# INVESTIGATIONS INTO THE SPECIFIC FIBROGENICITY OF MINE DUSTS IN HARDCOAL MINES OF COUNTRIES IN THE EUROPEAN COMMUNITY

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Results of investigations in the field of pneumoconiosis research in the last 30 years indicate that the mine dusts in the various deposits of European and American hardcoal mines have a different fibrogenicity.[1-4] Although epidemiological research into coal workers' pneumoconiosis in British coal mines (Figure 1) has revealed a clear relationship between the occurrence of radiological lung changes (ILO category 2 and higher) and the mass concentration of respirable dust in one and the same colliery, a significant difference is to be observed in the frequency of the occurrence of pneumoconiosis between collieries where either high rank coal (colliery T) or low rank coal (colliery Q) is mined with comparable dust exposure. In colliery T with high rank coal, the response is several times greater than that in colliery Q with low rank coal. At the same time, the curve of the occurrence of pneumoconiosis as a function of the dust concentration is far steeper for colliery T than for colliery Q. Similar relationships have been observed in Germany, France and the USA.

Even today, these differences cannot be explained by reference to the dust composition, even with the quartz contents determined by X-ray diffraction or infrared spectrography. As can be seen from the following table (Figure 2) of results from British investigations,[3] the frequency of the occurrence of pneumoconiosis (ILO category greater than 2/1) in the high rank coal seams (colliery W) is roughly 10 times higher than in the low rank coal seams (colliery Q), although the quartz content of the dust from colliery W is only one quarter of the value for the dust from colliery Q. The mineral content (ash content) of the dust from colliery W is also correspondingly lower than that of the dust from colliery Q. Similar results were obtained in earlier investigations[5,6] and also from a more recent investigation[7] in Germany as can be seen in the following table (Figure 3). Here again, as with the results of the British investigations, a ratio of 1:10 is to be observed in the prevalence of radiological changes (ILO category 2/1 and higher)—with an exposure time of 22 to 30 years—as a function of the rank of the coal and, at the same time, a lower cumulative quartz dust dose with higher prevalence.

The occurrence of the pneumoconiosis does not therefore correlate generally with the quartz content of the dusts. A more obvious correlation appears to exist with the specific petrographic characteristics of the stratigraphic horizons, which have a modifying effect on the fibrogenicity potential of the mine dusts. The rank of the coal could therefore be taken as an initial indicator for this potential.

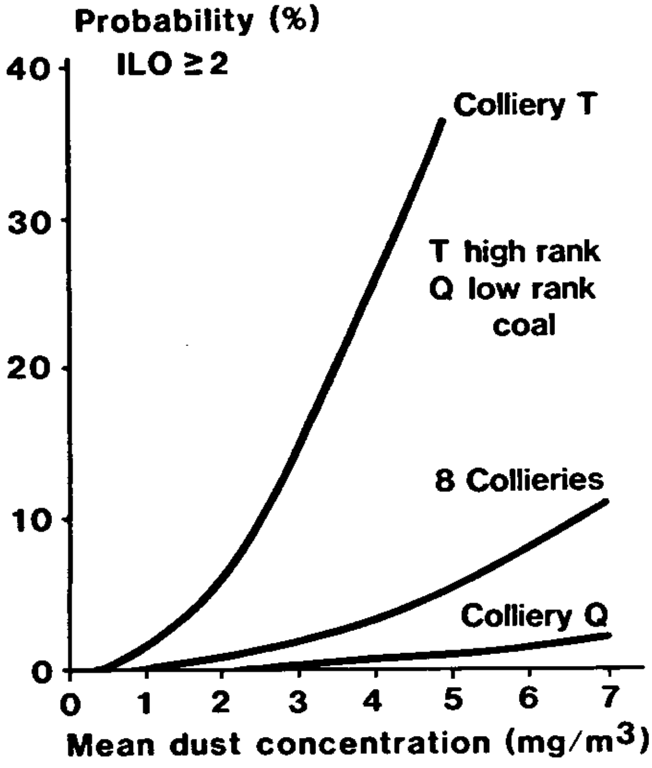


Figure 1. Estimated (percent) probabilities of category 2 or higher simple pneumoconioses, in relation to mean dust concentration.[3]

Cell experimental investigations (Figure 4) with mine dusts taken from various stratigraphic horizons of German hardcoal mines,[1,2] allow the following conclusions to be drawn, which also correspond to the results of the epidemiological investigations.

- The cytotoxicity (decrease in TTC reduction activity means an increase in cytotoxicity) of dust samples from one and the same stratigraphic horizon increases with increasing ash (mineral) and quartz content.

| Colliery | | Prevalence 2/1+ Observed/Predicted x 100 | Dust composition (%) | |
|---|---|---|---|---|
| | | | Quartz | Ash |
| Q (low rank) | | 18 | 5,4 | 70,6 |
| Y | | 67 | 3,6 | 32,7 |
| V | Seam 1 | 78 | 3,4 | 41,9 |
| | Seam 2 | 78 | 2,0 | 26,2 |
| W (high rank) | | 182 | 1,4 | 17,4 |

Figure 2. Prevalences of ILO categories 2/1+ in various British collieries with low resp. high rank coal and different composition of dust.[3]

| | Cohort size | Dust dosis (22/30 years) | | Prevalence | |
|---|---|---|---|---|---|
| | | c | $c_q$ | >1/1 ILO % | ≥2/1 ILO % |
| Group 1 (low rank seams) | 109 | 3700 | 3200 | 9,2 | 1,8 |
| Group 2 (high rank seams) | 109 | 5500 | 1800 | 31,2 | 18,8 |

Figure 3. Prevalences of ILO categories 1/1 and higher in German low rank coal resp. high rank coal seams in relation to mean total respirable dust doses c and respirable quartz dust doses $c_q$.[7]

- The same cytotoxicity of approx. 35% (65% TTC-RA) is observed for dusts from the later stratigraphic horizons with low rank coal (horst strata), however, only at roughly three times the ash (mineral) or quartz content of dusts from earlier horizons with high rank coal (lower Essen strata), as shown in Figure 5.

The petrographic characteristics of the stratigraphic horizons or deposits which could be responsible for the differences in specific cytotoxicity and fibrogenicity are:

- The quartzes from the various horizons have a different effect. Either the quartz from the earlier horizons with high rank coal has a far greater fibrogenicity, or the quartz from the later horizons with low rank coal has only a very low fibrogenicity. It was possible to show that structural impurities (Al ions in Si lattice positions among others) introduced into the lattice during the genesis of the quartzes or introduced subsequently into the surface by associated minerals led to a reduction in the cytotoxicity (Figure 6) and also, during *in vivo* experiments on animals, to a reduction in the fibrogenicity.[8,9] The less disturbed, i.e., the purer, the $SiO_4$ tetrahedra of a quartz are, the greater its specific fibrogenicity potential.
- X-ray diffractive, amorphous silica present in differing contents in mine dusts has an amplifying effect on the cytotoxicity[9] and fibrogenicity (Figure 7) compared with standard quartz DQ 12, as shown by the lymph node test in the animal experiment.[10]
- The other associated minerals occurring along with the quartzes in the mine dusts have an amplifying, i.e., additionally fibrogenous, effect in horizons with high rank coal (e.g. kaolinite) but an inhibiting effect in horizons with low rank coal.
- The individual particles of the respirable dust in the various horizons have either a differing heterogeneity (intergrowths) or are more homogeneous (with free active surfaces).

However, the relationships between the cytotoxicity of the dusts, their fibrogenicity and the results of the epidemiological investigations must be examined more closely.

On 1st October 1987, the first 2-year phase of a joint European research programme sponsored by the EEC was started to corroborate these non-contradictory and exemplary investigation results and to discover measurable properties of the mine dusts which allow the specific fibrogenicity potential to be definitively characterized. This programme is entitled "Characterization of Mine Dusts with Respect to their Specific Fibrogenicity." The research institutes involved in this programme are the Instituut voor Reddingswezen, Ergonomie en Arbeidshygiene (IREA), Hasselt/Belgium, the Centre d'Etudes et Recherches de Charbonnages de France (CERCHAR), Verneuil-en-Halatte/France, the Steinkohlenbergbauverein, Essen/W. Germany, the Silikose-Forschungsinstitut, Bochum/W. Germany and the Institute for Occupational Medicine, Edinburgh/UK.

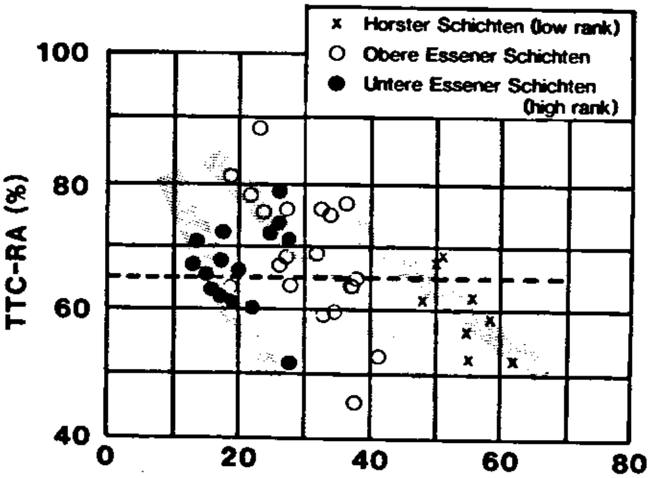


Figure 4. Cytotoxic effects (100-TTC-RA%) of German mine dusts from different stratographic horizons in relation to ash (mineral)-content.[2]

The sample material used in this programme is quartzes of various genesis and purity, mine dusts from earlier programmes (1960's and 1970's) and current mine dusts. The mine dust samples are or were taken using BAT-II samplers (CPM-3 samplers in France) from various strata over a number of weeks in such a way that at least 3 samples from each seam with different mineral (quartz) content can be examined in order to enable the trend of a dose/response relationship to be determined in a biological experiment. Dust samples from a total of 11 seams in the Ruhr coalfield and 4 seams in the Saar coalfield in Germany, 2 seams in Belgium, 3 seams in France and 4 seams in the UK were used.

The investigations incorporate:

- An extensive mineralogical, physical and chemical analysis, in particular of the individual particles, using electron microscopy (STEM) and laser-induced mass spectrometry (LAMMA),
- Luminescence measurements of their electron structure,
- The adsorption and desorption behaviour,
- Cytological, histological and biochemical *in vitro* reactions,

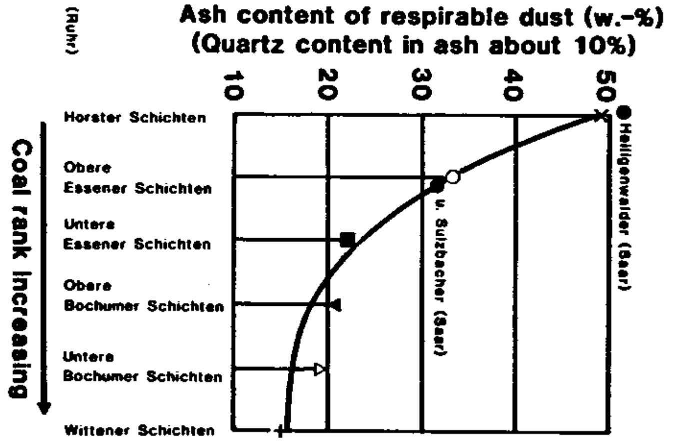


Figure 5. Ash (mineral)-content in German mine dusts from different stratographic horizons with same cytotoxic effect (35% depression of TTC-RA 120/Figure 4).[2]

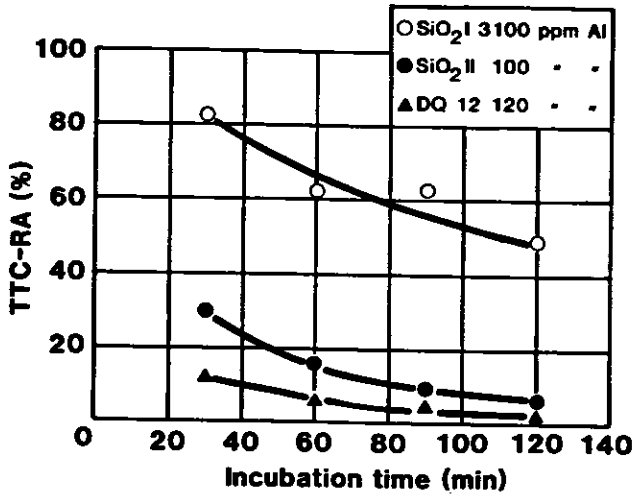
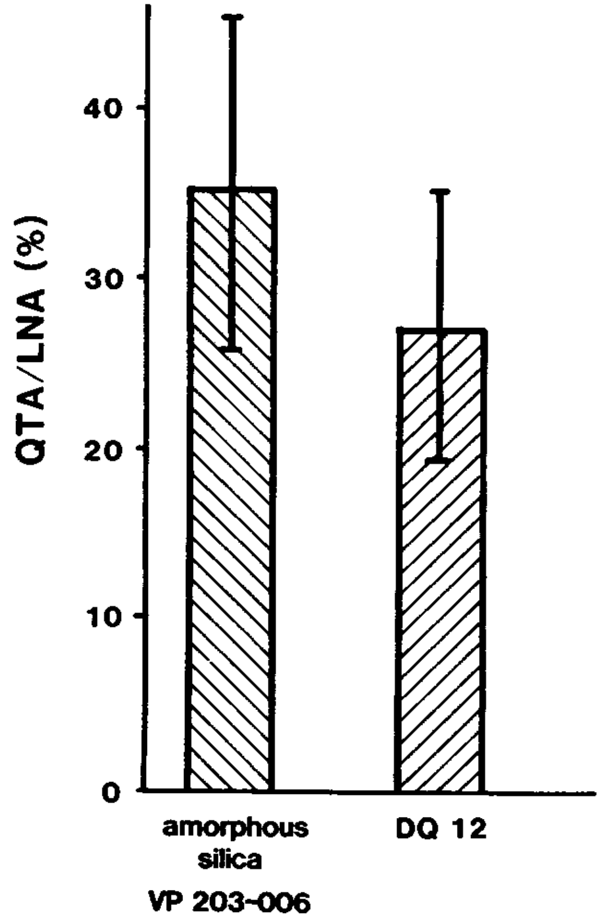


Figure 6. Cytotoxic effects (100-TCC-RA%) of 3 quartzes with different Al-content.[8]



Ffigure 7. Fibrogenic effects in lymph nodes of rats by amorphous silica (VP 203–006) and standard quartz DQ 12.[10]

- *In vivo* experiments, however only in a 2nd phase after positive completion of this lst phase, and
- Epidemiological examinations on selected groups of miners who, at least during the first 5 years of their employment, were exposed to dust in only one stratographic horizon.

