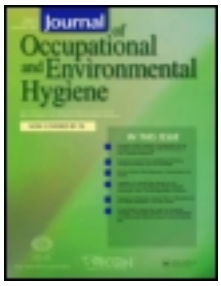
[Appendix 2]



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The Relationship between Various Exposure Metrics for Elongate Mineral Particles (EMP) in the Taconite Mining and Processing Industry

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The Relationship between Various Exposure Metrics for Elongate Mineral Particles (EMP) in the Taconite Mining and Processing Industry

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ABSTRACT

Different dimensions of elongate mineral particles (EMP) have been proposed as being relevant to respiratory health end-points such as mesothelioma and lung cancer. In this article, a methodology for converting personal EMP exposures measured using the NIOSH 7400/7402 methods to exposures based on other size-based definitions has been proposed and illustrated. Area monitoring for EMP in the taconite mines in Minnesota's Mesabi Iron Range was conducted using a Micro Orifice Uniform Deposit Impactor (MOUDI) size-fractionating sampler. EMP on stages of the MOUDI were counted and sized according to each EMP definition using an indirect-transfer transmission electron microscopy (ISO Method 13794). EMP were identified using energy-dispersive x-ray and electron diffraction analysis. Conversion factors between the

EMP counts based on different definitions were estimated using (1) a linear regression model across all locations and (2) a location-specific ratio of the count based on each EMP definition to the NIOSH 7400/7402 count. The highest fractions of EMP concentrations were found for EMP that were 1–3 µm in length and 0.2 – 0.5 µm in width. Therefore, the current standard NIOSH method 7400, which only counts EMP > 5 µm in length and ≥ 3 in aspect ratio, may underestimate amphibole EMP exposures. At the same time, there was a high degree of correlation between the exposures estimated according to the different size-based metrics. Therefore, the various dimensional definitions probably do not result in different dose-response relationships in epidemiological analyses. Given the high degree of correlation between the various metrics, a result consistent with prior research, a more reasonable metric might be the measurement of all EMP irrespective of size.

INTRODUCTION

A number of studies have been published on the relationship between exposure to asbestiform "fibers" and health effects such as lung cancer and mesothelioma. Since the term "fiber" has been controversial in the context of asbestos [(1)], the National Institute for Occupational Safety and Health (NIOSH) has recently proposed the use of the term "elongate mineral particles" or EMP to refer to mineral particles with a minimum aspect ratio of 3:1 that are of inhalable, thoracic, or respirable size [(2)].

The current regulations for asbestiform EMP are based on length (≥ 5 µm) and aspect ratio (> 3:1) measured using the NIOSH Method 7400, a counting protocol that has been criticized as lacking

a scientific basis [3, 4]. EMP dimensions are important because: (1) the different sizes of EMP penetrate to and deposit in different regions of the lung, (2) the macrophages cannot remove EMP from the lung when they are longer than the macrophage diameter, and (3) the lung cannot function properly when thinner EMP deposit in the alveolar region of the lung [5]. Other EMP characteristics related to toxic health effects include the morphological habit, chemical composition, and activity [5, 6, 7].

Minnesota counties in the vicinity of taconite mining operations have been found to have elevated age-adjusted rates for mesothelioma [8], a disease thought to be associated with exposure to asbestiform EMP. Studies measuring EMP dimensions have been relatively scarce [9]. However, due to the characteristics of the ore body, non-asbestiform EMP are a potentially major source of exposure and therefore, adverse health effects may be linked also to non-asbestiform EMP. To date, no study has conducted an extensive assessment of the relationship between non-asbestiform EMP and adverse health effects in taconite mining industry. In general, non-asbestiform cleavage fragments have not been thought to have high potential for disease [7, 10, 11].

In the taconite mining industry, cleavage fragments refer to the fractured mineral EMP created during the crushing and fracturing process [2]. Because no standard definition exists, distinguishing cleavage fragments from asbestiform EMP is challenging [12]. Even if a given EMP counting criterion is met, standard methods such as phase contrast microscopy (NIOSH method 7400) [13] or transmission electron microscopy (NIOSH method 7402) [14] cannot distinguish between non-asbestiform cleavage fragments and asbestiform EMP. Researchers

have found that non-asbestiform cleavage fragments are inactive in *in vitro* bioassays and that they have less strength and flexibility in morphologic analyses [6, 7]. A linear relationship has been found to exist between the width and length of cleavage fragments, while no such relation exists in asbestiform EMP [15]. The term "amphibole EMP" refers to a subset of double chain silicate minerals that can be asbestiform or non-asbestiform [2, 15]. Cleavage fragments are likely thicker than asbestiform EMP, while asbestiform EMP are likely to be longer and more flexible [4].

Because of the difficulty in distinguishing between asbestiform and non-asbestiform EMP, the relationship between the size of non-asbestiform EMP and carcinogenic lung disease is still not well understood [2]. Although the chemical composition of asbestiform and non-asbestiform EMP can be the same, they differ in their habit or morphology [16, 17]. Asbestiform EMP are "polyfilamentous" whereas non-asbestiform EMP display a "multidirectional" pattern [15].

No consensus exists regarding the most health-relevant, dimension-based exposure metric for EMP. Stanton et al. [18] ascribed carcinogenicity to EMP with a length greater than 8 µm and a diameter less than 0.25 µm. Berman et al. [19] suggested that asbestos EMP greater than 5 µm in length contributed to lung tumor risk, while those less than 5 µm did not contribute to the risk. A panel of experts convened by the Agency for Toxic Substances and Disease Registry [1] concluded that asbestos and synthetic vitreous fibers shorter than 5 µm are unlikely to cause cancer in humans. Chatfield [20] proposed a protocol that defined asbestiform EMP as those with widths between 0.04 µm and 1.5 µm in width and aspect ratio between 20 and 1000; EMP that

did not fall in these ranges are considered non-asbestiform EMP including cleavage fragments. The Occupational Safety and Health Administration [21] also defined the cleavage fragments as those with aspect ratio less than 20.

Other researchers have argued against ruling out the effect of short fibers. Suzuki et al. [22] concluded that shorter (≤ 5 μm) and thinner EMP (≤ 0.25 μm) were more strongly associated with malignant mesothelioma through analysis of lung and mesothelial tissues in human patients. Dement et al. [23] showed that exposures to EMPS with all combinations of dimensions (lengths ranging from < 1.5 μm to > 40 μm and widths ranging from < 0.25 μm to > 3.0 μm) were highly associated with lung cancer and asbestosis. This result led them to conclude that the traditional method, which only counts EMP longer than 5 μm, may be deficient. In fact, shorter EMP also contribute to health-relevant work exposures, a contribution that may be important [24].

