38	0	2	2	2	35	35	122	574	0	25	0	0
121	35	0	0	908	10524	0	0	0	0	180	180	2	2	10	10	17	17	415	2993	43	43	1	0.9
122	28	0	0	0	4291	39	605	0	0	144	144	0	2	4	4	28	28	4102	4102	0	0	0	0
123	801	0	9	0	129	0	78	0	1	98	106	0	0	0	0	0	19	329	364	1	5	0	0.5
124	193	0	0	31	320	0	517	0	5	170	228	0	0	0	0	0	13	379	493	4	4	0	0.5
125	40	0	0	471	9785	0	1265	0	0	142	180	8	9	8	8	67	229	264	2701	13	13	11	11.7
126	2977	0	0	102	326	0	0	0	0	84	87	1	1	0	0	6	11	134	168	7	7	2	2.3
127	175	0	0	135	1152	0	0	0	0	96	96	0	2	2	2	4	67	164	651	0	0	1	3.2
128	106	1254	1481	1250	5447	0	1217	0	0	123	191	2	2	2	2	15	112	142	1110	35	44	1	2.5
129	876	0	0	110	252	0	11	0	0	94	113	0	1	3	4	1	1	251	386	20	24	13	17.8
130	164	0	0	228	1156	0	0	0	0	143	143	0	0	1	1	4	4	168	581	9	9	5	9
131	515	0	0	0	575	0	0	0	0	77	113	0	2	0	3	0	0	166	446	1	18	2	12.7
132	69	0	0	259	2400	0	0	0	0	280	280	2	2	3	3	9	9	253	988	22	22	1	1.6
133	118	0	0	156	1223	0	0	0	0	123	123	0	0	2	2	2	2	202	551	12	12	7	9.7
134	157	0	0	74	714	0	0	0	0	201	201	1	1	2	2	3	12	242	531	10	10	9	13
135	1334	0	0	0	83	0	0	0	0	48	94	0	1	0	0	0	2	133	679	0	21	0	3.1
136	11873	0	0	25	39	0	0	0	0	60	68	1	1	0	0	5	5	399	445	8	10	2	2
137	86	0	0	152	2455	0	420	0	0	145	155	3	4	2	4	19	19	233	1029	2	2	3	3.3
138	76	0	0	129	3532	0	0	0	0	195	195	2	2	2	2	17	17	725	1949	6	6	2	2.4
139	633	71	71	144	373	0	0	0	0	88	112	0	2	1	1	6	6	143	298	20	22	1	1.5
140	276	27	116	68	581	0	0	0	0	114	114	1	1	1	1	4	4	276	467	8	8	1	1
141	64	0	0	180	2284	0	0	0	0	116	116	1	1	2	2	5	36	359	1193	0	0	1	1.5

142	53	0	0	395	4764	0	1536	0	0	163	247	4	4	4	8	7	67	343	1366	27	27	1	2.2
143	43	0	0	626	4929	0	915	0	0	119	158	0	0	4	4	3	3	282	1570	3	3	1	1.6
144	1618	0	0	29	157	0	0	0	0	106	108	1	1	1	1	0	2	273	354	17	17	1	1.1
145	89	1803	1803	880	3487	0	0	0	0	100	100	0	0	1	1	7	63	199	1307	6	21	0	0.9
146	208	0	0	47	1238	0	0	0	0	106	123	1	2	1	1	18	56	494	1824	9	9	3	4.1
147	43	0	0	145	4211	0	0	0	24	110	110	2	2	5	5	32	32	255	1903	0	0	5	5.4
148	58	2208	2646	563	4773	0	0	0	25	101	101	4	4	2	2	23	64	260	2189	7	7	2	2.6
149	1305	0	0	5	118	0	0	0	0	32	62	0	0	0	0	3	19	165	510	2	8	1	2.3
150	1893	0	0	0	37	0	0	0	0	40	76	0	0	0	0	1	9	203	646	5	16	1	3.5
151	41	0	0	184	11668	0	2152	0	119	83	164	6	6	2	2	8	441	231	1828	3	3	1	2.4
152	69	0	0	0	3196	0	0	0	17	2	2	0	1	1	2	2	68	0	1652	0	0	0	1.6
153	55	2262	2262	0	6706	0	0	0	0	0	23	1	1	0	6	2	2	0	1884	0	215	0	1.7
154	107	0	0	0	0	89	89	0	0	71	71	2	2	0	0	0	0	135	135	0	0	0	0
155	4052	0	9	0	0	23	78	0	0	21	36	0	0	0	0	0	0	160	220	9	9	0	0.6
156	145	0	3057	0	1412	32	316	0	0	113	154	0	0	0	1	0	74	120	120	0	93	0	0.6
157	96	0	1942	0	0	27	2309	0	0	39	197	0	0	0	1	0	22	0	0	0	0	0	1.9
158	201	1296	2614	0	518	14	1715	0	0	66	191	0	1	0	1	0	0	79	79	12	42	0	1.1
159	969	0	31	0	118	2	272	0	0	76	106	3	3	0	0	21	21	57	57	1	1	2	2.7
160	159	29	304	0	0	7	1217	0	0	38	141	1	1	0	1	4	4	105	109	0	0	1	2.7
161	105	0	1175	0	0	5	876	0	0	162	335	0	0	0	1	0	0	307	307	0	0	0	1.8
162	1561	0	49	0	0	4	140	0	0	63	85	0	0	0	0	0	0	156	163	0	0	0	0.3
163	79	0	459	0	0	5	2731	0	0	169	399	0	0	1	1	0	0	221	221	0	0	0	2
164	5068	0	6	0	0	3	54	0	0	200	430	0	0	0	0	0	0	42	54	0	0	0	0.2
165	77	0	330	0	0	23	2140	0	0	326	542	0	0	1	1	0	0	237	237	0	0	0	1.6
166	41	0	673	0	0	24	63	0	0	418	540	0	0	0	2	0	0	225	225	0	0	0	1.2