The aim of these investigations is to be able to characterize the specific fibrogenicity of a quartz dust or any other mineral dust using a measurable parameter k. In addition to the quartz concentration $c_q$ used alone to date to determine the fibrogenous risk potential $R_F$ of a dust

$$RF = f(c_q, t, I) \qquad (1)$$

(in addition to the exposure time t and individual susceptibility I), it would then be possible to introduce specific factors k, $k_1$ and $k_2$ for the fibrogenicity potential into this risk equation:

$$R = f(c, k_1 \cdot c_q, t, I) \qquad (2)$$

$$R = f(k_2 \cdot c, k_1 \cdot c_q\ t, I) \qquad (3)$$

or simply

$$R = f(k \cdot c, t, I) \qquad (3)$$

where c is the concentration of the total respirable dust and $c_q$ the concentration of the respirable quartz dust.

Based on this type of analysis, it would then be possible to obtain scientifically corroborated limit values which can be different for the individual stratigraphic horizons or deposits. Furthermore, this method could then be applied to all other sectors of industry in which fibrogenous dusts occur.

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# SEEKING THE "RANK FACTOR" IN CWP INCIDENCE: ROLE OF RESPIRABLE DUST PARTICLE PURITY

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## INTRODUCTION

The search for the cause(s) of coal workers' Pneumoconiosis continues despite the vast amount of research that has been undertaken over the past forty to fifty years. Good correlation between the mass of dust inhaled over workers' lifetimes and the incidence of the disease has led to the development of effective environmental standards and implementation of good procedures for the control of dust generation and dispersion in the working place. Research has only begun, however, to uncover the biochemical mechanisms involved in initiating the disease.

Correlation between the rank of the coal seam in which miners worked and disease incidence and severity is acknowledged worldwide, but the agents in the higher rank coal seams which cause CWP are still not defined. The basic components of the coal seams, i.e., the elements and minerals, are generally known, but the way in which they interact with human pulmonary cells is largely still a mystery.

The process of disease development is a biochemical one which depends on the characteristics of the pulmonary cells within their immediate biochemical environment and the characteristics of invading respirable coal mine dust particles, probably both chemical and physical ones. The cell-particle relationship, therefore, must be defined, and both cells and particles must be characterized according to properties which potentially are involved in the disease process.

Following such characterization, specific respirable dusts from different coal seams, which appear to have different effects on coal workers, must be obtained or constructed and then used in experiments designed to determine dust toxicity or the response of animals exposed to the dusts.

The results presented here focus on characterizing properties of respirable coal mine dust particles collected from operating longwalls in West Virginia and Virginia. Longwall panels were selected for analysis because the respirable dust in the coal face areas is largely uncontaminated by rock dust and analysis of the dust yields results unconfounded by rock dust.

The major emphasis has been placed on the characterization of mineral particles, although the analysis is presently being extended to include coal particles as well. This approach was selected because bulk analyses of dusts do not give the level of detail needed to unlock the key elemental and mineralogical roles in the disease process and because the role of minerals in disease development is contradictory at present. Another reason for this emphasis hinges on the known toxicity of quartz and the possible interaction of clay minerals with quartz in reducing overall dust toxicity.

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## THE ROLE OF QUARTZ AND OTHER MINERALS

Although a majority of the studies on the role of quartz in CWP suggest a relationship does exist, some researchers have found contradictory evidence.[1] Interaction of quartz with other minerals may be a factor in reducing the influence of quartz in the disease process. Martin et al.[2] showed that mineral matter in coal inhibited the fibrogenic effect of quartz on experimental rats. They suggested that the surface characteristics of quartz particles were modified, thereby reducing their fibrogenic response.

Leiteritz et al.[3] found that the incidence of CWP in West German mines was related to the concentration of quartz in the dust and to the fine dust concentration whenever similar mines were compared. Interestingly they found a lower amount of quartz in higher rank coals. Davis et al.[4] found that noncoal minerals, including quartz and clay minerals, increased in concentration as the severity of CWP increased. They concluded that quartz is not the only cause of disease progression. Reisner and Robock,[5] studying the cytotoxicity of respirable coal dusts in the German Federal Republic, found that cell damage was positively correlated with the mineral content of dust and with coal rank and age as the mineral content was controlled.

These results clearly indicate an interaction between quartz and other minerals, most probably the clay minerals, in the disease process. It is evident that the mix of the minerals in respirable dusts, the intimacy of the mixing (i.e., whether clay mineral particles are bonded to quartz particles, and where, or merely lying adjacent to them), and the size fractional relationship among the minerals may all be important aspects for continuing research. Further, a special relationship, which is yet to be defined, exists between these factors and the rank of the coal seam.

Toxicity tests on quartz, clay minerals, and specific mixes of the two, similar to the research presently being conducted by Wallace et al.,[6] and exposure-response experiments on animals using specific, representative mixes of minerals in dusts may be crucial to understanding and defining the mechanisms involved in CWP. The types of dusts which should be used in these experiments remain to be defined. They certainly need to be representative of the respirable dusts existing in working mines, and a number of different dusts, some from apparently harmless environments and others from toxic ones, need to be used and results compared. A methodology for characterizing and describing such representative dusts will be presented next.

## METHODOLOGY

In order to study the characteristics of respirable-sized dust particles, dust samples were collected from longwall faces in accordance with 30 CFR 70.2 precollector. Samples were taken at seven to nine different locations on panels depending on the manpower configuration and the distribution of ventilating air. In an effort to ensure the representativeness of the dust particles from a coal seam, full-shift sampling was done over three different shifts, resulting in twenty-one to twenty-seven samples being obtained from each panel (three at each location sampled).

Scanning electron microscopy interfaced with energy dispersive x-ray analysis (EDXA) was selected as the procedure best able to study dust particles in the detail required. Due to the limitations of the in-house scanning electron microscope (SEM), which had to be operated manually, a strategy of identifying and characterizing 100 mineral particles per sample was adopted during particle analysis. An image analysis system was not available, thus particle sizing and shape analysis had to be done manually.

Dust samples from two different longwall panels operating in coal seams of significantly different rank were analyzed under the SEM. The Beckley seam is of higher rank (14,670 Btu, 18.8% volatile matter) than the Pittsburgh seam (13,660 Btu, 39.7% volatile matter). An elemental spectrum had to be obtained under EDXA for anywhere from 800 to 1200 particles in order to determine their mineralogy before 100 minerals could be identified. After a mineral particle was identified, it was sized according to its breadth (Feret's diameter) and ordinally ranked for its general shape and angularity of its periphery.

Analysis of 2684 particles was made from the Pittsburgh seam samples, while 700 particles were analyzed from the Beckley seam before obstacles to further progress were encountered. The data was then analyzed for variations in mineralogy by location on each longwall panel, for variations in mineralogy by coal seam, for mineral particle size variation by location on each longwall panel, for mineral particle size variation by coal seam, for differences in shape and angularity, for size-fractional mineralogy, and for the purity of mineral particles with respect to normal stoichiometric elements. Descriptive statistics and analysis-of-variance (ANOVA) techniques were used to pinpoint differences in the various parameters.

## PRIMARY FINDINGS

Five major minerals were found to exist in the two coal seams: calcite, illite, kaolinite, quartz, and pyrite. The Beckley seam samples contained a substantially higher percentage of calcite particles (32.7% versus 20.7%) and kaolinite particles (10.3% versus 2.0%) than the Pittsburgh seam, while the relationship with illite particles (35.6% versus 55.8%) and pyrite particles (1.3% versus 6.0%) was reversed. The percentage of quartz particles (8.5% versus 8.7%) was virtually the same for each seam.

Although the percentage of quartz particles was the same for each seam, the purity of the quartz particles was very different. Only 28% of the quartz particles analyzed for the Pittsburgh seam were uncontaminated by non-stoichiometric elements, whereas 64% of those analyzed for the higher rank Beckley seam were uncontaminated. This information could potentially explain the contradictions on the role of quartz in CWP incidence since stoichiometric quartz is known to be cytotoxic. Contaminated, and hence toxically turned off, quartz particles may also be the key factor in defining the protective role that clay minerals have appeared to play in some studies.

In general, twice as many mineral particles are pure in the Beckley seam samples compared to those analyzed from the Pittsburgh seam. This finding is consistent for each mineral. This may be the rank factor alluded to in much of the literature concerning CWP, and this rank factor may indicate the geologic processes occurring during the time the minerals were formed. Hence, quartz occurring in the Beckley coal seam and in the overlying sandstone stratum may have been formed without major interaction with alumino-silicate clays, which were found to be the chief contaminants in the Pittsburgh seam quartz particles analyzed.

The mineralogy of respirable-sized particles was found to vary significantly by location on a longwall panel. For example, in the Pittsburgh seam the percentage of illite particles increased quadratically at a decreasing rate along the coal face from the fresh air point at the headgate (13%) to the farthest point along the face at the tailgate (77%). The percentage of quartz particles, on the other hand, remained nearly constant at each location. The amount of contamination of mineral particles by nonstoichiometric elements also remained nearly constant along the panel.

A major conclusion is that mineral particles comprising strata formed by sedimentary processes are as heterogeneous as coal particles. There is no such thing as standard quartz, illite, kaolinite, pyrite and calcite particles. This is especially true in the lower rank coal seams in West Virginia as compared to the higher rank coal seams. Therefore a trend exists toward relative mineral particle purity according to the rank of the coal seam.

Mineral particles identified from the Beckley seam were smaller, in general, than those from the Pittsburgh seam (1.36 versus 1.58 micrometers), although the size of quartz particles in the Beckley seam were larger than those in the Pittsburgh seam. Mineral particle size was also found to be significantly smaller at the tailgate location than at any other location on a longwall panel (1.17 versus 1.58 micrometers for the Pittsburgh seam). Particle mineralogy by size fraction was found to vary significantly, although differently for each coal seam studied.

## CONTINUING RESEARCH

Research priority for this project is now focusing on determining the percentage of all respirable dust particles that are single phase or multi-phase minerals versus single phase organic or multi-phase organic-inorganic complexes. This effort is being accomplished using a semi-automated procedure involving particle imaging followed by EDXA. Both coal and noncoal particles are being analyzed, and size fractional variations are being pursued. The mineral purity factor will be developed further, but additional emphasis will be focused on determining the extent of inorganic inclusions in organic particles. Initial results are just being obtained from this procedure, which will analyze 2000 particles per sample. Important size distributions for both coal and mineral particles will result from this research.

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# THE INFLUENCE OF SHAPE, SIZE AND COMPOSITION OF INDIVIDUAL DUST PARTICLES ON THE HARMFULNESS OF COALMINE DUSTS: DEVELOPMENT OF METHODS OF ANALYSIS

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## INTRODUCTION

The relationship between dust exposure and the probability of developing simple pneumoconiosis in UK coalmines was established in the field research programme over a period of more than 25 years (Figure 1). However, as demonstrated by Hurley et al. (1982) and Walton et al. (1977) the data for certain collieries do not fit the model developed for the main group. Some high rank collieries have higher probability of developing pneumoconiosis than predicted by the model from dust exposure data while other low rank collieries have much lower probabilities. Similar patterns are found for the same collieries in the development of progressive massive fibrosis.

Table I shows details from two of these anomalous collieries. Colliery Q is a low rank coalmine with moderate dust and quartz exposure but low prevalence of pneumoconiosis and observed:expected ratio of about 18% in the Hurley (1982) model. In the Walton (1977) model when other factors are involved including the mineralogy and interactions between minerals the observed:expected ratio is close to 100%. In contrast, with Colliery W, a high rank mine, the dust exposure is moderate and the quartz exposure is low but the prevalences are high and the observed:expected ratios from both models are around 200%. Thus while the dust composition data may be used to explain the lower prevalence in some low rank mines there is still no satisfactory explana-

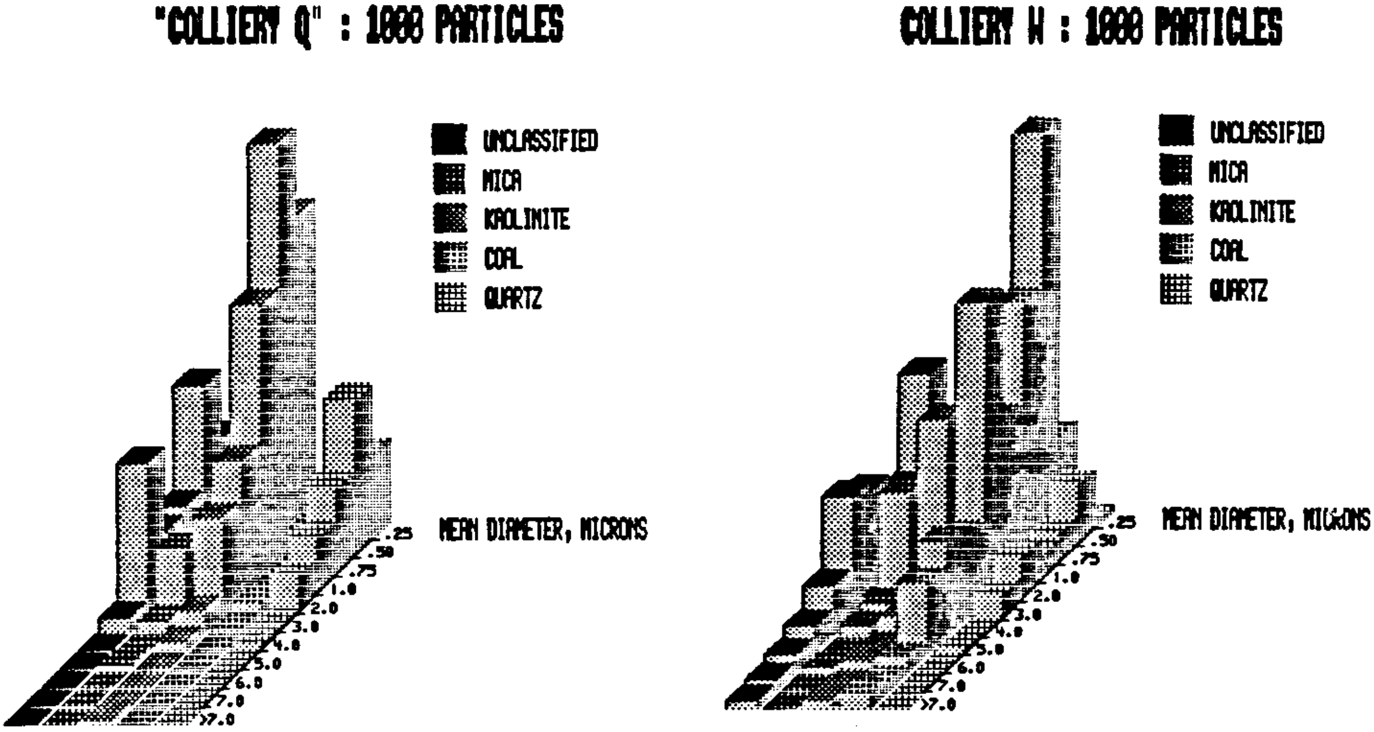

Figure 1a and b. Typical particle size and composition data derived for a low rank coalmine Colliery Q and a high rank coalmine Colliery W showing clear differences in the coal quartz and kaolinite particle sizes and proportions.