Table 1 summarizes the dimension-based EMP definitions that will be used in this paper. In Figure 1, the same four size-based EMP definitions are compared using a typical sample collected for this study. Each graph shows the same particle counts from five stages of a Micro Orifice Uniform Deposit Impactor (MOUDI) cascade impactor (Model 125R MOUDI-II, MSP Co., Shoreview, MN), overlaid by a polygon that indicates one of the size-based definitions. There are no overlapping areas between the NIOSH and Suzuki et al. definitions or the Chatfield EMP and Cleavage Fragment definitions. Typically, few EMP were identified by the NIOSH and Chatfield definitions, while many were identified by the Suzuki and Cleavage fragment

definitions. A lack of consensus on the appropriate exposure metric can partially explain the different exposure-response relationships obtained in an epidemiological study [25].

The aims of this study were to: 1) determine the size distribution by length and width of EMP as measured using the MOUDI in different representative locations in each of the six taconite mines currently operating in the Mesabi Iron Range; 2) develop a methodology to determine the relationships between the standard NIOSH Methods 7400/7402-based EMP exposure metric to the other dimension-based EMP exposure metrics.

The current research was carried out as part of an epidemiological study investigating the relationship between exposures to EMP during the mining and processing of taconite ore and respiratory diseases.

METHODS

Sampling sites

The mineralogy of the Mesabi Iron Range in northeastern Minnesota changes from east to west with distinct metamorphic mineralogical zones. In the eastern zone, the iron ore contains amphibole, whereas the ore in the western zone contains predominantly phyllosilicates that are not regulated as asbestiform or amphibole EMP [26, 27].

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Based on the mineralogical characteristics of the two zones, taconite mining may potentially lead to exposures to different types of EMP. Our exposure assessment strategy attempted to capture this difference. Currently, one mine operates in the eastern zone (mine A) and five mines in the western zone (mines B, C, D, E, and F). The first criterion for the sampling design was to determine locations for area sampling representing the eastern and western zones that generally corresponded to the similar exposure groups (SEGs), the basis for personal sampling [(28)]. The term location is used to refer to physical places where the area measurements were obtained for each SEG. Table 2 lists the number of area MOUDI samples taken and the number of locations sampled by mine.

Sampling design

Area samples, taken during normal operating conditions at locations representative of each SEG, were collected in up to two samples per location. The samples were obtained using a MOUDI impactor. The cut sizes of the 13 impactor stages ranged from 0.010 µm to 10 µm. Based on observations of stage loading and to be able to assess a broad range of aerodynamic particle size intervals within a budget that limited the number of samples, we chose stages 3, 5, 7, 9 and 11 – corresponding to size intervals of 3.2-5.6 µm, 1.0-1.8 µm, 0.32-0.56 µm, 0.10-0.18 µm, and 0.032-0.056 µm, respectively – for further microscopic analysis. The inlet flow rate from the attached vacuum pump (Model R 5, Busch USA, Virginia Beach, VA, USA) was ~10 L/min and the duration of each sample was 4 hours. The impaction plate used a hydrophilic polycarbonate membrane filter (Isopore Co., Billerica, MA, USA) suitable for analyzing the chrysotile,

amphibole, and non-amphibole EMP on each stage. The after-filter for the impactor used polytetrafluoroethylene (PTFE) filters with laminated PTFE supports (SKC Inc., Eighty Four, PA, USA).

Analytical methods

The ISO 13794 method, adopted to analyze the impactor samples, provides details of EMP dimension, structure type, and mineral type for each EMP regardless of the EMP dimension [(29)]. ISO 13794 immerses the whole filter in water and re-filters the particles suspended in the water through a secondary filter. Thus, the particles are indirectly but evenly distributed across the surface of the secondary filter. Eypert_Blaison et al. [(33)] compared previous studies in which the direct versus the indirect methods of measuring EMP were used. They found that the indirect method resulted in higher measurements of EMP than the direct, suggesting several reasons for the difference. With the indirect method, large structures can be separated by calcinations, ultrasonic dispersion, and re-dispersion procedures during TEM preparation [(32)]. Conversely, with the direct method, organic debris can hide EMP because the filters are not ashed [(34)]. Eypert_Blaison et al. [(33)] also compared studies in which the direct versus the indirect methods of measuring chrysotile were used, finding that the direct method resulted in higher measurements than the polycarbonate indirect method although the majority of EMP in this study were amphibole. They pointed out that EMP can be lost during evaporation using indirect method, while the EMP density for an overloaded filter is underestimated using the direct method.

However, they did not identify any differences between the two methods due to EMP size distribution.

The resolution limit of ISO 13794 is 0.3 µm in length and 0.1 µm in width, and we counted all fibers > 0.3 µm in length with an aspect ratio > 3. Therefore, our data have both counts and sizes of each EMP for each of the MOUDI stages analyzed in each location. All analyses were carried out at an American Industrial Hygiene Association-accredited laboratory (EMSL Analytical Inc., Minneapolis, MN, USA).

ISO 13794 classifies EMP into three distinct categories: chrysotile, amphibole, and non-amphibole EMP. The non-amphibole EMP do not include the chrysotile EMP. The amphibole EMP were further classified into five types: amosite/cummingtonite-grunerite, crocidolite/riebeckite, tremolite asbestiform/tremolite, anthophyllite asbestiform/ anthophyllite, and actinolite asbestiform/actinolite. The transmission electron microscopy (TEM) method for identifying each EMP using ISO 13794 was the same as that used for NIOSH Method 7402 [(14)], based on the diffraction pattern and chemical spectrum for each EMP. Each type of amphibole EMP has a certain ratio of Na, Mg, Si, Ca, and Fe. EMP that did not fit in either the chrysotile or amphibole category were classified as non-amphibole.

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Data management

The impactor data were used to determine the distribution of EMP sizes and derive the relationships between various dimension-based EMP exposure metrics. The assessment of cumulative exposure according to these metrics will enable epidemiological testing of various hypotheses regarding the health-relevance of different sizes of EMP.

Since only five chrysotile EMP (also known as a common commercial asbestiform EMP) were found from a total of 2931 identified EMP, we excluded these particles from further analysis. While EMP analysis using TEM can distinguish between amphibole and non-amphibole EMP, it cannot distinguish between asbestiform and non-asbestiform EMP. Therefore, in this paper, "total EMP" refers to both amphibole and non-amphibole/non-chrysotile EMP and "amphibole EMP" refers to amosite/cummingtonite-grunerite and actinolite asbestiform/actinolite EMP (which were the only types of amphiboles found in our samples).

For each sample, grid openings were analyzed for EMP until either the 100th particle was counted or the required analytical sensitivity was achieved, whichever occurred first [(29)]. The EMP count was normalized by the number of grid openings analyzed in each substrate for each location by zone. If more than one sample was obtained at a location in a mine, we tallied the EMP for all samples and then divided by the corresponding number of samples to obtain the average EMP count for that location. If a sampled location was representative of more than one SEG, we assigned the data from the sampled location to all the relevant SEGs.