3. Experimental Design, Materials, and Methods

3.1. Sample Collection and Preparation

A total of 171 sets of respirable dust samples were collected in 25 underground coal mines across five distinct regions of the United States: northern Appalachia (region "A", which includes US Mine Safety and Health Administration [MSHA] districts 2 and 3; mines 5, 6, 16-18), mid-central Appalachia (B, MSHA district 4; mines 1-4, 15), south-central Appalachia (C, MSHA districts 5, 7 and 12; mines 7-14, 21, 22, 25), mid-western basin (D, MSHA district 8; mines 19 and 20), and western basin (E, MSHA district 9; mines 23 and 24). Each set represents a unique sampling event (i.e., specific sampling location in a specific mine). Sample collection was targeted in five standard locations per mine: intake airway (I), just outby of the primary production area (including the headgate of a longwall section) or along the mantrip track; feeder (F), near the feeder breaker or along the main conveyor belt; production (P), just downwind of an active continuous miner or near the midface of a longwall section; roof bolter (B), just downwind of an active bolter; and return airway (R), just outby of the primary production area (including the tailgate of a longwall section). In some mines, one or more of the targeted locations could not be sampled; in some cases, multiple sampling events were conducted in given location (e.g., on two separate shifts).

A detailed description of the mines and sampling protocol is reported in the literature [1]. Briefly, each sample set consists of multiple samples collected simultaneously in the same location. Each sample was collected using a personal air pump (Escort ELF model; Zefon International, Ocala, FL) with a 10-mm nylon Dorr-Oliver cyclone (Zefon International, Ocala, FL), which produces a d_{50} cut size of about 3.5 µm at the sampling flow rate of 2 L/min. A rigid frame was used to mount all samplers used to collect a given set, such that the inlet of all cyclones was positioned within about 15 cm of each other and oriented in the same direction. The samples analyzed for this report were collected directly onto 37-mm polycarbonate filters (PC, track-etched with nominal 0.4-µm pore size) over a continuous 2-4 hr period.

For analysis of particle size and mineralogy, a circular subsection (8-9mm diameter) was cut from one PC filter sample from each set, mounted on an aluminum stub, and sputter-coated with Au/Pd. Another PC filter from each set was used for the metals and trace elements analysis.