Table I

Epidemiological Data from a High Rank and a Low Rank Colliery
Showing the Differences in Fit to their Statistical Models
Devised by Walton (1977) and Hurley (1982)

| | **Colliery W**<br>**High Rank** | **Colliery Q**<br>**Low Rank** | |
|---|---|---|---|
| **Dust Concentration (mg $m^{-3}$)** | 5.0 | 5.9 | ) **Mean of** |
| **% Quartz** | 1.4 | 5.4 | ) **11 years** |
| **% Ash** | 17.4 | 70.6 | ) |
| **Observed/Expected 2/1+** | 182 | 18 | |
| **Observed/Expected 0/1+** | 214 | 99 | |
| **Prevalence 0/1+** | 18.3 | 3.3 | |
| **Prevalence 2/1+** | 6.0 | 0.8 | |

tion for the higher prevalence of simple pneumoconiosis and PMF in the high rank mines.

In the early stages of PFR programme it was observed that many of the coal particles on thermal precipitator slides from high rank collieries appeared to be very much larger than expected. These observations were also reflected in the ratios between particle number and respirable dust mass concentrations established in the programme (Jones 1974). Similarly, particles with large projected areas have now been seen in a range of respirable mine dust samples and in dust samples recovered from the lungs of coalminers suggesting that these large particles in fact have small aerodynamic diameters. It seems likely that these particles from high rank collieries are plate-like. If so then like long fibres they may also be difficult for lung clearance. This factor has not yet been considered in epidemiological studies.

In the current project, supported by the European Coal and Steel Community, we are setting up a method to measure, rapidly and automatically, the shape, size and composition of individual particles in the dusts and to find out if the differences suggested by these earlier observations are real.

## METHODS

The method is based upon a Cambridge Instruments Scanning Electron Microscope with a Link Analytical LZ-5 light element X-ray detector, both controlled by a Link AN 10,000 multi-channel analyser and image processor.

The method has been applied to airborne dust samples loaded on polycarbonate membrane filters examined using a secondary or back-scattered electron image in the first instance.

The EM image signal is acquired by the AN 10,000 and digitised. The signal intensity at each point is measured and features are detected by searching for intensities above a given threshold level. The positions and sizes of all features larger than a given minimum are recorded to a results file and the computer then directs the electron beam to each one in turn. The X-rays generated are collected and analysed and the chemical composition is recorded with the other data in the file. This is repeated over many fields of view, building up data on more than 1000 particles in each sample.

The results file is then compared with a set of chemical classes based upon the compositions of the expected minerals. Each particle is classified and size distributions are prepared for each class. The time taken for each set of 1000 particles is about 2 hours.

A large number of factors are defined by the user for feature detection, feature analysis and data evaluation.

## FEATURE DETECTION

For feature detection, only the setting of the threshold signal levels caused any real difficulties. Initial work with the secondary electron images showed insufficient contrast between the smaller particles and the background making particle detection very unreliable. We avoided this and other problems by adopting the techniques of Scanning Transmission Electron Microscopy to the SEM. Routine sample preparation methods identical to those used for airborne asbestos dusts were used.

The advantages of this change include low image noise and very low sensitivity of the thresholding routines to gross signal level changes. In addition, there is a very much lower level of background noise in the X-ray counting routines. The disadvantages are in the additional sample preparation necessary although there are possibilities for direct impaction of samples on to pre-coated TEM grids.

## X-RAY ACQUISITION

Only small sections of the full 10 KeV X-ray spectrum are necessary for particle identification. The minerals of interest in these samples are Coal, Quartz, Kaolinite and Mica, and the elements required to identify them are carbon, oxygen, aluminum, silica and potassium. Suitable X-ray energy ranges or windows are set up to count only the X-ray's characteristic of these elements and to provide suitable background counts for subtraction from the element counts.

X-ray counts of any duration can be made up to hundreds of seconds but counts for 1500 milliseconds have been found to be sufficient for classification and suitably fast.

## DATA PROCESSING

Mineral composition limits must be described for all of the elements in the X-ray window file for each mineral of interest in order that the particles may be classified. The ranges of compositions, established as a standards file, are shown in Table II and illustrate some important features. The ranges do not represent precise compositions for a number of reasons. Firstly, the classes need to be broad to accommodate the statistical variation found with low X-ray counts resulting from short analysis times. Secondly, the breadth allows for a degree of cross-contamination of X-ray spectra by adjacent particles or by fine particles on the surfaces of larger ones. At the same time, however, a wide range of compound particles will remain unclassified as will such minerals as $CaCO_3$ and $FeS_2$.

## SIZE DISTRIBUTION

The only remaining task for the data processing is the size distribution calculation. This may be undertaken for any of a range of parameters, including maximum, minimum or mean Feret diameters, projected area, circumference etc., or any ratio of these. Any range of sizes in any number of size classes may be established for the calculations for each of the mineral categories. In the development work carried out to date mean projected diameter has been used to describe particle size.

## RESULTS

Typical results are shown in Figures 1a and 1b for the high rank and low rank coalmine dusts. The low rank coalmine dusts from Colliery Q show a low proportion of coal particles, high kaolinite and moderate mica and quartz. Almost all of the particles are less than 2 µm mean projected diameter. In contrast the high rank dust from Colliery W contains a much higher proportion of coal particles, very low amounts of quartz and moderate-low amounts of kaolinite and mica. While most of the mineral particles are less than µm in projected diameter a large number of the coal particles are considerably larger and would constitute an even higher proportion of the mass. The actual mineral compositions of both dusts as determined by normal analytical methods are reasonably consistent with the proportions in the mineral categories shown.

The two respirable dusts illustrated here should contain similar aerodynamic size ranges since they were collected in the same way using the same sampling instruments. The proposed explanation for the larger particles in the anthracite dust is that they are flat and plate-like and therefore aerodynamically small. Confirmation of this from three dimensional image analysis is also in progress.

## DISCUSSION AND CONCLUSIONS

There is no single unifying physico-chemical theory of toxicology for all coal mine dusts. For example, the proportion of quartz in the dust, the nature of its surface, the mitigating effects of the clay minerals and the nature of the coal particle surface (oxygen free radicals) have all been used to explain observed differences in toxicity. If large plate-like particles, like long thin fibres, are difficult for lung clearance mechanisms to handle then the shape and size of the coal particles may also be important factors. This may then be the simplest hypothesis to explain the prevalence of coalworkers simple pneumoconiosis and progressive massive fibrosis in South Wales.

The preliminary conclusions from the study so far are two:

1. The method as established using STEM can be used to provide rapid automatic shape, size and composition information on a large number of individual particles.

Table II

Composition Ranges of "Standard" Minerals Used for Discrimination of Minerals from X-ray Counts

| | **Coal** | **Quartz** | **Kaolin** | **Mica** |
|---|---|---|---|---|
| **C** | 20 - 100 | <20 | <30 | <30 |
| **O** | <35 | 15 - 80 | 15 - 70 | 15 - 70 |
| **Al** | <20 | <15 | 15 - 70 | 15 - 70 |
| **Si** | <20 | 15 - 80 | 15 - 70 | 15 - 70 |
| **K** | <5 | <5 | <5 | 5 - 30 |

**Ranges are broad and do not reflect precise compositions:**
**Allows for low X-ray counts (short analysis times)**
**Allows for some compound particles, adjacent particles, sub-µm particles on surfaces of larger ones**
**Still wide range of 'unclassified' compositions [e.g. Ca $CO_3$, $TiO_2$]**

2. There appear to be real differences in the shapes and sizes of the coal minerals in dusts from coalmines of different rank.

Once it has been fully developed we believe that this method of dust analysis will have a useful role to play in epidemiological studies of coalworkers' pneumoconiosis.

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# HARDGROVE GRINDABILITY INDEX OF COAL AND ITS RELATIONSHIP WITH COAL WORKERS' PNEUMOCONIOSIS

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The prevalence of coal workers' pneumoconiosis (CWP) is generally considered to be directly related to the rank of coal, (1) mass of respirable dust, (2) free silica in coal, (3) and a few other factors such as trace elements (4) and diesel fuel emission.

The present study emphasizes on the mass of dust and dust generation potential using indirect measuring technique. Hardgrove Grindability Index (HGI) of coal is a measurement of the resistance to abrasion and crushing and therefore an indirect measurement of generation potential of fine particles and dust upon crushing. HGI is used in this study because of two reasons: (1) there is a large published data bank available, and (2) there seems to be a better correlation between HGI and coal dust level than any other factors. Rank of coal has long been recognized to have positive correlation with coal workers' pneumoconiosis (CWP). HGI values of coal is not only controlled by rank but also by mineral (ash) content and maceral composition and the size of vitrain bands.[5] Dull coals (durain rich coals) tend to be hard and exhibit lower HGI than bright coals (vitrain rich coals) of the same rank. Mineral matter, particularly dispersed, fine grained mineral matter, acting similar to other inertinite macerals, tend to cause a decrease of the HGI of the coal. More than 2000 entries of HGI of United States coals were published by the U.S. Bureau of Mines and about half of them came from the states of Pennsylvania and West Virginia.[6] There are sufficient data points from these two states to make an acceptable 2 evaluation of any relationship between HGI and CWP, also based on published data.[7]

Results of evaluation of available Pennsylvania and West Virginia data indicate that there is a very good correlation between Hardgrove Grindability Indexes of coals (Figure 2) and the prevalence of CWP (Figure 3). Both plots are presented here on a county basis for easy comparison because the CWP result was published on a county basis. High HGI values come from high rank bituminous coals (low volaltile bituminous) occurring in those counties along the Allegheny front immediately west of the Appalachian folding belt that coincide well with the rank distribution of the coals. No petrographic data are available for detailed studies of the effect of maceral composition and HGI. Lower Kittanning coal is generally known in this region as a dull coal and rich in durain. On the other hand the Lower Freeport coal is a bright coal. Figure 1a is a plot of the frequency distribution of HGI of all available Lower Kittanning and Lower Freeport coals in Pennsylvania and West Virginia. There seems to be more Lower Kittanning coals exhibiting low HGI number than Lower Freeport coals, suggesting potential effect of the differences in petrographic composition. To further refine the technique and to remove the strong influence of rank differences, data from a single county (Clearfield County, Pennsylvania) were plotted (Figure 1a) and exhibit similar result. Petrographic effect is minimized in very high rank (also with high HGI, for example low volatile bituminous) coals because of the convergence of macerals in which individual macerals are progressively indistinguishable. Comparison between Lower Kittanning coal and Upper Freeport coal (also a bright coal) shows a similar result.

In summary, Hardgrove Grindability Index can be a good indicator of prevalence of coal workers' pneumoconiosis because it encompasses many of the dust generating factors such as rank, ash content, petrographic composition, and other factors yet to be isolated. If all other factors are the same the coal that tends to generate the most dust could also cause more CWP.

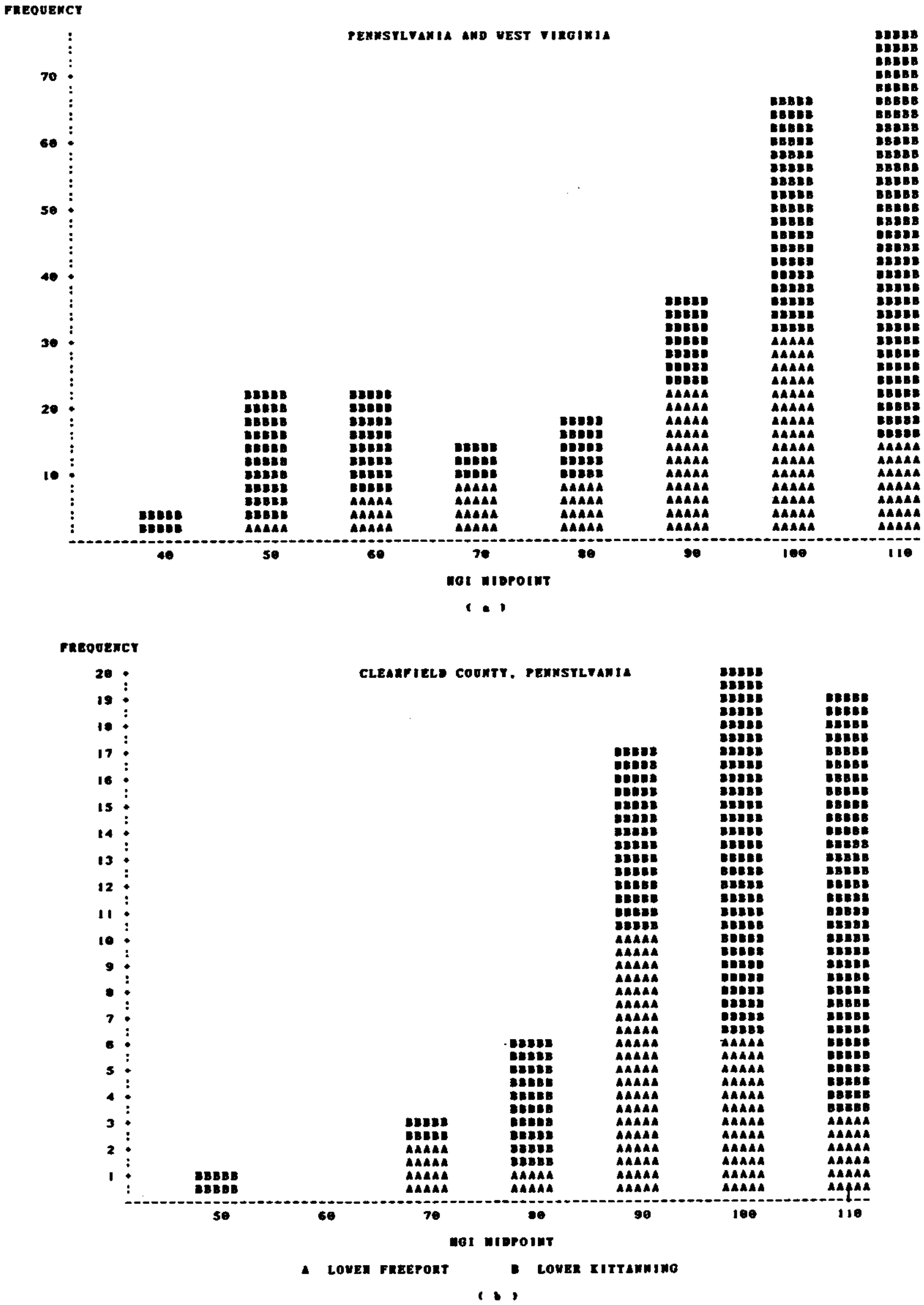

Figure 1. HGI frequency distribution of Lower Freeport and Lower Kittanning coals.