Because we analyzed only selected stages, the non-analyzed stages were estimated as an arithmetic average of the two adjacent stages. As we show later, this interpolation was justifiable because the counts of EMP on alternate MOUDI stages were not significantly different. Only the data from stages 3 to 11 were considered because EMP were not analyzed on stages 1, 2, 12, and 13. Size-integrated EMP counts for an impactor sample were the sum of the counts for all stages between 3 and 11. Total (or amphibole) EMP were counted for each stage using dimension-based metrics. We converted the normalized EMP count from number of particles to concentration (particles/cm^3) by dividing the number of EMP per sample by the product of the grid opening area and the number of grid openings observed, multiplying by the effective area of the secondary filter, and then dividing by the sampled air volume.

Statistical analyses

All analyses reported here were conducted using SAS version 9.3 (SAS Institute, Cary, NC, USA). Statistical significance was defined by levels of 0.05 or lower.

A two-way ANOVA was used to examine whether concentration differences between stages of the MOUDI are significant for each location. We started with two main effects (impactor stage and location) and then included the interaction of impactor stage and location to see if the interaction term was significant. In addition, Tukey's studentized range was used for pair-wise comparisons of the log of the number concentration of EMP collected on each stage.

The associations between NIOSH 7400 and various total EMP concentration metrics such as Suzuki, Chatfield, and Cleavage Fragments were assessed (a) using a simple linear regression based on the log-transformed exposure concentrations across all locations in the eastern zone (Equation 1) and (b) the ratio of the log-transformed exposure concentrations according to each alternative metric and the NIOSH 7400 metric across all locations in the western zone (Equation 2). Since no amphibole EMP were counted by the NIOSH metrics in the western zone, the regression approach characterizes an overall conversion factor for total EMP across locations in each zone:

$C_{Definition}=a_1 (C_{NIOSH\ EMP})^b$ for total and amphibole EMP in the eastern zone

(Equation 1)

$C_{Definition}=a_2 (C_{NIOSH\ EMP})$ for total EMP in the western zone

(Equation 2)

where $C_{Definition}$ = concentration of total EMP that meet a specific size definition, $C_{NIOSH\ EMP}$ = concentration of total EMP that meet the NIOSH 7400 definition, a_1= intercept based on linear regression between the log-transformed concentrations $C_{Definition}$ and $C_{NIOSH\ EMP}$, b = slope based on the linear regression, and a_2 = ratio of concentration of each size EMP definition to NIOSH 7400 definition based on linear regression.

The second approach characterizes the ratio of each size-based EMP definition to NIOSH 7400 and 7402 by location (Equation 3). In this way there is a separate conversion factor for each location for both total and amphibole EMP in both zones.

$C_{Definition} = a_i C_{NIOSH\ EMP}$ for the ith location

(Equation 3)

RESULTS

To assess the relationship between the EMP size distribution and stages, we performed pair-wise comparisons of the counts on the various stages. Ten stage comparisons (combinations of stages 3, 5, 7, 9, and 11) were carried out for both the total and amphibole EMP concentration. Only two pair-wise stage comparisons (stages 5 vs. 9, and 5 vs. 11) out of 10 in the eastern zone and one pair-wise stage (stages 3 vs. 11) out of 10 in the western zone were significantly different at $\alpha = 0.05$. The interaction of location and stage was not a significant variable in either geologic zone (p-value: 0.9980 in the east, 0.3967 in the west). When the model was run without the interaction term, the two main effects (stage and location) were significant variables in both zones.

Considerable differences between the two geological zones were found for total and amphibole EMP. Figure 2 shows the combined EMP concentration of stages 3 through 11 by location in the eastern zone, with a reference line indicating the NIOSH recommended exposure limit (REL) of

0.1 particles/cm^3 for EMP. The reference line is shown only as a benchmark, and not to show compliance as the MOUDI measurements are area samples obtained over 4 hours. However, the total EMP classification does not necessarily refer to regulated asbestiform EMP, because the NIOSH 7400 cannot differentiate between asbestiform and non-asbestiform EMP and also refers to a very specific range of lengths and widths. We only present EMP results in the eastern zone because the concentrations of EMP in the eastern zone are markedly higher than that in the western zone. The highest exposure location per department was selected in Figures 2 – 3 (all locations are available in the online supplement). Pair-wise comparisons using a t-test indicated that average total and amphibole EMP concentrations at similar locations were significantly different between the two zones (p-value < 0.0001).

When a location had a high concentration of total EMP, there was also a high concentration of amphibole EMP (e.g., location corresponding to *Railroad* SEG). In the eastern zone, the highest concentration for total and amphibole EMP was found in the location corresponding to *Operating technician* SEG, which was at least 2.4 times higher than the second highest concentration found in the *Railroad* location. In the western zone, the location corresponding to the *Boiler technician* SEG was the only location in which the concentration of total EMP exceeded the NIOSH recommended exposure limit (REL) of 0.1 EMP/cm^3, while none of the locations had amphibole EMP concentrations exceeding the REL.

The total and amphibole EMP concentrations for the four different size-based definitions in the eastern zone are shown in Figure 3. Again, the reference line indicates the REL for EMP and the

y-axis scale for amphibole EMP is different than for total EMP. Much lower concentrations were observed for the NIOSH and Chatfield definitions than the Suzuki, in which shorter EMP are counted. The highest concentrations for total and amphibole EMP were observed for the Cleavage Fragment definition. Again, the location corresponding to *Operating technician* SEG had the highest EMP concentration for all size-based definitions. In the western zone, we observed substantially lower concentrations of both total and amphibole EMP for all definitions. For total EMP, the concentration levels for most of the locations in the eastern zone were above the REL, while all but one location in the western zone had concentration levels below the REL. Figure 3 indicates that the relative magnitudes of the concentrations of total and amphibole EMP at selected locations were similar using the different exposure definitions. It is emphasized that the EMP concentrations in this paper are not personal exposure samples; therefore, the values do not necessarily mean that workers are exposed above the REL.

Figure 4 provides the coefficients of determination (R^2) between various dimension-based EMP definitions for total and amphibole EMP in the eastern zone for log-transformed concentration data. High R^2 were found for total and amphibole EMP (R^2 ranges: 0.90 - 0.98 and 0.84 - 0.99, respectively), consistent with the pattern of concentrations across locations according to the different EMP definitions in the eastern zone displayed in Figure 3. The slightly lower correlation among definitions for amphibole EMP might be taken as an indication that the concentrations based on these definitions are more independent, the low amphibole EMP concentration in the western zone support such an interpretation. The coefficients of determination for total EMP concentration in the western zone with log-transformed

concentration data are shown in Figure 5. Except for one coefficient of determination between the Suzuki and Cleavage Fragment definitions ($R^2 = 0.88$), the coefficients of determination for total EMP in the western zone were low ($R^2 = 0.14 - 0.58$).