3.2. SEM-EDX Analysis to Determine Particle Size and Mineralogy Distributions

The SEM-EDX analysis was conducted in two phases, submicron and supramicron, each having a dedicated computer-controlled routine. Both routines used the same instrumentation and software, a FEI Quanta 600 FEG environmental SEM (FEI, Hillsboro, OR) equipped with a Bruker Quantax 400 EDX spectroscope (operated in backscatter mode) and Esprit software (Version 1.9; Bruker, Ewing, NJ). The analysis on each sample (i.e., the filter subsection mentioned above) proceeded as follows:

- Initially, the SEM stage was moved to the center of the sample stub, which was designated as "Frame 0" (Figure 1). Analysis began at Frame 0 and then proceeded through subsequent frames (i.e., up to Frame 79 for submicron analysis and up to Frame 39 for supramicron analysis). The frame positioning was pre-set to avoid user bias and ensure that data was collected across multiple areas of the sample; all frames were positioned within a 7-mm diameter circle allow tolerance in the case that the sample was not perfectly mounted in the center of the stub. For submicron analysis, interior frames were spaced about 0.5 mm apart and the spacing was increased moving toward the sample edge. The magnification was set to 10,000× such that each frame was approximately 140.25 µm^2. For supramicron analysis, frames were spaced about 1 mm apart and magnification was set to 1,000× such that each frame was approximately 14025 µm^2. Table 5 summarizes the other key parameters for each routine.

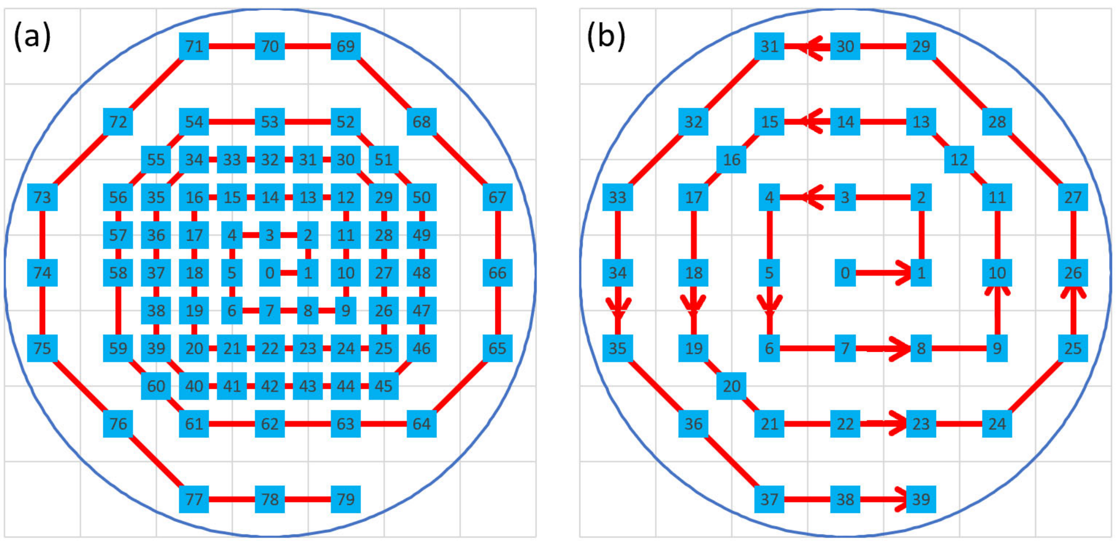


Figure 1 Frame positions for computer-controlled SEM (a)submicron and (b)supramicron particle analysis routines. Blue circles have a 7-mm diameter. The span between adjacent frames is generally 0.5 mm for submicron and 1.0 mm for supramicron particle analysis routines.

- In each frame, 30 or 50 particles were analyzed for submicron and supramicron routines, respectively. The aim was to analyze approximately 300 submicron particles and approximately 500 supramicron particles per sample; therefore, at least 10 frames were analyzed in both routines.

Table 5. Description of sub- and supramicron particle analysis routines using SEM-EDX analysis routines (reproduced from [1]).

Parameters	Submicron	Supramicron
Magnification	10,000×	1,000×
Voltage (kV)	10	15
Spot size	5.5	
Working distance (mm)	12.5	
Brightness	92.5%	
Contrast	60-70%	
Recorded elements for normalized atomic % value	C, O, Al, Si, Mg, Ca, Ti, Fe	
Particle size based on longest dimension (nm)	100-1,000	1,000-10,000
Frame area (μm^2)	140.25	14,025
Minimum number of frames	10	
Maximum number of frames	80	40
Maximum number of particles per frame	30	50
Total number of particles aimed	300	500