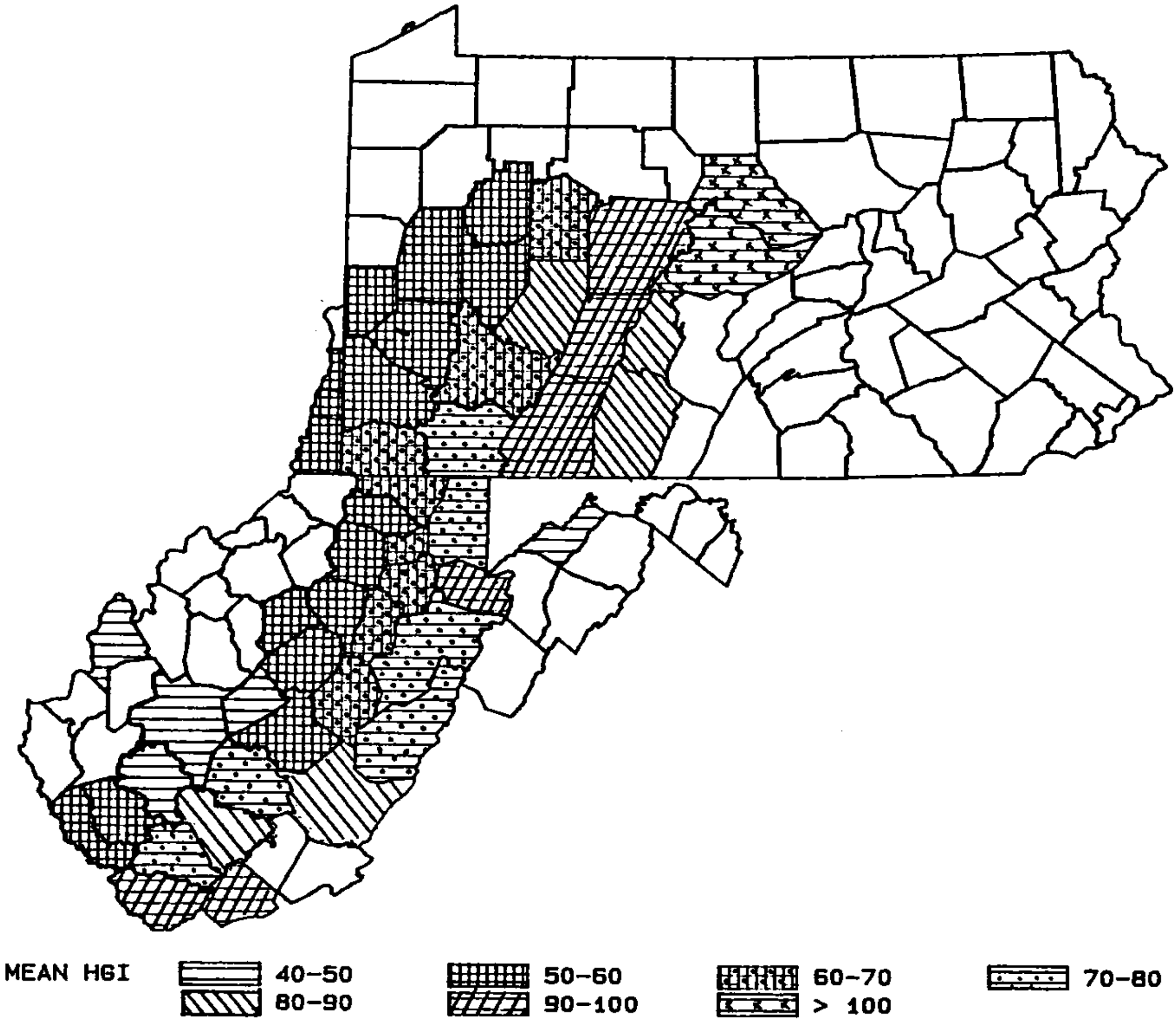

Figure 2. HGI averages for WV and PA.

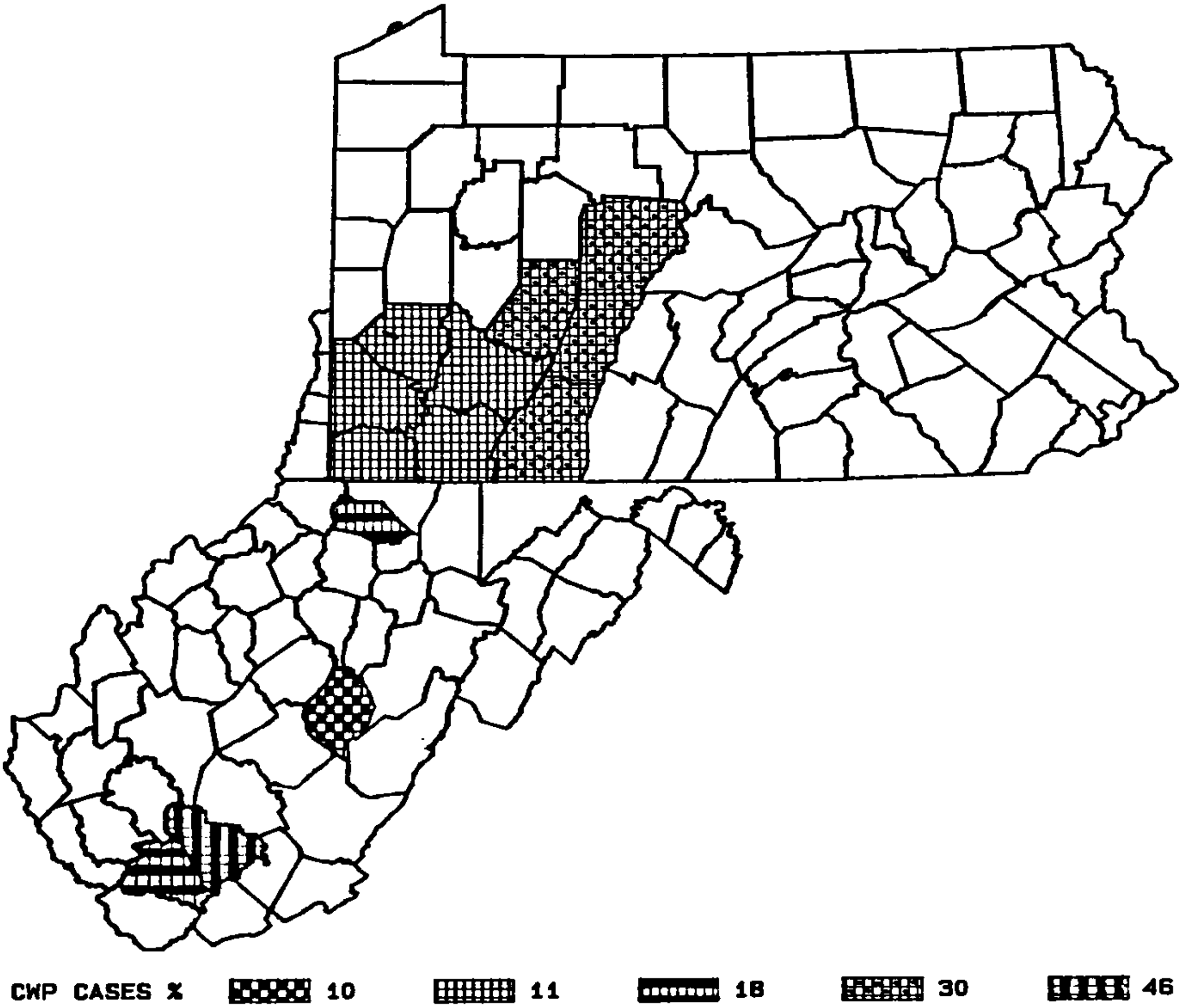

Figure 3. CWP cases for WV and PA.

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# MINERAL CONTENT VARIABILITY OF COAL MINE DUST BY COAL SEAM, SAMPLING LOCATION, AND PARTICLE SIZE

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## ABSTRACT

Size-selective airborne dust samples were collected using 4-stage cassette impactors at 9 different locations in each of five coal seams in order to determine the mineral content and its variability as a function of coal seam, sampling location, and particle size. Coal seams investigated were the Upper Freeport, Pittsburgh, Kittaning, Coalburg, and Pocahontas. Mineralogical analyses were done by an X-ray powder diffraction photographic technique.

Common minerals found in coal mine dust were illite, calcite, kaolinite, quartz, siderite, dolomite, gypsum, anhydrite, and pyrite. Among these minerals, illite and calcite were the two dominating minerals followed by kaolinite and quartz. It was found that mineral content was significantly affected by coal seam and particle size. In contrast, no statistical significance was found between samples collected at different locations within a section and sections within a mine. The variability of the mineral content was found to be quite high, with the mineral specific CV being .5 or greater.

## INTRODUCTION

In spite of its long history of incidence, and the large amount of research conducted on it, coal workers' pneumoconiosis (CWP) is still prevalent among coal miners and needs further research to develop effective preventive and remedial measures. Although the dose-response relationship between simple CWP and coal mine dust has been established,[1] the causal agent(s) and the mechanism(s) involved in the progression of simple to complicated CWP are not yet defined. Many plausible hypotheses have been made to explain the mechanisms involved in the disease progression, however, none of them is satisfactory. In addition, the source of variation in occurrence of CWP among miners in different geographic areas,[2] rank of coal seam,[3,4] and job[5] remain unsolved.

As a result, more epidemiological and medical efforts have been directed toward identifying the causal agent(s) by investigating the physical and chemical properties of coal mine dust. Among the many factors investigated, the mineralogical composition has received attention because past epidemiological and post-mortem lung tissue studies have shown some correlation with the prevalence and severity of CWP and the quantity and type of minerals found in the coal mine dust.[6-8] Such findings have inspired many toxicological studies designed to assess toxicity of coal minerals *in vivo* and *in vitro*. These investigations have reported toxicity of coal minerals although contradictory toxicological results on the toxicity of each mineral by itself and in combination have been reported.[9-12] Although these studies have provided us useful medical and toxicological information of coal minerals, some of them should be interpreted cautiously because the experimental settings used may not be comparable to actual mine situations. Furthermore, the studies provided little or no information about the "dust" used (i.e., dust size, mineral content, etc.). A recent review of one hundred toxicological studies and experiments designed to assess the health effects of exposure to coal and coal related minerals[13] revealed that, in many cases (43%), the geographic location where the test substance was obtained was not reported. Rarely was a specific seam or mine identified. The review also disclosed that many studies (67%) did not list how the test substance was obtained or created. Among those preparation methods reported, grinding or crushing of bulk coal samples was the main preparation technique used (22%). It was followed by collecting airborne mine dust at the coal face or in the return airway. Compounding the interpretation problem is the fact that the mineral composition of the dust was not incorporated for assessing dust toxicity in the majority of studies. In addition, most studies concentrated on evaluating the toxicity of a limited number of minerals and were not designed to assess the interaction of those minerals found in coal mine dust.

It is clear that toxicological studies designed to evaluate the effect of coal mine dust with different mineral combinations and concentrations are currently lacking because the mineral content as well as variability of coal mine dust has not been defined. Therefore, the purpose of this research was to identify the mineral matter contained in coal mine dust, to establish the variability of each mineral, and to find those factors that affect mineral content changes so that the results can be used as the basis of dust selection in toxicological research.

## EXPERIMENTAL PROCEDURE

### Sampling Location

Details of the sampling methodology have been reported previously.[14,15] Thus, only a brief summary is provided here. Six working sections from five mines, one section from each of four mines and two sections in one mine, located in the Appalachian bituminous coal field were investigated. In each mine, dust samples were collected at nine different locations on a continuous mining section. These locations include: the intake airway, the dinner hole, immediately before and after the continuous miner, immediately before and after the roofbolter, the feeder, the haulageway, and the return airway. These locations were selected such that the researchers could monitor the contribution of mining activities to the changes observed in the mineral content of the dust in the mine air as it moves from the intake side of the face to the return side. These locations are illustrated in Figure 1.

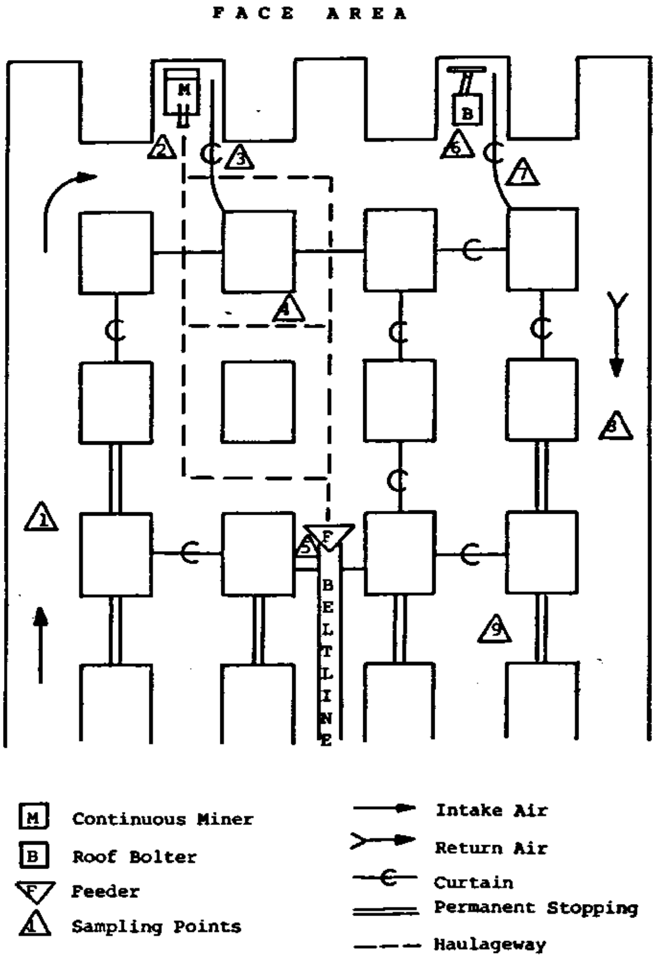

Figure 1. A typical continuous mining section layout and the sampling locations selected for study.