A regression model (Equation 1) was derived to relate EMP log-concentrations based on NIOSH 7400 to the log-concentrations based on each of the other size-based EMP definitions for total and amphibole EMP (Table 3). This regression equation is applicable across all locations for total EMP in the eastern zone. However, the regression coefficients for both the intercept and slope were not significant for total EMP in the western zone. Therefore, we used a different regression model (Equation 2) to determine a ratio of concentration based on each size-based definition to NIOSH 7400 EMP concentration across all locations in the western zone.
The regression coefficients for both total and amphibole EMP in the eastern zone were calculated for uncertainty estimates. The 95% confidence intervals are only shown for the slope. The largest range for the slope estimates was found for the amphibole Chatfield EMP (95% CI= 0.296, 1.707). The concentrations of amphibole EMP in the western zone identified by NIOSH 7400 were zero, except at the location corresponding to the *Concentrator operator* SEG (0.0002 particles/cm^3). Therefore, no regression parameters were estimated for amphibole EMP in the western zone.

Table 4 shows the ratios of EMP concentrations based on the Suzuki, Chatfield, and Cleavage fragment definitions to EMP concentration based on the NIOSH 7400 and 7402 definitions, by location for total and amphibole EMP in both zones. Many of these ratios for amphibole EMP

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are not available in the western zone because amphibole EMP were not observed in the MOUDI measurements in this zone. For both total and amphibole EMP, the ratios for the Cleavage Fragment definition generally have the largest values, followed by those for the Suzuki and Chatfield definitions. Also, the ratios for total EMP tend to be larger than those for amphibole EMP.

Figure 6 presents the size distribution by length and width of total EMP for six representative locations (one in each department except office/control room) in the eastern zone. For each location, the fraction of the total EMP in each of 25 categories (5 lengths x 5 widths) was calculated. The sum of the fractions of amosite/cummingtonite-grunerite (black) and non-amphibole/non-chrysotile EMP (gray) over all the categories is equal to one. Interestingly, the size distribution category 1- 3 µm in length and 0.2 - 0.5 µm in width contained the highest fraction of total EMP for all locations in the eastern zone.

DISCUSSION

This study is the first comprehensive assessment of the size distributions of EMP in the six currently operating mines in the Mesabi Iron Range. In addition, this is the first attempt to understand the relationships between exposures based on different EMP exposure metrics.

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Comparison between total and amphibole EMP by zones

The concentrations of both total and amphibole EMP were much higher in the eastern zone than in the western, which is consistent with the geological differences between the zones. Higher amphibole EMP concentrations are found in the mining processes than in the shop areas. Both total and amphibole Cleavage Fragment concentrations are clearly higher in the mining processes, consistent with the generation of cleavage fragments in the mining and processing of ore. Some of these EMP may conform to the regulatory fiber definition of length greater than 5 µm and aspect ratio of at least 3:1 even if they are not asbestiform in habit.

Comparison between various dimension-based EMP exposure metrics

We found that relative magnitudes of concentrations across locations were similar for the different dimension-based EMP definitions in the eastern zone, a similarity that was more obvious in the mining processes (e.g., mining, crushing, concentration, and pelletizing) than in the shop area. Even though the measured levels of total and amphibole EMP concentrations using each size-based definition were different, the metrics themselves were highly correlated in our study. The high correlation among these EMP concentration metrics will limit the ability of epidemiological analyses in determining relative differences in health effect due to exposure to different EMP metrics. In other words, the effects of correlated relationships are not identifiable. Quinn et al. [25] showed that the ranges of the R^2 for different size-based EMP definitions were 0.02-0.89. They explained the impact of relationships among the alternative definitions in epidemiologic analysis. If one EMP definition is more closely related to the health effects, there

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would be a loss of power using another EMP definition. Dement et al. [23] showed that exposures to various combinations of EMP dimensions (lengths ranging from < 1.5 µm to > 40 µm and widths ranging from < 0.25 µm to > 3.0 µm) were all highly associated with lung cancer and asbestosis. This could be because of correlation between the exposures based on various length and width combinations in that study. Stayner et al. [30] also conceded that the main limitation of their study about the fiber dimension in an asbestos textile plant was the high degree of correlation between size-specific cumulative exposure measures.

Relationship between NIOSH and other definitions

We assessed both the concentration and size distribution of EMP using different size-based definitions to understand the relationships among these exposure metrics. The various size-based definitions resulted in different EMP counts, implying that the specific definition used had a significant effect on the EMP exposure levels. The relationships developed from this analysis will be used to convert historical personal exposure data measured by NIOSH 7400 to the alternative exposure metrics for use in epidemiological analyses.

Quinn et al. [25] and Dement et al. [23] presented an “adjustment factor” or a simple ratio of PCM to TEM for the EMP dimension categories. In our study, the relationship between each EMP dimension index and the NIOSH 7400 definition was created by 1) a location-specific ratio, and 2) a regression model based on data across all locations. The regression equation led to ratios that were in the same range as the location-specific ratios.

The ratios varied by zone. The high ratios and wide range for total EMP using the Cleavage Fragment definition in the western zone indicate that cleavage fragments are a greater percentage of what is in the western zone. On the other hand, ratios for amphibole EMP detected using the Chatfield definition in the eastern zone had a narrow range, indicating that relatively few Chatfield EMP in this zone. We also found that the short EMP metrics (Suzuki and Cleavage fragments) were more likely to have high ratios. Thus, exposures based on these metrics are likely to be greater than those measured by the standard analytical methods that do not count short EMP.

Taconite EMP size distribution

The highest count fractions of EMP concentration were found for particles with 1 - 3 µm in length and 0.2 - 0.5 µm across locations. This refers to short EMP and is a subset of the Suzuki and Cleavage Fragment definitions. Dodson et al. [31] concluded that asbestos EMP of all lengths induce pathological responses and cautioned against ignoring EMP shorter than 5 µm in length, as they constitute most of the contributions of EMP to exposure. In our measurements, most of the EMP are shorter than 5 µm in length and 1 µm in width. The NIOSH methods only count EMP greater than 5 µm in length; thus, shorter EMP are not counted. No significant differences existed in the EMP length and width distributions across locations.

Limitations of this study

This study was conducted using area measurements with limited number of samples for each location in each mine to understand the relationship between the various exposure metrics that can then be applied to personal exposure measurements based on NIOSH 7400. Ideally, each personal sample would have been analyzed using ISO 13794 to obtain EMP counts according to each exposure metric. However, this approach was not feasible due to budgetary constraints. Furthermore, the area samples from stages 1, 2, 12, 13 were not analyzed, and the potential impact of this might underestimate the EMP exposures. However, the area measurements provided useful insights, chief among them that the exposures based on various metrics were significantly correlated with each other. Since asbestiform and non-asbestiform amphibole EMP are chemically identical, our data based on TEM analysis cannot distinguish between them. This method includes expanded characterization of elemental composition with energy dispersive X-ray analysis and crystalline structure by selected area electron diffraction. While laboratories typically claim to distinguish between asbestiform and non-asbestiform EMP using TEM, a more conservative assessment is that this method can identify amphibole versus non-amphibole EMP (in addition to chrysotile EMP). For instance, although the contracted laboratory used the terms "amosite" and "actinolite", common in asbestos terminology, "amosite" can mean either amosite (asbestiform) or cummingtonite-grunerite (non-asbestiform). The Chatfield definition can classify each amphibole EMP into asbestiform and non-asbestiform categories using TEM. It is important to note, however, that asbestiform amphibole EMP can typically be identified using scanning electron microscopy. However, these methods have not been extensively validated.