- For each particle, its length, width, projected area, particle location, and the elemental spectra were recorded. The dimensions were used to determine projected area diameter.
- Normalized atomic percentages of eight elements (per [3]) (i.e., C, O, Al, Si, Mg, Ca, Ti, Fe) were used to classify particles into in nine pre-defined classes (per [1]): carbonaceous (C); mixed carbonaceous (MC); aluminosilicates, kaolinite-like (ASK) or other (ASO); other silicates (SLO); silica (S); heavy minerals (M), which mostly included metal sulfides or oxides; carbonates (CB), or other (O). These are referred to as "mineralogy" classes herein, though it is acknowledged mineralogy is inferred from the elemental spectra. The classification criteria are listed in Table 6 and verified with high-purity reference materials (Table 7) for both submicron and supramicron particles [1]. It should be noted that submicron particles in the C class may include both carbonaceous dust (i.e., coal and organic matter) and diesel particulates, which can sometimes be identified based on their characteristic morphology [1,2].

Table 6. Mineralogy classification criteria (reproduced from [1]).

Class	Routine	Normalized Atomic%							
		C	O	Al	Si	Ca	Mg	Ti	Fe
C	Sub	≥75	<29	≤0.30	≤0.30	≤0.41	≤0.50	≤0.50	≤0.50
	Supra							≤0.06	≤0.15
MC	Sub			<0.44	<0.44	≤1.00	≤0.50	≤1.00	≤1.00
	Supra			<0.35	<0.35	≤0.50	≤0.50	≤0.60	≤0.60
ASK[1]	Sub			≥0.44, (≥37)	≥0.44, (≥42)	(<16)	(<4)	(<8)	(<10)
	Supra			≥0.35, (≥39)	≥0.35, (≥32)	(<8)	(<15)	(<13)	(<13)
ASO[1]	Sub			≥0.44, (<37)	≥0.44, (<42)	(≥16)	(≥4)	(≥8)	(≥10)
	Supra			≥0.35, (<39)	≥0.35, (<32)	(≥8)	(≥15)	(≥13)	(≥13)
SLO[2]	Sub				≥0.50				
	Supra				≥0.33				
S[3]	Sub				≥0.50				
	Supra				≥0.33				
M	Sub			>1.00				>1.00	>1.00
	Supra								
CB	Sub	<88	>9			>1.00	>0.50		
	Supra					>0.50			

[1]To differentiate ASK from ASO, additional limits for Al, Si, Mg, Ca, Ti and Fe are shown in parenthesis (normalized to exclude C and O)
[2]Additional limits for SLO: Si/(Al+Si+Mg+Ca+Ti+Fe) < 0.5
[3]Additional limits for S: Al/Si < 1/3 and Si/(Al+Si+Mg+Ca+Ti+Fe) ≥ 0.5

Table 7. Classification results on submicron and supramicron particles in respirable dust samples generated in the laboratory using high-purity reference materials (reproduced from [1]). Results are shown for both submicron and supramicron SEM analysis routines. Dominant mineralogy class is shaded grey.

Reference Material	SEM Routine	Mineralogy Distribution (%)								
		C	MC	ASK	ASO	SLO	S	M	CB	O
Coal	Sub	91	6	3	0	0	0	0	0	0
	Supra	99	1	0	0	0	0	0	0	0
Kaolinite	Sub	9	3	85	3	0	0	0	0	0
	Supra	0	0	98	2	0	0	0	0	0
Quartz	Sub	1	1	3	3	2	91	0	0	0
	Supra	1	1	1	1	0	94	0	0	0
Calcite	Sub	1	1	0	0	0	1	0	97	0
	Supra	1	0	0	0	0	0	0	99	0
Rock Dust	Sub	1	1	0	17	0	0	0	78	3
	Supra	6	3	0	1	0	0	0	88	1

- Following completion of the sub- and supramicron particle analysis, the resulting datasets were merged to allow description of particle size and mineralogy distributions across the entire analyzed range. This was done by normalizing both datasets on the basis of number of particles per analyzed filter area. Finally, the data were split into the size bins included in Tables 3a-i.

3.3. Metals and Trace Elements Analysis

Analysis of potentially bioaccessible and total acid-soluble metals and trace elements in the respirable dust samples was conducted using two sequential digestions followed by inductively-coupled mass spectroscopy on the digestates. The first digestion was in a simulated lung fluid (SLF) and the second was in a 4-acid solution. Following is a detailed description of the entire method used to prepare and analyze the samples.