### Sampling Equipment

Dust samples were collected using 4-stage cassette impactors. They were designed and constructed using a modification of an original design described by Jones et al.[16] Figure 2 shows the exploded view of the cassette impactor. Aluminum foil was used as the collection substrate for stages 1 to 4 and a standard 37 mm, 5 μm pore size, polyvinyl chloride (PVC) membrane filter was used for the last stage. The aluminum substrates were brush coated with a mixture of Apiezon L grease and toluene to reduce particle bounce. Sampling was done at a flow rate of 5 liters per minute (LPM) to improve dust collection efficiency by increasing the Reynolds number (Re). The flow rate was maintained with three MSA Model G air sampling pumps connected with tubing and Y-connectors to a pre-calibrated precision rotameter. Figure 2 also shows the in-mine dust sampling equipment arrangement.

### Analysis

The analytical method used for the characterization of the mineral content of the dust samples was X-ray diffraction powder photography. The X-ray machine used was a Norelco X-ray generator type 12045/3 manufactured by North American Philips Electronic Instruments. The camera used was a Debye-Scherrer geometry camera of 114.6 mm diameter. The diffraction pattern was recorded on Kodak diagnostic, direct exposure DEF 329, GBX-2 film. Dust samples were mounted onto a thin glass spline mounted in wax on a copper stud. The sample to be X-rayed was placed into the camera which was then mounted on the X-ray generator. The aligned, rotating sample was exposed to copper Ka radiation monochromated with an Ni filter for 5 hours. The X-ray unit was operated at a voltage of 35 kilo-volts at 20 milliamperes. Each mineral was identified from the location of its diffraction line by comparing the film spectrum with the spectral data reported in the American Society of Testing Materials (ASTM) Powder Data File. Semi-quantitative estimation of the minerals present in the dust sample was accomplished by measuring the intensity of the diffraction lines and using weighting factors to compensate for the differences in the diffraction intensity of individual minerals. In this procedure, the intensity of each mineral was measured from the developed X-ray films using a microphotodensitometer. The height of each peak was measured after the estimated background which occurs primarily because of the grease and the organic coal dust matrix was subtracted. The raw intensities were then multiplied by weighting factors determined by Renton.[17] Mineral percentages were then calculated using the sum of weighted intensities as the denominator.

## RESULTS AND DISCUSSION

Mineral matter here is defined as the inorganic and discrete mineral grains. In this study, only those minerals commonly found with relatively high abundance (>0.1%) were analyzed.

The minerals identified in the airborne coal mine dust samples were calcite, illite, kaolinite, quartz, siderite, dolomite, gypsum, anhydrite, and pyrite. The term illite describes an illite

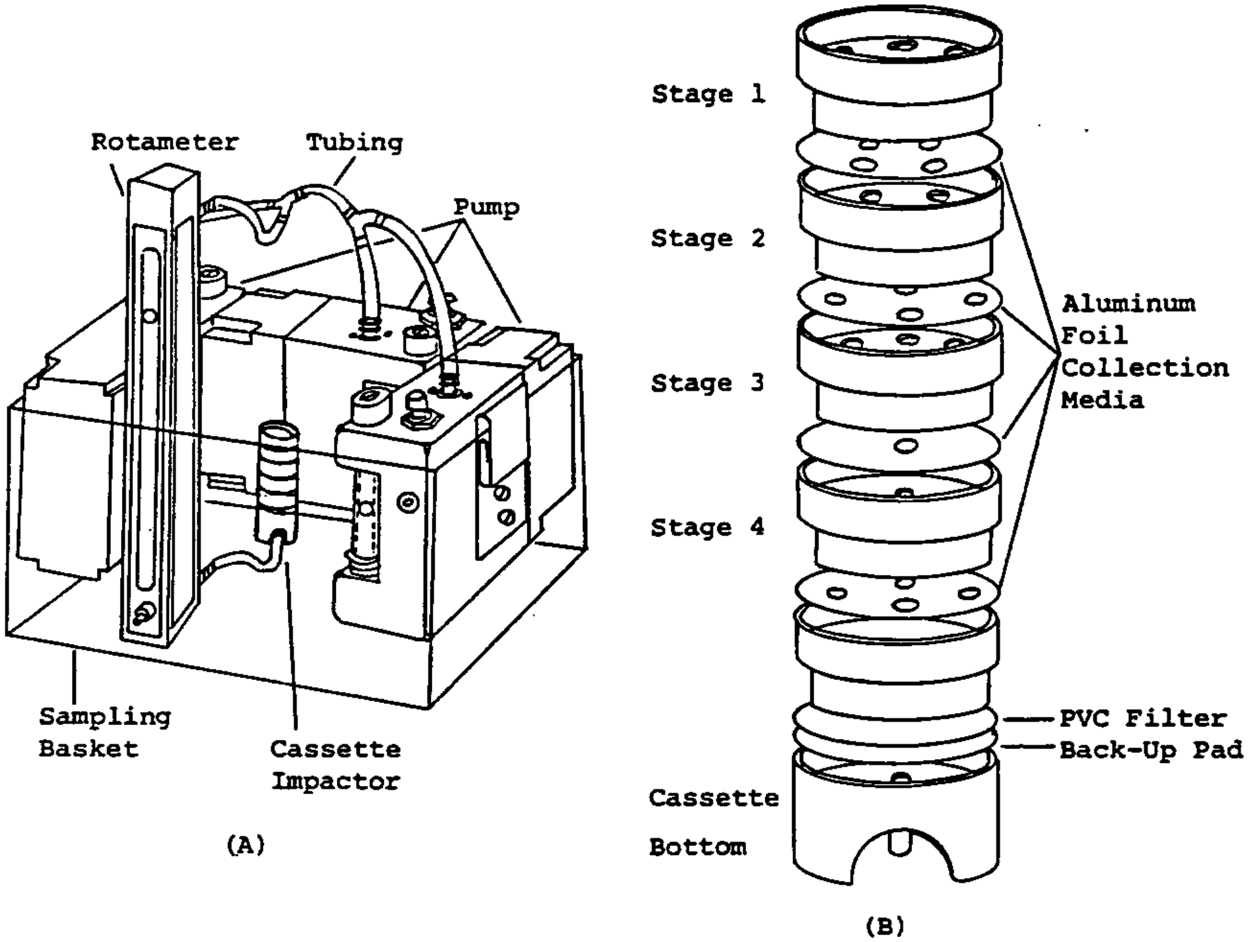

Figure 2. (A) Assembled in-mine sampling arrangement showing the pumps, tubing, and impactor. (B) Exploded view of the cassette impactor.

dominated mixed layered clay. The distribution of each mineral is depicted in Figure 3. Among these minerals, illite was the most dominant consisting of 43% +/− 22% of the mineral matter found in the samples collected. The other major mineral was calcite which amounted to 28% +/− 24% of the sample material. Similarly, kaolinite accounted for 9% +/− 8% of the sampled material, while quartz accounted for 4% +/− 4%. For these four minerals, the coefficient of variation (CV) ranged from 0.5 to 1.0. This is indicative of the very high variability found in the mineral content of coal mine dust. Other minor minerals, with concentrations ranging from 1 to 10%, included dolomite, siderite, and gypsum. The CVs for these minerals ranged from .68 to 1.47. Trace minerals with less than 1% concentration were anhydrite and pyrite.

In order to find factors affecting mineral content changes, the data were analyzed by a two-level nested-crossed analysis of variance (ANOVA). Subsequently, differences in mean values were evaluated using the Duncan's Multiple range Test. The ANOVA revealed that the coal seam factor was the cause of significant changes in mineral content for almost all minerals in the coal mine dust sampled in the region. Two exceptions were gypsum and siderite. A statistically significant high percentage of illite was found in the Coalburg seam while the lowest concentration was found in the Pittsburgh seam. Distribution patterns similar to that of illite were also observed for kaolinite and quartz. The distribution pattern for calcite, however, was almost opposite the patterns observed for the silicate minerals. Calcite content was the highest in the Pittsburgh seam while the lowest concentration was found in the Pocahontas and the Coalburg seams. In the case of such minerals as dolomite, anhydrite, and pyrite, they were more coal seam specific. The Pocahontas seam contained a significantly high (about 4 to 7 times) percentage of dolomite while more anhydrite was found in the Coalburg seam. Reportable amounts of pyrite were found only in the Upper-Freeport and the Pocahontas seams. The distribution of mineral content by coal seam is provided in Figure 4.

The working sections within a mine did not cause significant mineral content changes. No statistically significant changes in mineral content were observed between two working sections within a mine located in the Pittsburgh seam for all minerals except calcite. Similarly, the overall effect of

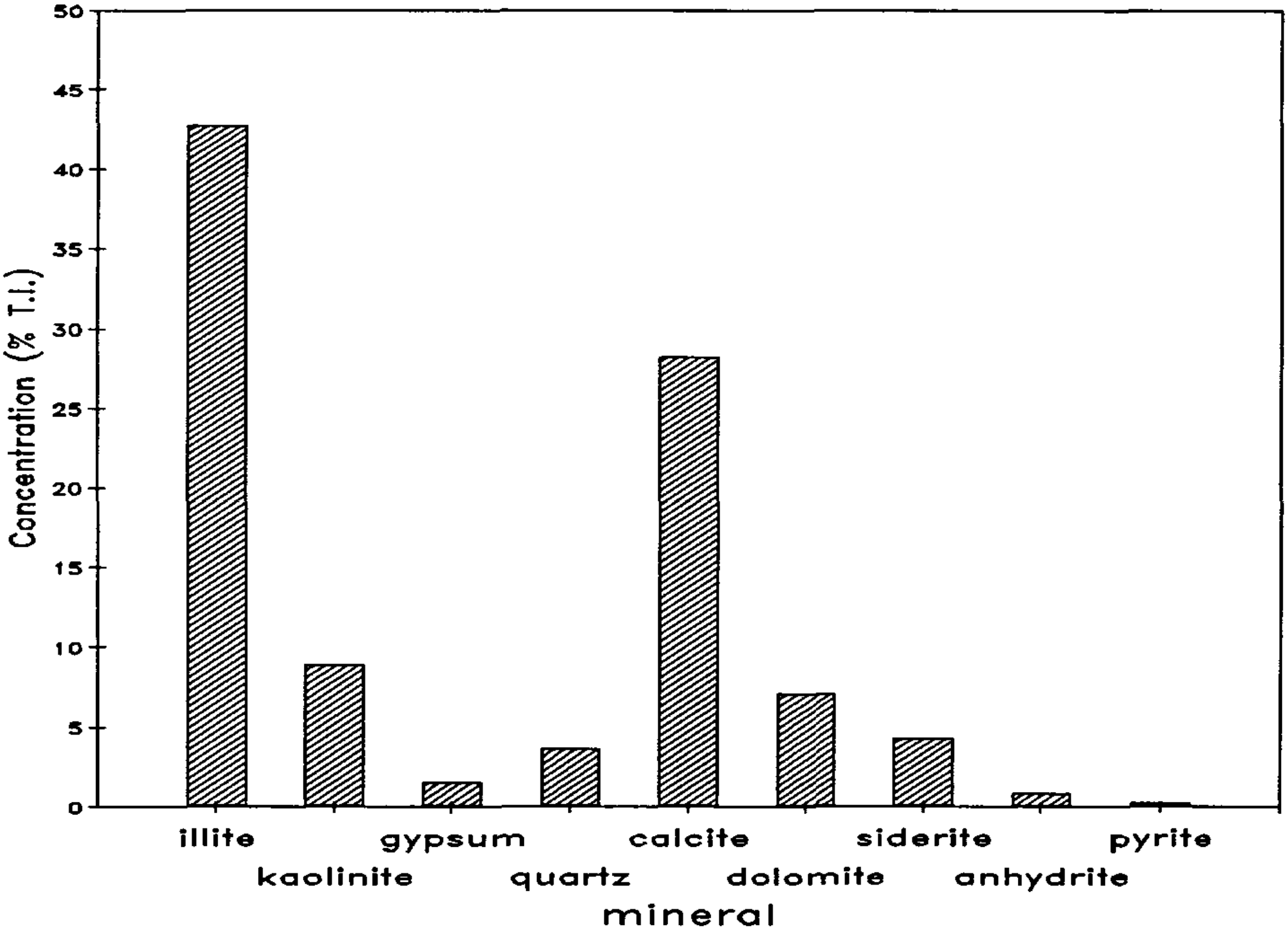

Figure 3.Overall mineral content distribution of the Appalachian bituminous coal field.

sampling location on mineral content was not statistically significant. Although no statistically significant difference was found, some trends were observed. The distribution of mineral content by sampling location is illustrated in Figure 5. High percentages of illite, kaolinite, and quartz in the samples collected near the continuous miner were observed while their concentrations were low in the samples collected in the return airway. Illite and kaolinite percentages were also relatively high in the feeder area. These results suggest that coal cutting and dumping liberates those minerals contained in coal. Their concentration then decreases as they gradually mix with other minerals as the mine air travels to the return airway. Quartz and siderite were relatively rich in the intake air samples while dolomite was found in the samples from near the roofbolter. Calcite content was the lowest in the coal producing area and the highest in the samples from the return air followed by the samples from the dinner hole. No particular patterns were observed for gypsum, anhydrite, and pyrite.

The distribution of mineral content by particle size is provided in Figure 6. The results of the ANOVA showed significant size effects on the mineral content for all minerals except kaolinite and pyrite. Subsequent analysis showed that the illite concentration was the highest in the size range of 1 to 3.5 µm followed by the size range of 6.6 to 10 um. The pattern for gypsum was similar to that of illite. For calcite, the trend was exactly opposite of the illite pattern. The highest concentration was found at the top stage (over 10 µm) followed by the third stage (3.5 to 6.6 µm). The pattern for quartz was similar to that of calcite although the highest concentration was observed on the third stage. Dolomite content was the highest on the top stage while no statistically significant difference in mineral content was found among the rest of the stages. The trend for siderite was exactly opposite of that of dolomite. Although some patterns of mineral content change as a function of particle size were observed, no general relationship was established.