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While we expect that most of amphibole EMP are likely non-asbestiform in the Iron Range, this is based on past studies [9, 27].

CONCLUSIONS

Many size-based definitions have been proposed for assessing concentrations of EMP. We chose four different EMP dimensional definitions, including the NIOSH standard method to understand the relationships between these metrics. These four have been proposed as being relevant to respiratory diseases such as mesothelioma and lung cancer. Conversion factors were calculated using both simple linear regressions across all locations and the ratio of the exposure according to each definition to the NIOSH 7400 definition for each location, and these two approaches yielded similar results.

Both the total and amphibole EMP concentrations were much higher in the eastern zone than in the western zone. The highest fractions of EMP concentrations were found for EMP that were 1-3 µm in length and 0.2 - 0.5 µm in width, which is a subset of Suzuki and Cleavage Fragment definitions. Therefore, the EMP counts based on the current standard NIOSH 7400 method are much lower than counts based on shorter EMP.

Similar exposure patterns were observed based on different EMP size definitions, consistent with the high degree of correlation between these EMP exposures. Therefore, the independent effects of the EMP of these various sizes will not be identifiable in epidemiological analysis. Given the

high degree of correlation between the various metrics, consistent with previous work by other researchers, a more reasonable metric might be to measure all EMP irrespective of size.

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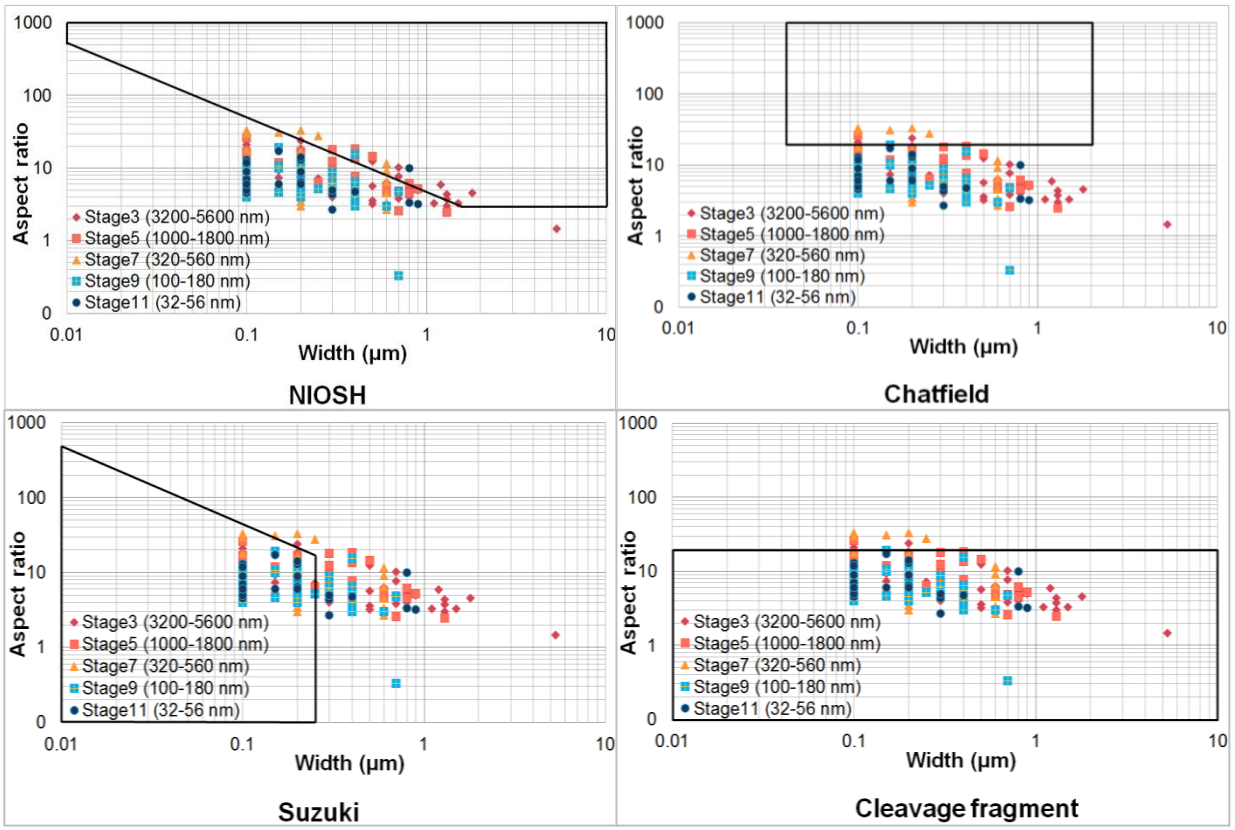

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Figure 1 Comparison of total EMP count for four dimension-based EMP exposure metrics by MOUDI impactor stage for the *Crusher Maintenance* location in mine A. The black boxes indicate the dimension-based EMP exposure metrics based on each definitions.