For dust recovery and the digestion in SLF, the procedure described in [2] was followed. Briefly:

- One PC filter from each sample set was weighed to establish a pre-weight prior to dust recovery.
- Each filter was placed into a glass digestion tube and rinsed with 18 MΩ water. Enough water was added to fully submerse the filter (about 5 mL). The tubes were capped and placed in an ultrasonic bath for 1 hr (@ constant 37 °C), then centrifuged for 10 min (@ 3000 rpm) to settle the liberated dust. Tubes were uncapped and water was evaporated in a clean oven (@ 120 °C).
- Dry filters were carefully removed and re-weighed to determine recovered dust mass by difference with the pre-weight.
- SLF solution was prepared per [4] using the reagents shown in Table 8. SLF was prepared using 18 MΩ water as the base. It was stored at 3 °C and used within 1 week of preparation.

Table 8. Simulated lung fluid (SLF) recipe and order of chemical addition (reproduced from [2]).

Addition	Chemical	Formula	Concentration/L
1	Ammonium chloride	NH_4Cl	535 mg
2	Sodium chloride	$NaCl$	6780 mg
3	Sodium bicarbonate	$NaHCO_3$	1770 mg
4	Sodium carbonate	Na_2CO_3	630 mg
5	Sodium dihydrogen phosphate monohydrate	$NaH_2PO_4 \cdot H_2O$	166 mg
6	Sodium citrate dihydrate	Na_3-citrate$\cdot 2H_2O$	59 mg
7	Glycine	$C_2H_5NO_2$	450 mg
8	Sulfuric acid	H_2SO_4	51 mg (27.7 µL)
9	Calcium chloride dihydrate	$CaCl_2 \cdot 2H_2O$	29 mg

- The SLF solution was added to each digestion tube containing only the recovered dry dust, as well as tubes prepared as blank (clean PC filter) and matrix (SLF only) samples. The SLF solution volume was determined using a 1/10,000 ratio between the dust mass (g) and SLF volume (mL) ratio. This solid/liquid ratio was chosen per [5]. For samples with recovered dust weight below 200 µg, a standard SLF volume of 2 mL was used.

- The tubes were capped and placed in the ultrasonic bath for 24 hours (@ constant 37 °C), and then centrifuged for 10 min (@ 3000 rpm).
- A 5 mL aliquot of the SLF digestate in each tube was taken by syringe using a PTFE filter (0.1 µm pore size) to trap any suspended dust particles. The filtered digestate was then added to new ICP tube and diluted with 18 MΩ water to a volume of 9.8 mL. Finally, 0.2 mL HNO_3 (trace metal grade) was added to each ICP sample to achieve 2% acid by volume (and a total sample volume of 10 mL).

The second digestion followed a procedure modified from [6] to digest the remaining dust from each sample (i.e., that not digested by the SLF) using four concentrated acids (i.e., HNO_3, $HClO_4$, HF and HCl):

- The 4-acid digestion procedure began with pre-digestion step. The PTFE filter used to trap dust from each SLF sample was placed into a 50-mL Teflon vessel. The filter and vessel walls were then rinsed by pipetting a minimal volume of 18 MΩ water (i.e., 1-2 mL). Then, 750 µL of concentrated HCl (trace metal grade) was added to each vessel. The vessels were capped and placed into a hot block with individual wells, and left under a fume hood overnight at 60°C. Vessels were allowed to cool to room temperature and then transferred to an $HClO_4$-rated fume hood for the rest of acid digestion procedure.
- The next three acids (all trace metal grade) were added to each vessel in this order: 500 µL of HNO_3, 250 µL of $HClO_4$, and 500 µL of HF. Contents were swirled gently and then the vessels were placed into the hot block at room temperature. They were heated (uncapped) to 110°C and left at temperature until incipient dryness.
- After the vessels cooled, another 250 µL of the $HClO_4$ was added to each. The vessel walls were then rinsed by pipetting a minimal volume of 18 MΩ water (i.e., 1-2mL and gently swirled before being placed back into the hot block. The vessels were then heated (uncapped) to 150°C and left at temperature until incipient dryness.
- After the vessels cooled, another 250 µL of the HNO_3 was added to each. They were left uncapped for 5 mins, and then swirled to mix. Then 6 mL of 18 MΩ water was added to each vessel, and it was swirled again. Next, 25µL of H_2O_2 (30%) was added and the reaction was allowed to subside before the vessels were capped tightly and shaken. They were placed in an oven for 1 hr (@ 100°C).
- Finally, the total volume of digestate in each vessel was transferred to a new ICP tube and acidified to 2% HNO_3 (by volume).