## CONCLUSIONS

This research identified nine common minerals associated with coal mine dust in samples collected on continuous mining section in the Appalachian Bituminous coal field. Among these minerals, illite and calcite followed by kaolinite and

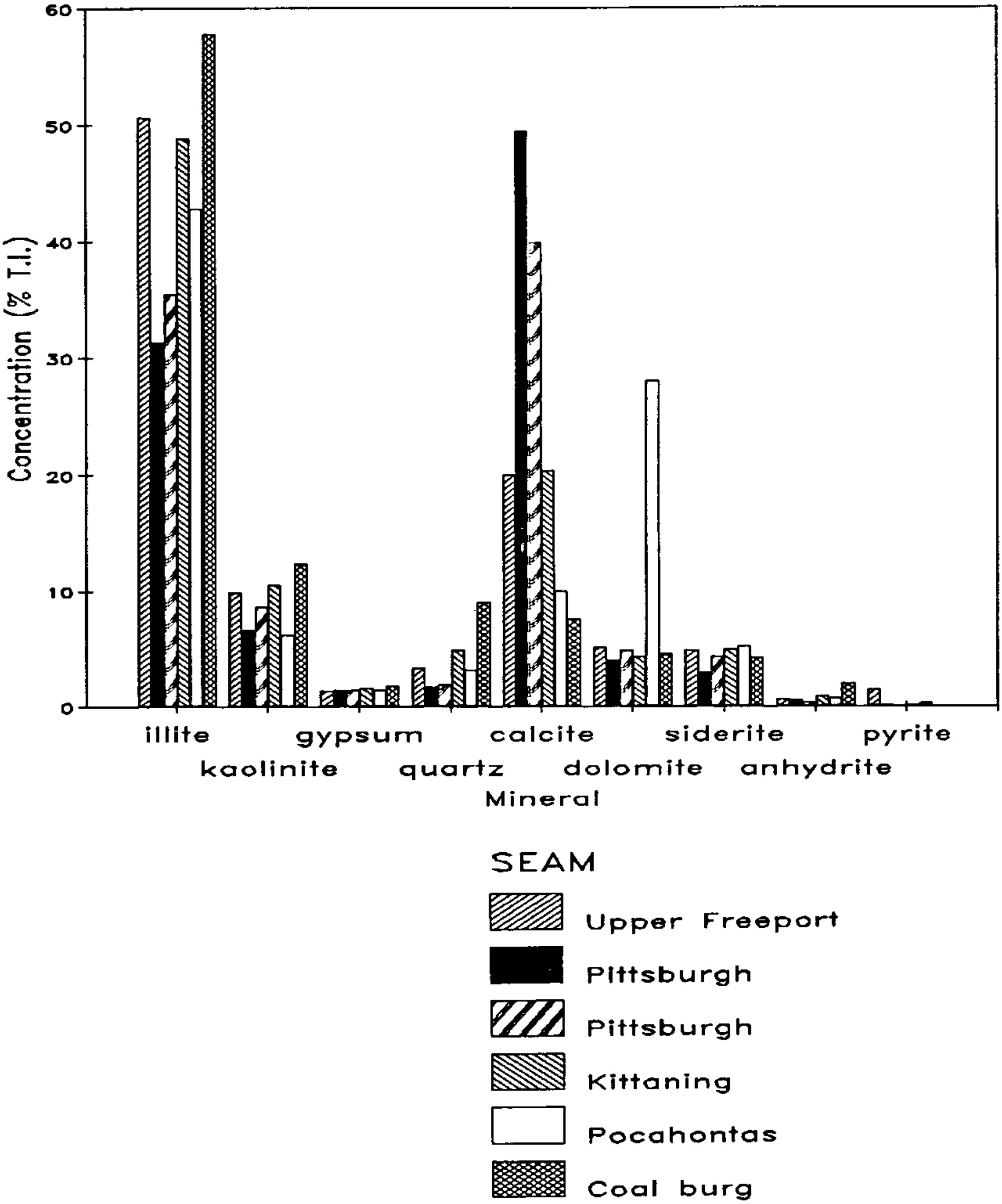

Figure 4. Mineral content distribution by coal seam.

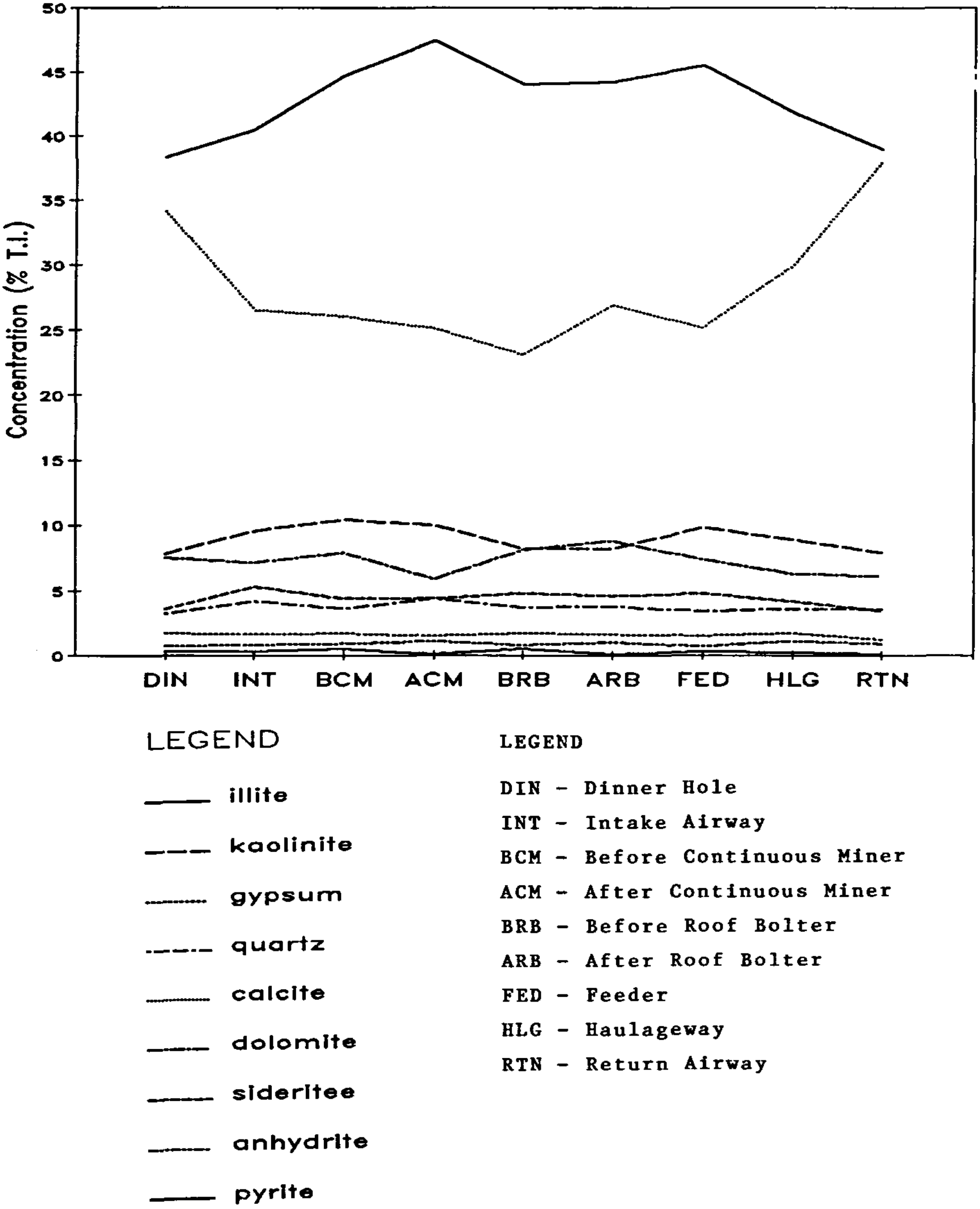

Figure 5. Mineral content distribution by sampling location.

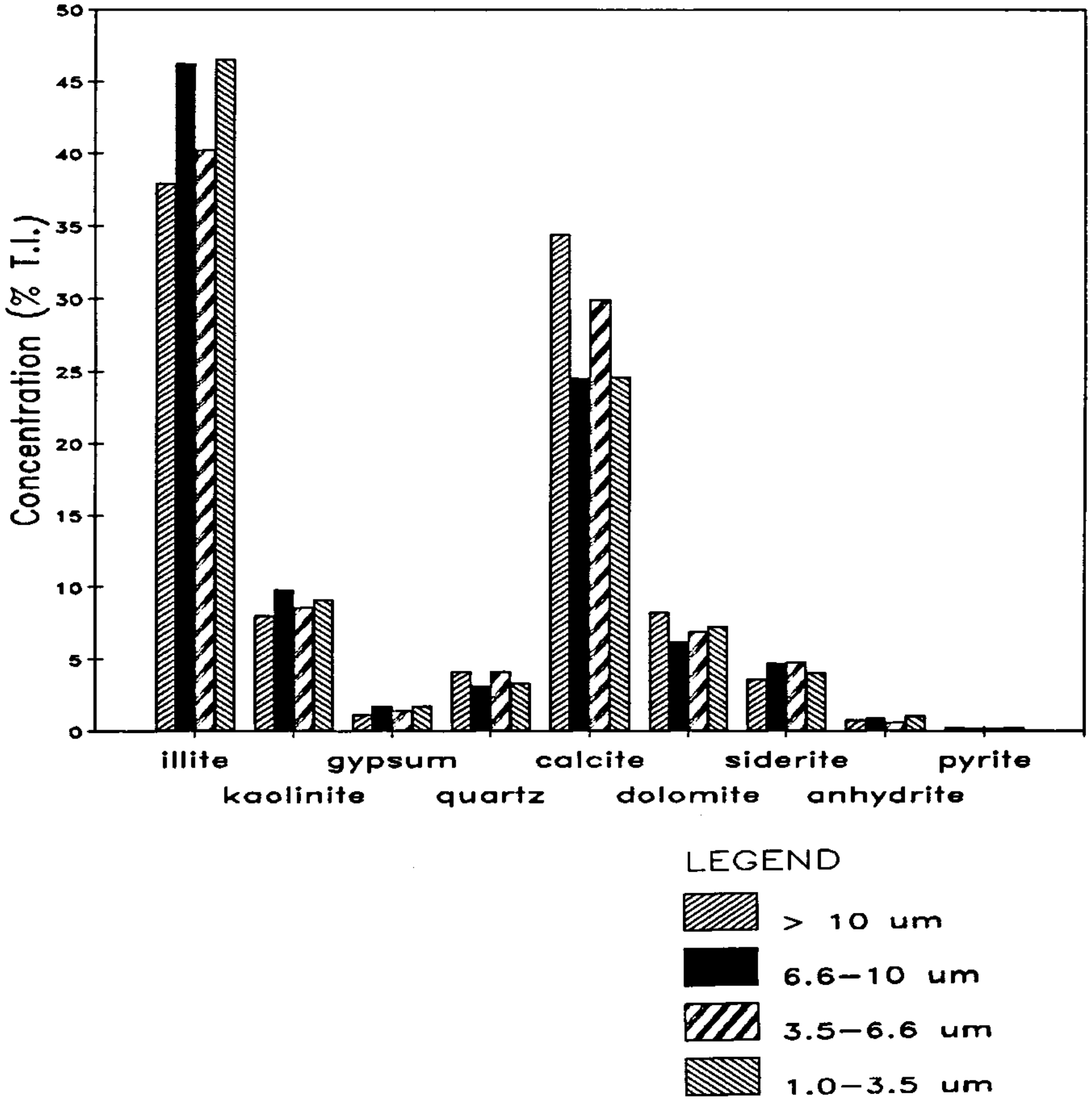

Figure 6. Mineral content distribution by particle size.

quartz are the dominant minerals. The relative abundance of all minerals except siderite and gypsum, however, is coal seam specific. This indicates the existence of coal seam variability. Also, mineral content was affected by particle size although no general relationship was established. The influence of sampling location upon changes in mineral content proved to be minimal. Likewise, no statistically significant difference was found between two working sections within a mine. However, significant variations in particle size distribution and respirable dust concentration were observed between sampling locations.

The results of this research clearly indicate that mineral content is highly variable and dependent upon coal seam and particle size. Therefore, it is clear that much of the previous medical and toxicological research on coal mine dust and CWP, which failed to consider, evaluate, and report the size specific mineral content of the administered dust can supply only limited information about the causal relationship between CWP and coal mine dust. This has left significant gaps in our knowledge of CWP causation and is at least to some degree, responsible for the conflicting results obtained by some of the past research. Thus, it is imperative that future research of this kind should carefully consider the physical and chemical nature of the "dust" used, and report in detail on the "dust" source, preparation method, and nature. This will allow appropriate interpretations to be drawn from the results, and subsequent research can be based upon it.

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# A COMPARATIVE ANALYSIS OF THE ELEMENTAL COMPOSITION OF MINING-GENERATED AND LABORATORY-GENERATED COAL MINE DUST

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## ABSTRACT

The occurrence of Coal Worker's Pneumoconiosis (CWP) has been potentially linked with several characteristics of coal such as rank, volatility, percent content of ash and non-coal components, quartz content, and the presence of several trace elements. According to the National Research Council, numerous epidemiological studies indicate that the incidence of CWP varies significantly with the composition and/or the concentration of the coal mine dust.[4]

Although advances in dust-suppression techniques have markedly reduced respirable-dust levels in underground coal mines, the National Research Council has concluded that chemical characteristics of respirable dust from different coal seams should be studied. With this objective in mind, research has been conducted in underground coal mines located in the eastern and midwestern United States, and in the laboratory to characterize the elemental composition of mining-generated airborne dust and laboratory-generated dust derived from samples taken from these mines. The goal of the research is to determine if a relationship exists between mining-generated and laboratory-generated dust from the same mine.

## INTRODUCTION

The Federal Coal Mine Health and Safety Act of 1969 was enacted to ensure healthier and safer working conditions for miners. In 1970, it provided for an underground respirable dust standard of 3.0 mg/m$^3$ in active coal mine workings and was subsequently lowered, in 1972, to 2.0 mg/m$^3$ as long as the mine dust contained less than five percent quartz. Despite the current respirable-dust standard, coal miners are continually being diagnosed as developing CWP. The amounts of black-lung compensation payments continue to rise and are approaching two billion dollars annually. The compensation payments are partially funded by an excise tax on coal. Currently, the tax amounts to $1.10 per ton for coal mined underground and $0.55 per ton for surface-mined coal.

Coal extraction by a continuous mining machine (CMM) is the most common underground method in the U.S. industry today and accounts for slightly more than two-thirds of the nation's deep-mining production (Figure 1).[1] Even if longwall mining should become more commonplace, it can proceed only after ventilation and access entries have been driven by CMMs.

The Mine Safety and Health Administration (MSHA) is required to inspect all underground coal mines four times each year and collect dust samples twice each year. MSHA inspectors also sample specific occupations in a mining operation that are typically exposed to the highest respirable-dust concentrations and which create potential hazards to the individuals assigned to these worksites. Such occupations are referred to as designated occupations (DO). Examples of DO would include the continuous-miner operator and the continuous-miner-operator helper. Additionally, the MSHA inspectors sample other underground occupations suspected to have high dust exposures such as roof bolters. These are referred to as nondesignated occupations (NDO).[8]

Thus, since a CMM operator and helper have DO, and other personnel such as the section foreman and the shuttle-car operators may be exposed to dust generated by the CMM as well, the purpose of this paper will be to discuss the relationship between the elemental composition of mining-generated airborne dust sampled from the immediate ventilation return of a CMM and laboratory-generated dust derived from channel samples taken from the mines. The elemental composition of the dust in the immediate ventilation return was chosen to be compared to the laboratory-generated respirable dust because it is close to the dust generating source, which is the CMM, and samples can be safely taken from this area.