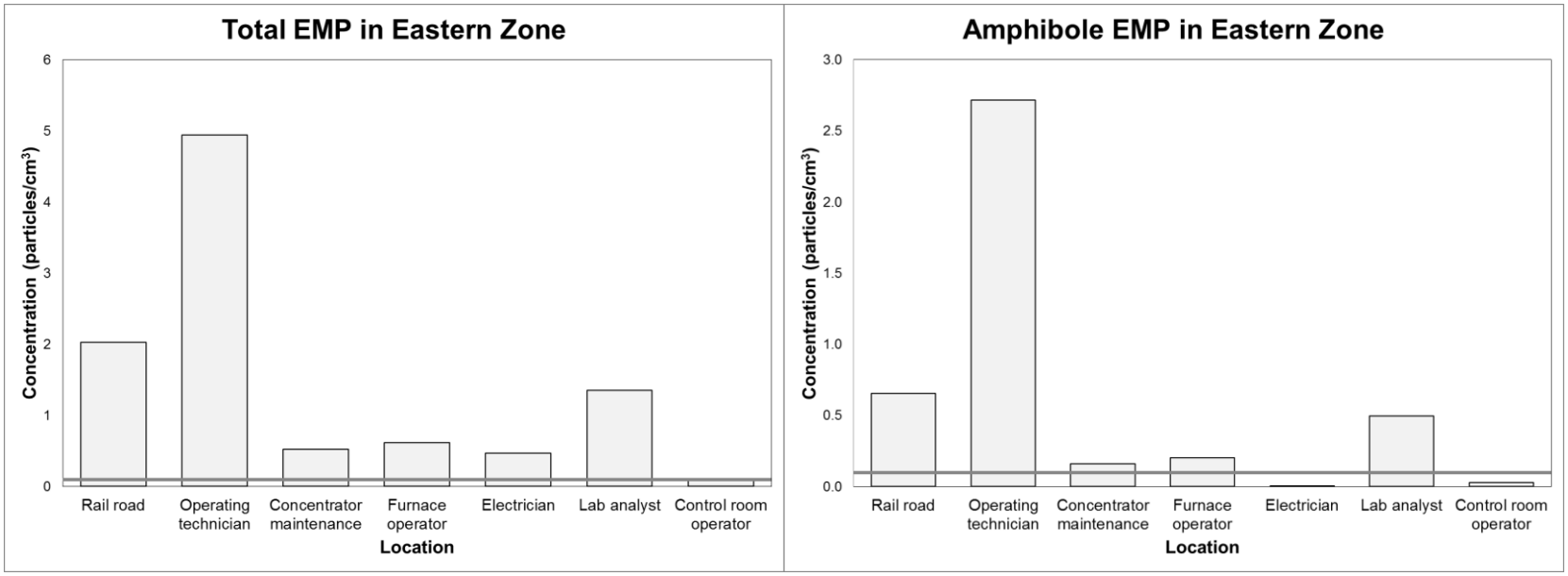

Figure 2 Total and amphibole EMP concentration by selected location in the eastern zone (line indicates the NIOSH REL for NIOSH 7400/7402 EMP= 0.1 particles/cm^3)

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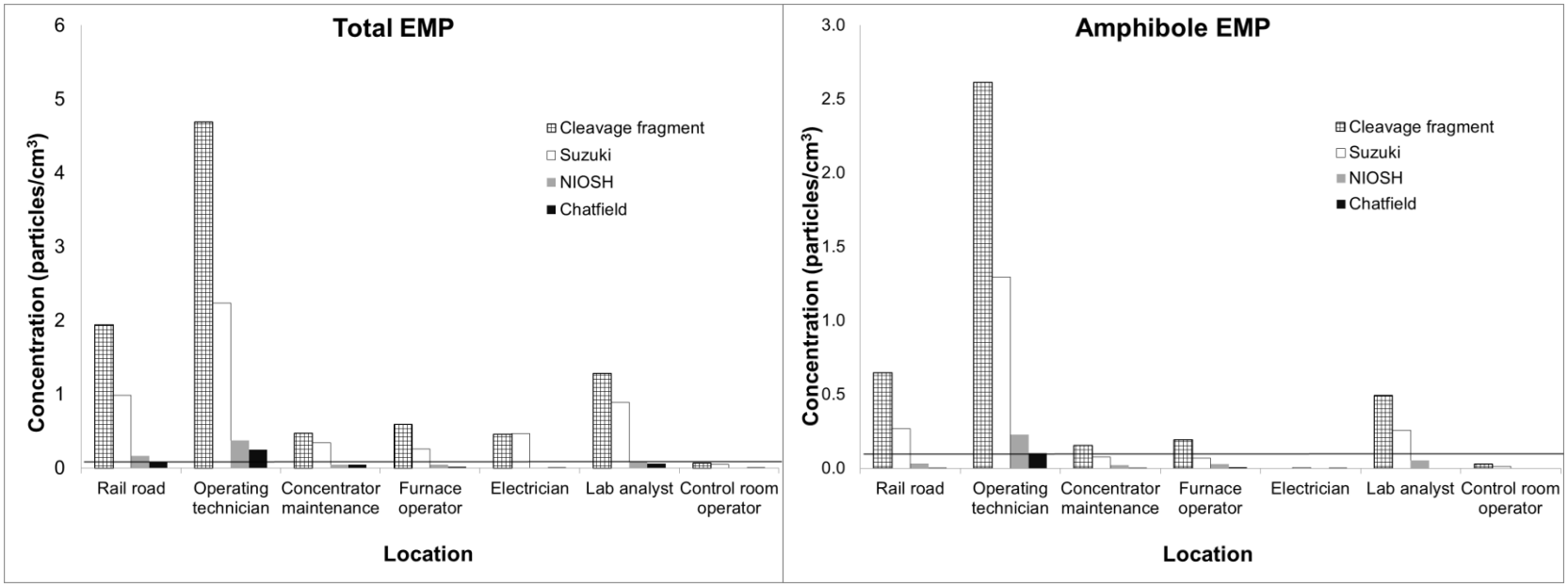

Figure 3 Total and amphibole EMP concentration by size-based definitions in eastern zone (line indicates the NIOSH REL for NIOSH 7400 EMP = 0.1 particles/cm^3)

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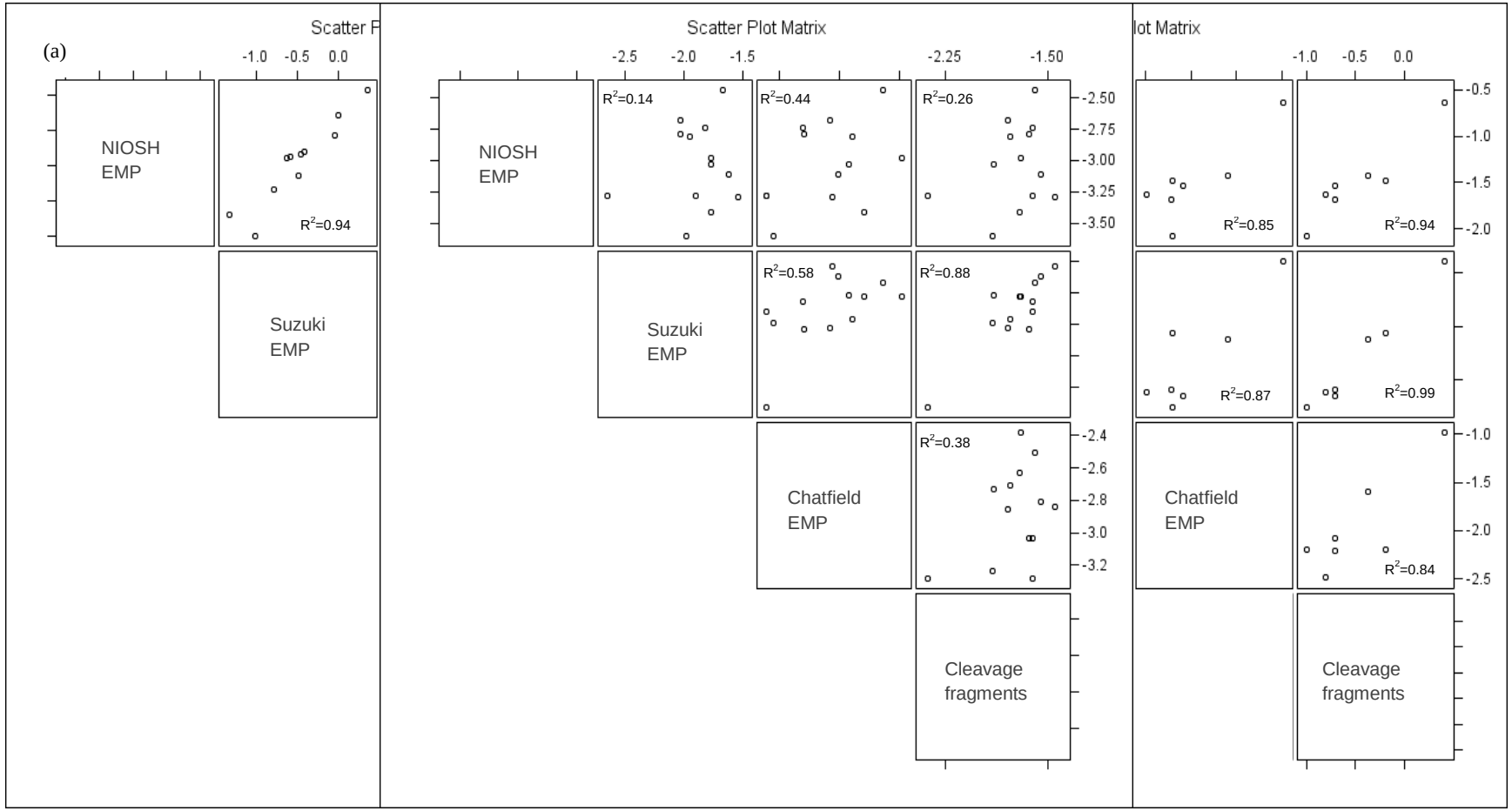