To analyze the SLF and 4-acid digestates, a Thermo Electron iCAP-RQ ICP-MS instrument was used (Thermo Fisher Scientific, Waltham, MA):

- For each batch of ICP samples, three blank PC filters and blank solutions were also prepared using the SLF and 4-acid digestion procedures described above. Their results were used for blank and matrix corrections, respectively.
- The ICP results (µg/L in the digestate solutions) were corrected and then transformed into dry dust concentrations (µg/g) using the dust mass recovered from each filter sample. The concentration determined from the SLF digestate is regarded as potentially bioaccessible; and the sum of the concentration from the SLF and 4-acid digestates is regarded as total acid-soluble concentration. It is noted that, due to relatively low sample masses for the current dataset, results in Tables 4a-b should be regarded as estimated concentrations.
- Based on data from researchers at the U.S. Geological Survey (USGS), the 4-acid digestion procedure followed here (i.e., with open vessel during HF digestion) is predicted to result in Si loss of about 90% (H. Lowers and Z. Arslan, personal communication, September 16, 2021). To enable general comparisons between Si and other elements (i.e., in Tables 4a-b), this factor was used to further correct the Si data herein. However, the reported Si concentrations should accordingly be viewed with some caution.
- The elements that were measured by ICP-MS and reported here are listed in Table 9 with their respective method reporting level (MRL). MRLs are based on the calibration curve for each element, which is generated using a series of standard solutions; the limits of detection are generally about one order of magnitude lower. (Note that other elements, including Ca, Na, P, Ti, S and Cl, can be measured by ICP-MS, but were not included in the analysis presented here due to significant interferences from the digestion solutions.) In addition to ICP-MS calibration prior to sample analysis, check standards and blank samples were run between every set of 10 samples analyzed to ensure that there was no significant instrument drift or carryover contamination between samples.

Table 9. MRLs for elements included in ICP-MS analysis.

MRL (µg/L)	Element	MRL (µg/L)	Element
0.1	V, Cr, Mn, Co, Ni, As, Ag, Cd, Sn, Ba, Pb, U	5	Mg, Fe
0.5	Sr	10	Al, K
1	Cu, Zn, Se	500	Si

4. References

[1] E. Sarver, C. Keles, S. Ghaychi Afrouz, Particle size and mineralogy distributions in respirable dust samples from 25 US underground coal mines, Int. J. Coal Geol. 247 (2021) 103851. https://doi.org/10.1016/j.coal.2021.103851

[2] E. Sarver, C. Keles, M. Rezaee, Characteristics of respirable dust in eight Appalachian coal mines: A dataset including particle size and mineralogy distributions, and metal and trace element mass concentrations, Data in Brief 25 (2019) 104032. https://doi.org/10.1016/j.dib.2019.104032

[3] V. Johann-Essex, C. Keles, E. Sarver, A Computer-controlled SEM-EDX routine for characterizing respirable coal mine dust, Minerals 7(1) (2017), 15 pages. http://dx.doi.org/10.3390/min7010015

[4] S.A. Morman, V.H. Garrison, G.S. Plumlee, Trace metals in Saharan dust: the use of in vitro bioaccessibility extractions to assess potential health risks in a dustier world: Chapter 3, in: L.L. McConnell, J. Dachs, C.J. Hapeman (Eds.), Occurrence, Fate and Impact of Atmospheric Pollutants on Environmental and Human Health (Volume 1149), American Chemical Society, Washington, DC, 2013, pp. 41-58. https://doi.org/10.1021/bk-2013-1149.ch003

[5] C. Julien, P. Esperanza, M. Bruno, L.Y. Alleman, Development of an in vitro method to estimate lung bioaccessibility of metals from atmospheric particles. J. Environ. Monit. 13 (2011) 621–630. http://dx.doi.org/10.1039/c0em00439a

[6] P.H. Briggs (2002) The determination of forty elements in geological and botanical samples by inductively coupled plasma-atomic emission spectrometry, chap. G of Taggart, J.E., Jr., ed., Analytical methods for chemical analysis of geologic and other materials, U.S. Geological Survey: U.S. Geological Survey Open-File Report 02-223, 18 p. [http://pubs.usgs.gov/of/2002/ofr-02-0223/G01fortyelementICP-AESsolid_M.pdf]