The potential contributions of this research to the coal mining industry are: 1) after more fundamental knowledge of the cause(s) of CWP is learned, in particular certain trace elements, the laboratory-generated respirable dust could be used to identify a potentially hazardous coal seam. Also, this research could possibly aid in understanding the fundamental causes of CWP by producing mining-simulated samples of coal dust which could be used in epidemiological studies,

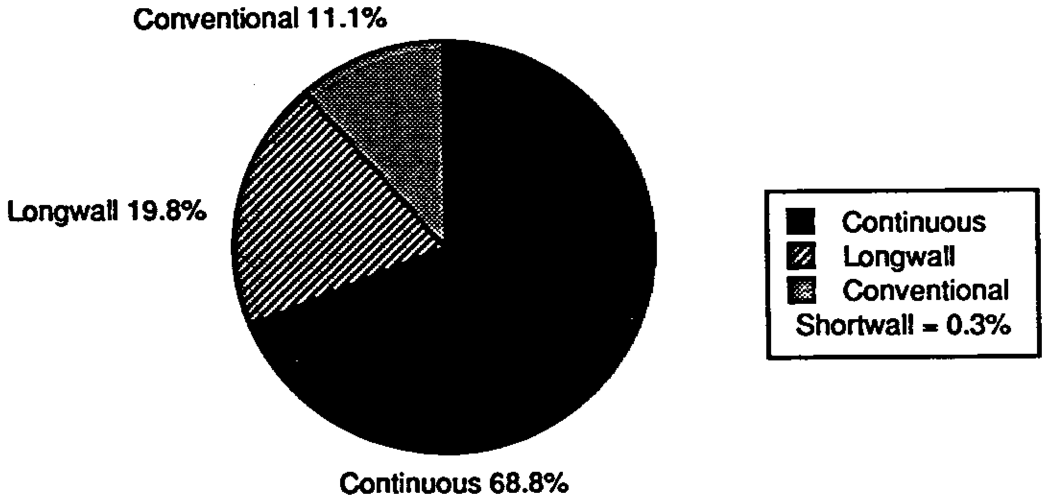

Figure 1. Underground coal production by type of mining for 1983.

and 2) assuming there is no difference in the elemental composition of a drill-core sample and a channel sample from the same location, a mining company could predict a new mine's respirable dust elemental composition in the immediate ventilation return by using exploratory drill-core samples of the roof, coal, and floor rock to prepare the laboratory dust. Ventilation engineers could then use engineering design and control measures during pre-mine planning to reduce the incidence and severity of CWP by better ventilating the potentially hazardous coal seam. If this proper planning prevented any future changes to the ventilation equipment and mine design, much time and money could be saved.

## SCOPE OF WORK

To investigate the variability of the chemical characteristics of respirable dust, airborne dust samples from eight underground coal mines located in the eastern and midwestern United States were collected with eight-stage Sierra Model 298 Marple cascade impactors marketed by Andersen Samplers, Inc. as well as twenty-five channel samples of mined material. Each channel sample was removed from the middle of the coal face before mining occurred. Sampling of the mining-generated dust was conducted by Lee.[3] He sampled the entire working sections, primarily for characterization purposes, to obtain information on the locational variability of dust characteristics. Research performed for this study used the elemental analyses of the mining-generated dusts he sampled in the immediate ventilation returns of CMMs.

The procedure that was used to produce the laboratory-generated respirable dust was based on the Hardgrove grindability test, a test which reflects the pulverizing characteristics of coal. This test was chosen for several reasons. First, it is repeatable and reproducible; a consistent amount of input energy is used as well as a specified size range of feed material to be crushed (the channel samples). Secondly, it is thought to generate secondary dust in a way similar to that of the crushing and grinding of the coal and rock as they pass through the arc-shaped cutting path of the CMM's cutter head. The potential effect on dust generation by this secondary grinding mechanism may be at least as much as that produced by primary fragmentation, which is dust produced by the cutting action of the bit against the coal or rock.[5] Finally, the Hardgrove grindability test is well known and is used in the coal industry to guide mineral-processing engineers in estimating the capacity of mills used to grind coal.

One hypothesis of dust researchers in the Generic Technology Center for Respirable Dust is that the elemental as well as the physical characteristics of coal-mine dust will make a difference in the incidence and severity of the Black-Lung disease. Coal-mine dust is generated not only from coal, but also from any rock partings contained within the seam or any roof or floor material mined with the coal. Thus, coal-mine dust may not have the same elemental characteristics as the coal being mined.

Given that hypothesis, mixtures proportional to each thickness mined of roof, coal, and floor rock derived from the channel samples of the face areas from which the respirable dusts were generated by the CMMs were used to produce the feed material which was pulverized in the Hardgrove machine. The minus 400-mesh fraction ($<37$ μm) (U.S. series standard test sieve) that was collected after pulverization was placed in a fluidized-bed aerosol generator manufactured by TSI, Incorporated and dispersed in an aerosol test chamber manufactured by Elpram Systems, Inc. While the minus 400-mesh fraction was being dispersed in the aerosol test chamber, it was also being sampled with the eight-stage cascade impactors in the same manner as the dust in the immediate ventilation return of the CMM was sampled.

The elemental composition of the mining-generated dust and the laboratory-generated dust were determined from stages 3, 5, and 7 (10 μm, 3.5 μm, and 1.0 μm aerodynamic diameter, respectively) of a cascade impactor by the Proton induced X-Ray Emission (PIXE) method by the Element Analysis Corporation of Tallahassee, Florida. To perform a PIXE analysis, a beam of protons is used to excite the atoms in the dust mass of a particular impactor stage. The X-rays emitted as a result of this excitation are analyzed to determine the elements that originated the various wave lengths. The number of X-rays that are emitted in a particular range are counted and the amount of each element in the dust can be calculated with an error that can be determined statistically for each element.

The PIXE method quantifies the mass of all elements simultaneously but had one limitation: the commercially available analysis is set up to determine only those elements that have an atomic number greater than or equal to sodium. The PIXE analysis is also a nondestructive method; therefore, any elements contained in the volatile material of the dust samples are not lost by an ashing procedure and dust samples can be archived for future use. The PIXE method easily gives a multi-element analysis from small dust masses which ranged from 5 to 120 µg.

## DESCRIPTION OF THE ANALYSIS

After receiving the elemental analyses from the outside company, the data were grouped by mine, channel-sample location, and stage according to their identification as a laboratory-generated dust or a mining-generated dust. Since each element's weight fraction of the total dust mass had an associated error plus or minus its weight fraction, a range of values occurred for an element's weight fraction. For example, if iron's weight fraction of a dust mass was 2.50E-3 (0.00250) with a ±10 percent error, then the range of iron in the dust mass was from 2.25E-3 to 2.75E-3. For a few elements in quantities near the PIXE analysis' detection limits, an error of more than 100 percent occurred. This presented no problem when adding more than 100 percent of the original weight fraction to itself, but when subtracting it, the weight fraction of the particular element was entered as zero, since a negative quantity of an element does not physically exist.

After a range of values for a particular element was calculated, all weight-fraction ranges of the mining-generated and the laboratory-generated dust from the same mine and particular impactor stage were sorted for a minimum and a maximum weight fraction value. Once these two values were identified, their average value could be calculated and column graphs drawn. The height of the column represented the average value, and the difference between the maximum value and the average value represented the error bar value. See Figures 2 through 5 for examples of some of the elemental values.

In those cases where the weight fraction of the laboratory-generated dust fell short of or exceeded the range of the mining-generated dust, or did not appear at all as in the cases for Na, Sb, and Ba, they were considered unsuccessful predictors. Thus, for the 32 elements, the laboratory-generated dust was considered a successful predictor 73% of the time for stage 3, 65% of the time for stage 5, and 57% of the time for stage 7. This resulted in an overall predictability of 65% (see Table I).

## CONCLUSIONS AND RECOMMENDATIONS

1. By observing Figure 2, the carbon or the organic fraction in the coal dust appears to increase with decreasing dust size.
2. Although different researchers have differing opinions on the elements which they believe have an impact in contracting CWP, evidence indicates that elements such as Pb, Ni, and Zn are contained in greater amounts in bituminous-coal-miner's lungs than normal concentrations of these elements.[6,7] As such, the standard procedure developed to produce a laboratory-generated dust appears to predict well the concentrations of Pb, Ni, and Zn (with 100%, 85%, and 81% accuracy, respectively) in the immediate ventilation return of a CMM (see Figures 3, 4, and 5). Thus, potential problem mines or coal seams may be identified during planning stages.
3. Ba, Sb, Cd, and Na were the most difficult elements for the laboratory-generated dust to produce in detectible amounts. They were detected in the mining-generated dust in mines where roof and floor rock were mined with the coal seam. Poor detection of these elements in the laboratory-generated dust may be due to inadequate grinding of the rock component during sample pulverization. It is recommended that a refined procedure of the one used in this research be developed to better predict the mining-generated dust when rock is concurrently mined with the coal seam.

An important contribution of this research was that it developed and described a standard procedure, or a tool, which has shown promise for measuring the variability of the elemental composition of coal mine dust in the immediate ventilation return of a CMM through the use of a laboratory-based process.[2]

To make the successful technology transfer in which a coal mining company is able to predict a proposed mine's airborne dust elemental characteristics in the immediate ventilation return of a CMM by using the standard procedure presented in this paper, it is recommended that core samples of a coal property be used to prepare the laboratory-generated dust which would be compared to the mining-generated dust sampled as close as possible to the core location after mine development. This would allow verification of the assumption that a core sample could successfully be used in place of a channel sample to produce laboratory-generated dust that has elemental characteristics similar to those of dust sampled in the immediate ventilation return of a CMM.

Finally, it is recommended that researchers investigating the significance of elements and chemical variations on cell cultures and live animals use a laboratory-generated dust which is similar in composition to actual mine dust to perform their studies. This would better represent the dust that miners actually breathe.

As medical investigations continue to find the cause(s) of CWP, and if a portion of the cause(s) is found to be certain elements or particular concentrations of those elements in the mine dust, then the successful application of this research will contribute to the reduction in incidence and severity of CWP and reduce the cost of attaining that goal.

## Table I
## Lab Dust in the Same Concentration Range as Mine Dust

| Element | Stage 3 | Stage 5 | Stage 7 | Totals |
|---|---|---|---|---|
| C | hypothetical | hypothetical | hypothetical | - |
| O | hypotthetical | hypothetical | hypothetical | - |
| Na | 0/3 = 0% | 0/2 = 0% | 0/1 = 0% | 0/6 = 0% |
| Mg | 6/8 = 75% | 5/9 = 56% | 1/8 = 13% | 12/25 = 48% |
| Al | 5/9 = 56% | 5/9 = 56% | 5/9 = 56% | 15/27 = 56% |
| Si | 6/9 = 67% | 4/9 = 44% | 3/9 = 33% | 13/27 = 48% |
| P | 9/9 = 100% | 8/9 = 89% | 7/9 = 78% | 24/27 = 89% |
| S | 6/9 = 67% | 7/9 = 78% | 6/9 = 67% | 19/27 = 70% |
| Cl | 8/9 = 89% | 5/9 = 56% | 7/9 = 78% | 20/27 = 74% |
| K | 4/9 = 44% | 3/9 = 33% | 1/9 = 11% | 8/27 = 30% |
| Ca | 4/9 = 44% | 3/9 = 33% | 0/9 = 0% | 7/27 = 26% |
| Ti | 6/9 = 67% | 5/9 = 56% | 6/9 = 67% | 17/27 = 63% |
| V | 4/6 = 67% | 4/7 = 57% | 0/6 = 0% | 8/19 = 42% |
| Cr | 2/7 = 29% | 5/8 = 63% | 1/8 = 13% | 8/23 = 35% |
| Mn | 6/9 = 67% | 5/9 = 56% | 4/9 = 44% | 15/27 = 56% |
| Fe | 5/9 = 56% | 3/9 = 33% | 3/9 = 33% | 11/27 = 41% |
| Ni | 7/9 = 78% | 7/9 = 78% | 9/9 = 100% | 23/27 = 85% |
| Cu | data suspect | data suspect | data suspect | - |
| Zn | 8/9 = 89% | 8/9 = 89% | 6/9 = 67% | 22/27 = 81% |
| Ga | 9/9 = 100% | 9/9 = 100% | 8/8 = 100% | 26/26 = 100% |
| Ge | 7/7 = 100% | 6/6 = 100% | 5/6 = 83% | 18/19 = 95% |
| As | 6/6 = 100% | 4/4 = 100% | 5/5 = 100% | 15/15 = 100% |
| Se | 6/6 = 100% | 4/5 = 80% | 5/6 = 83% | 15/17 = 88% |
| Br | 6/9 = 67% | 5/9 = 56% | 8/9 = 89% | 19/27 = 70% |
| Rb | 8/9 = 89% | 5/8 = 63% | 6/9 = 67% | 19/26 = 73% |
| Sr | 9/9 = 100% | 7/9 = 78% | 6/9 = 67% | 22/27 = 81% |
| Zr | 9/9 = 100% | 7/8 = 88% | 7/8 = 88% | 23/25 = 92% |
| Mo | 7/7 = 100% | 7/8 = 88% | 3/6 = 50% | 17/21 = 81% |
| Cd | 1/6 = 17% | 0/2 = 0% | 0/1 = 0% | 1/9 = 11% |
| Sb | 0/3 = 0% | 0/3 = 0% | 0/3 = 0% | 0/9 = 0% |
| Ba | 0/1 = 0% | 0/1 = 0% | 0/1 = 0% | 0/3 = 0% |
| Pb | 9/9 = 100% | 9/9 = 100% | 9/9 = 100% | 27/27 = 100% |
| Totals | 163/222 = 73% | 140/215 = 65% | 121/211 = 57% | 424/648 = 65% |