Figure 4 Coefficients of determination between (a) total and (b) amphibole EMP definitions in the eastern zone

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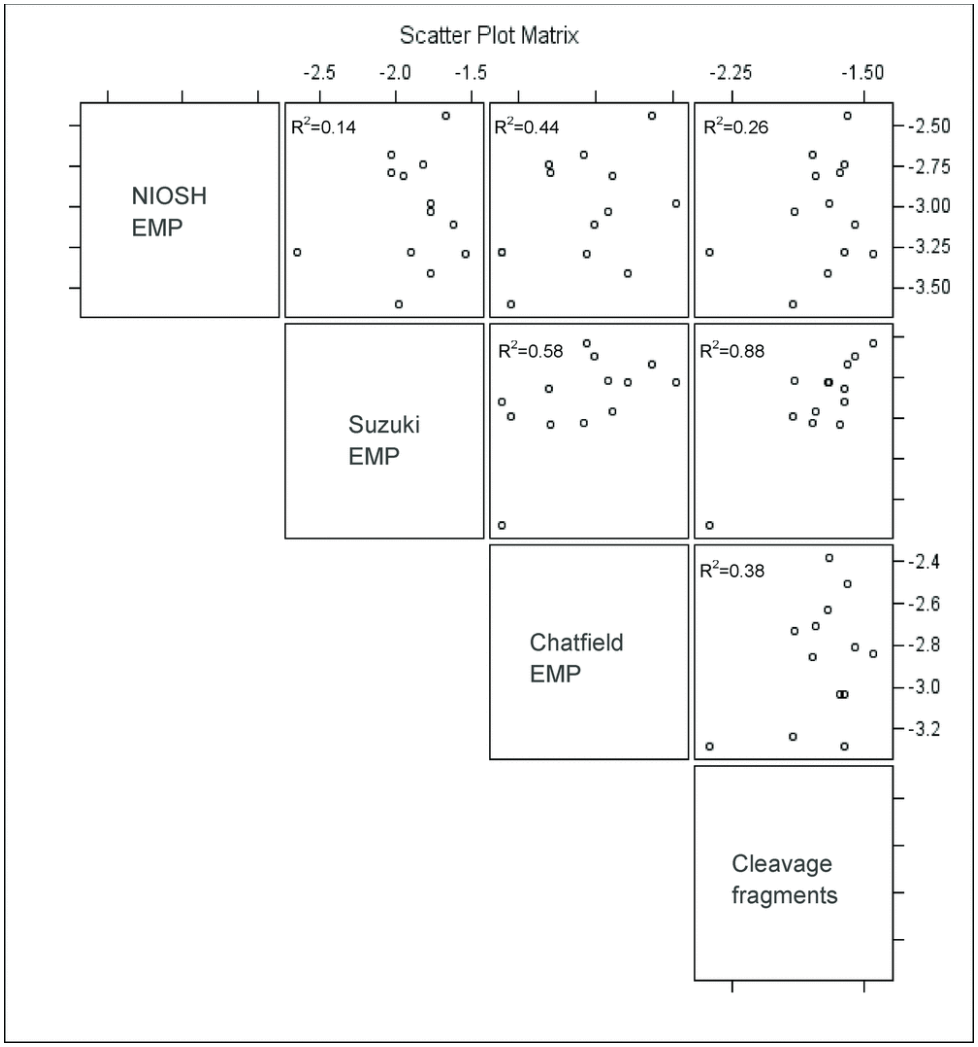

Figure 5. Coefficients of determination between total EMP definitions in the western zone

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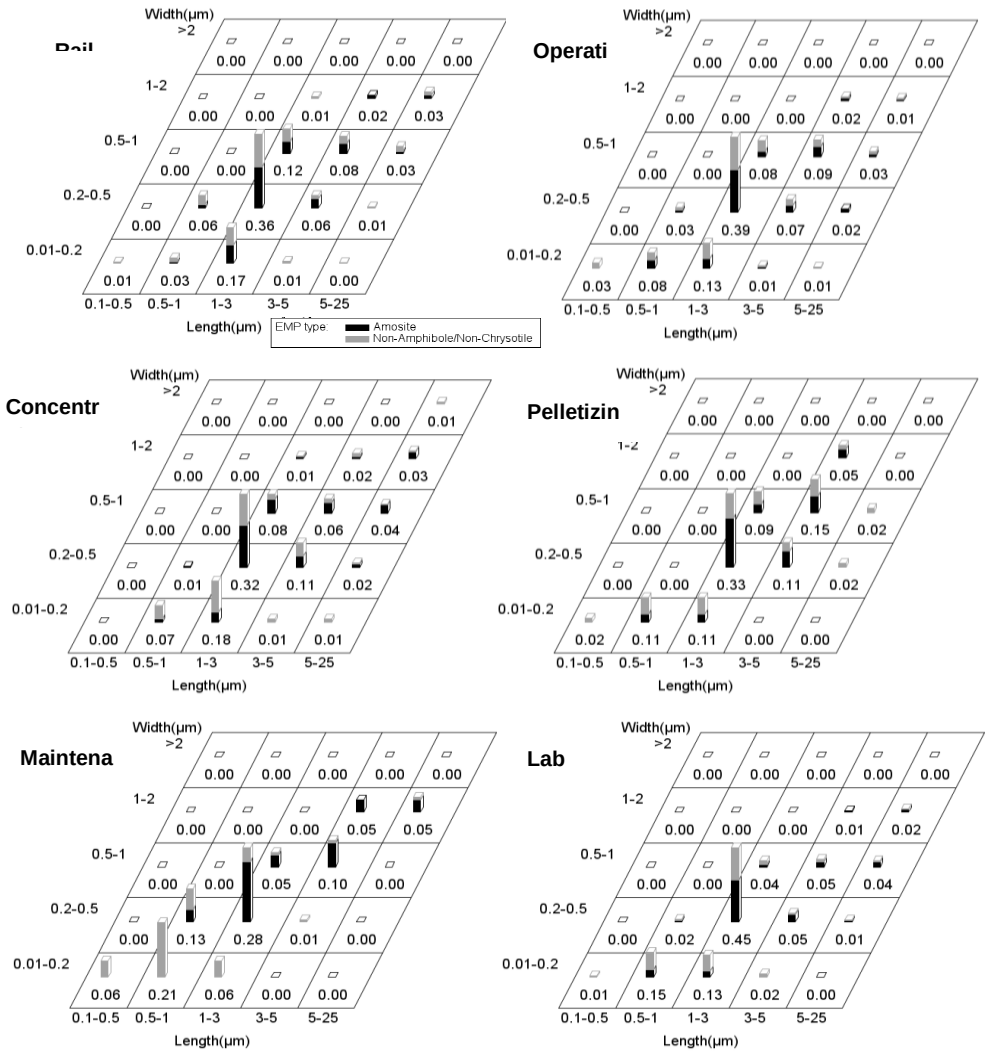