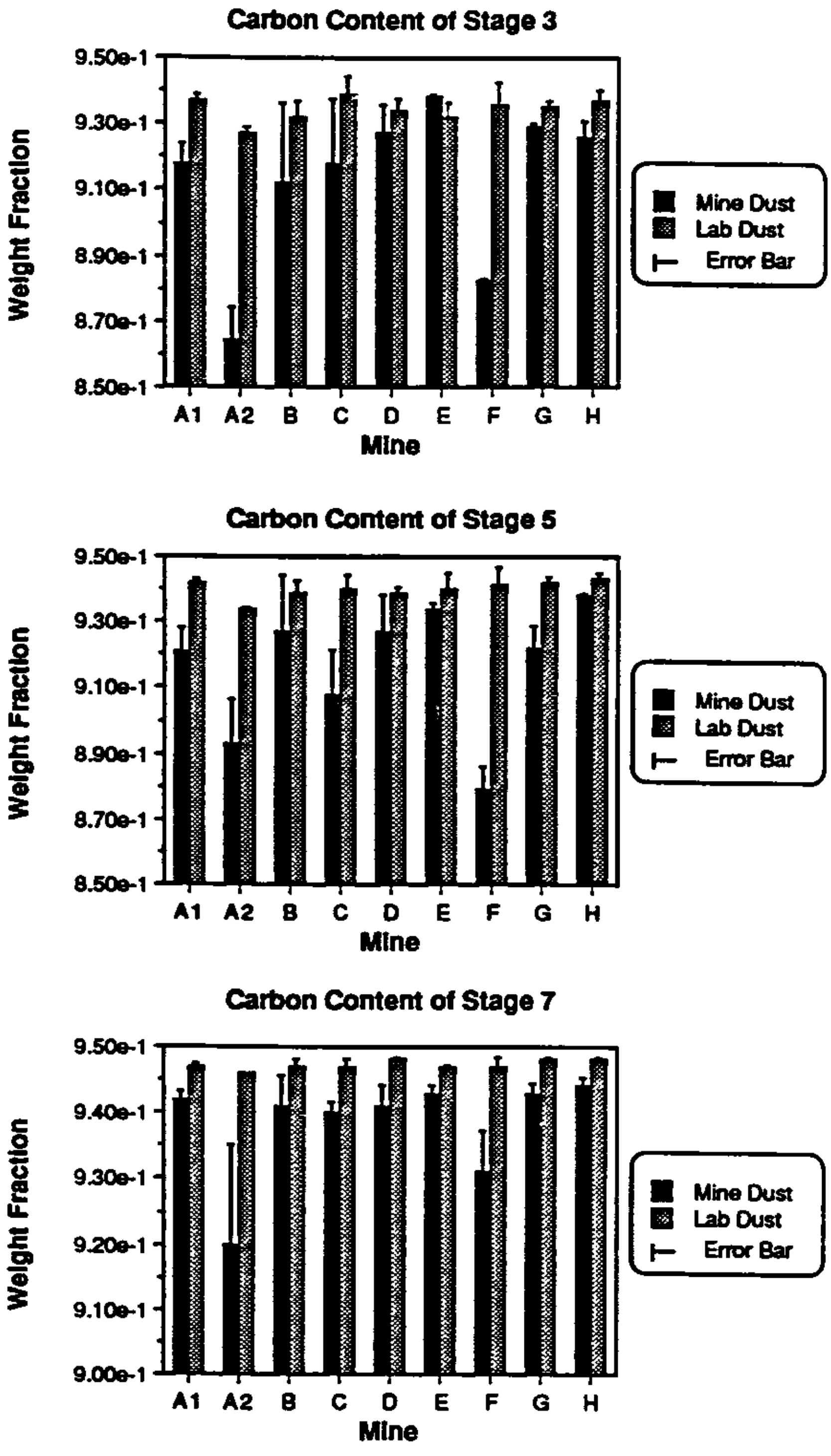

Figure 2. Carbon content by stage and mine.

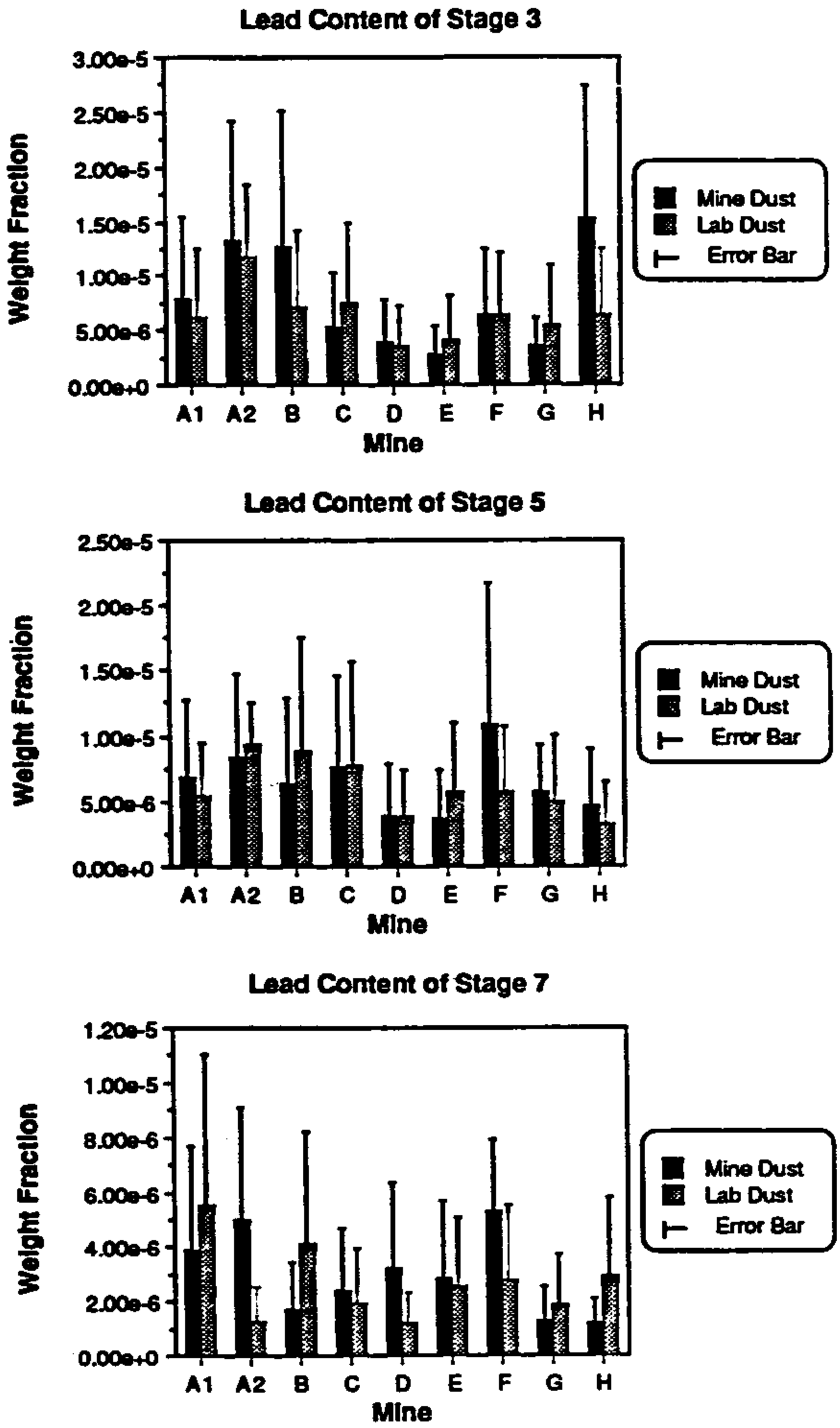

Figure 3. Lead content by stage and mine.

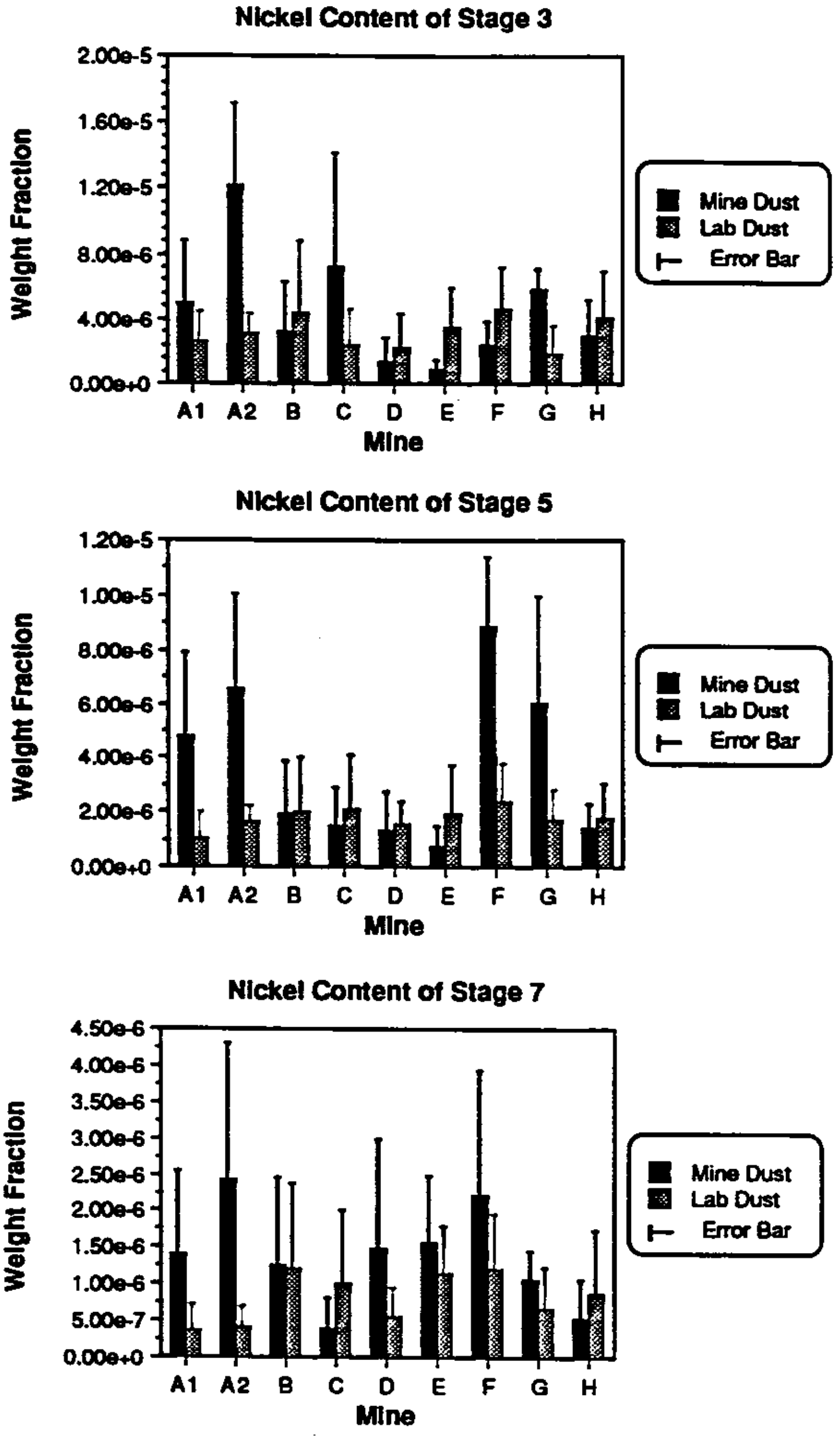

Figure 4. Nickel content by stage and mine.

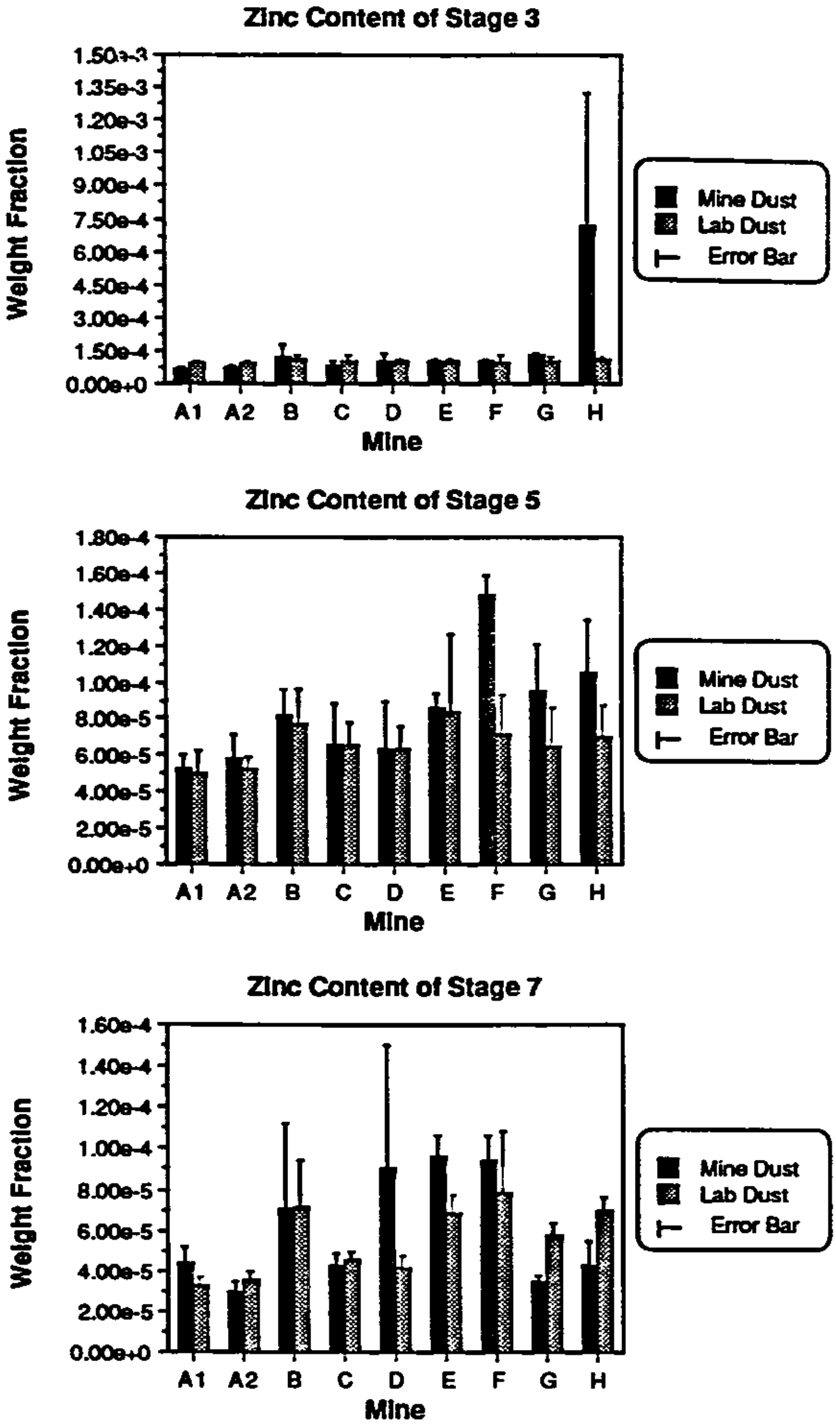

Figure 5. Zinc content by stage and mine.

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