Figure 6. EMP size distribution by location in eastern zone. The height of the bar in each cell represents the fraction of the total EMP for that combination of length and width.

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Table 1 Characteristics of four dimension-based EMP metrics

Size-based definition	**Width (μm)**	**Length (μm)**	**Aspect ratio**	**Analysis methods** [a]
NIOSH EMP	-	> 5	≥ 3	PCM, TEM
Suzuki et al. EMP	W ≤ 0.25	≤ 5	-	PCM, TEM
Chatfield EMP	0.04 < W < 1.5	-	20 < AR < 1000	TEM
Cleavage fragment	-	-	AR ≤ 20	TEM

[a] PCM: Phase contrast microscopy

TEM: Transmission electron microscopy

Table 2 Number of MOUDI samples and locations by mine

Mine	# MOUDI samples	# Locations
A	23	17
B	14	14
C	20	16
D	12	11
E	15	15
F	8	8

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Table 3 Regression coefficient equations between NIOSH and other definitions for total EMP in eastern zone

$C_{Definition}$[a]	Total EMP		Amphibole EMP	
	East[e] (95% CI)	West[b]	East (95% CI)	West[c]
$C_{Suzuki\ EMP}$	$3.83C_{NIOSH\ EMP}^{0.744}$ (95% CI= 0.543, 0.945)	$13.0C_{NIOSH\ EMP}$	$6.40C_{NIOSH\ EMP}^{1.089}$ (95% CI= 0.829, 1.349)	NA[d]
$C_{Chatfield\ EMP}$	$0.501C_{NIOSH\ EMP}^{0.894}$ (95% CI= 0.600, 1.189)	$1.46C_{NIOSH\ EMP}$	$0.328C_{NIOSH\ EMP}^{1.011}$ (95% CI= 0.269, 1.707)	NA
$C_{Cleavage\ fragment}$	$8.28C_{NIOSH\ EMP}^{0.819}$ (95% CI= 0.662, 0.977)	$14.9C_{NIOSH\ EMP}$	$11.9C_{NIOSH\ EMP}^{1.062}$ (95% CI= 0.788, 1.335)	NA

[a] Concentration of all ISO 13794 EMP that meet a specific EMP size definition.

[b] Regression coefficients from both intercept and slope are not statistically significant at p-value = 0.5.

[c] Concentrations of amphibole by NIOSH are zero in the western zone except at the *Concentrator operator* location.

[d] Not applicable (NA) because no regression parameters are estimated for amphibole EMP in the western zone."

[e] The 95% confidence intervals are only shown for the slope (appearing as the exponent in this table).

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Table 4. Ratios of EMP concentrations based Suzuki, Chatfield, and Cleavage Fragment definitions to EMP concentrations based on NIOSH 7400 and 7402 definitions

Department	**Location**	**Total EMP**						**Amphibole EMP**					
		East			**West**			**East**			**West**		
		Suzuki	Chatfield	Cleavage	Suzuki	Chatfield	Cleavage	Suzuki	Chatfield	Cleavage	Suzuki	Chatfield	Cleavage
Mining	Basin operator	.	.	.	.	.	.	.	.[a]	.	.	.	.
	Mining operator 1	.	.	.	.	.	.	.	.	.	.	.	.
	Mining operator 2	.	.	.	.	.	.	.	.	.	.	.	.
	Rail road	6.0	0.48	12	.	.	.	8.2	0.19	20	.	.	.
Crushing	Crusher maintenance	7.6	0.60	12	8.4	0.51	14	6.5	0.68	11	.	.	.
	Crusher operator	6.0	0.69	12	4.5	0.67	7.7	3.9	0.30	9.5	—	—	—
	Operating technician	6.0	0.67	13	.	.	.	5.7	0.45	11	.	.	.
Concentrating	Concentrator maintenance	7.5	1.1	10	24	1.0	47	3.2	0.14	6.7	—	—	—
	Concentrator operator	.	.	.	57	2.8	71	.	.	.	2.0	0	3.8
Pelletizing	Balling drum operator	—	—	—	43	6.0	50	—	—[b]	—	—	—	—
	Dock man	—	—	—	0.0	0	1.0	—	—	—	—	—	—
	Furnace operator	6.2	0.35	14	7.1	1.3	11	2.4	0.29	6.7	—	—	—

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	Pelletizing maintenance	11	0.43	21	5.7	0.57	14	—	—	—	—	—	—
	Pelletizing operator	14	1.9	18	5.8	0.86	7.0	6.5	0.75	12	—	—	—
Shop mobile	Boiler technician	.	.	.	—	—	—	.	.	.	—	—	—
	Carpenter	.	.	.	4.8	0	5.8	.	.	.	—	—	—
	Electrician	—	—	—	—	—	—	—	—	—	—	—	—
	Lubricate technician	.	.	.	.	.	.	.	.	.	.	.	.
	Maintenance technician	31	1.0	39	31	2.0	36	3.3	0 [c]	12	—	—	—
	Pipefitter/Plumber	.	.	.	.	.	.	.	.	.	.	.	.
	Repairman	.	.	.	—	—	—	.	.	.	—	—	—
	Supervisor	.	.	.	.	.	.	.	.	.	.	.	.
Shop stationary	Auto mechanic	—	—	—	41	2.3	50	—	—	—	—	—	—
	Lab analyst	11	0.70	16	16	4.0	19	4.8	0	9.2	—	—	—
	Warehouse technician	—	—	—	18	2.0	14	—	—	—	—	—	—
Office/Control room	Control room operator	—	—	—	—	—	—	—	—	—	—	—	—
	Office staff	7. 7	0	13	4.3	1.0	8.0	0	0	3.7	—	—	—

[a] : No samples/ [b] : NIOSH is zero / [c] : Suzuki, Chatfield, or Cleavage fragment is zero.

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