ASSESSING PRESENT-DAY AND HISTORICAL EXPOSURES OF WORKERS TO TACONITE DUST IN THE IRON MINING INDUSTRY IN NORTHEASTERN MINNESOTA

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1. Health Studies and Exposure Assessment in Taconite Mining Industry

Minnesota counties in proximity to where taconite is mined have elevated age-adjusted rates for mesothelioma (Case *et al*., 2011). The apparent increase in cases has been of concern to communities within the Mesabi Range, many of which are situated immediately adjacent to the mining operations. There have been ongoing and unresolved concerns about health risks from taconite mining for the last three decades driven by limited epidemiological assessments and lack of quantitative exposure data. Reports of potential health issues related to taconite mining, especially to airborne taconite dust, began to appear in the 1980s. Initial reports concerned with pneumoconiosis (Clark *et al*., 1980; Higgins *et al*., 1983) were followed by mortality assessments in specific mining companies (Higgins *et al*., 1981; Cooper *et al*., 1988; Cooper *et al*., 1992). While these studies found no excesses of asbestos-related diseases, they had minimal to no information regarding worker exposures and had limited statistical power. The Minnesota Department of Health (MDH) conducted studies to understand the taconite workers' health in response to the rise in cases of mesothelioma between 1988 and 1996 under the Minnesota Cancer Surveillance System (MCSS, 1999). They found 17 workers who were diagnosed with mesothelioma during this period (MDH, 2003).

Since the term "fiber" has been controversial in the context of asbestos (e.g. Eastern Research Group, 2003), 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 any mineral particle with a minimum aspect ratio of 3:1 that is of inhalable, thoracic, or respirable size (NIOSH, 2011).

In 1973, asbestiform amphibole elongate mineral particles (EMP) were found in Lake Superior where tailings (residual waste) from the Northshore mine were disposed (Cook *et al*., 1974). Since then, a number of papers have focused on ingestion exposure, but the tests on rat gastrointestinal organs exposed to EMP showed no significant evidence of carcinogenic effects (Hilding *et al*. 1981). Similarly, in Gamble & Gibbs's (2008) review, there is a lack of evidence associating gastrointestinal cancers with EMP exposure, although lung cancer and mesothelioma are highly associated. Instead, most research on the Northshore mine disposal facility at the tailings basin focuses on inhalation exposure, which is the most common route of exposure to EMP. Therefore, this study limits its discussion to inhalation exposure.

Due to the possible use of commercial asbestiform EMP for insulation, asbestos cement, roof coatings, gaskets, etc., in the mining process, and due to the fact that most of the taconite processing facilities in the Mesabi Iron Range were built starting in the 1950s, the MDH conducted studies to distinguish between commercial asbestiform and naturally occurring asbestiform EMP from taconite dust. Specifically, Brunner *et al*. (2003) and Brunner *et al*. (2008) concluded that commercial asbestiform exposure may be more relevant to mesothelioma than taconite dust exposure. Brunner *et al*. (2008) reported on the relationship between asbestiform EMP and mesothelioma in the industry, finding that pleural mesothelioma occurred in 88% (15 out of 17) of cases and 12% of taconite miners were diagnosed with peritoneal mesothelioma. Interestingly, workers with exposures to non-commercial EMP or cleavage fragments in the taconite mining industry have been less likely to receive a mesothelioma diagnosis (Gibbs & Berry, 2008).

Only a few studies have investigated exposure to respirable dust and respirable silica (quartz) in the taconite mining industry. Sheehy (1986) conducted respirable quartz exposure sampling and found that respirable silica exposures in the taconite industry have often exceeded 0.1 mg/m^3, which was the standard NIOSH recommendation at that time. Sheehy and McJilton (1990) reported that the concentrations of silica-containing dust exposures were above acceptable limits in mines, crushers, and concentrators, but not in pellet plants. The limits referenced included the Recommended Exposure Limit (REL), set by the NIOSH, and the Permissible Exposure Limit (PEL), set by the Mine Safety and Health Administration (MSHA). Silica quartz concentrations ranged from less than 0.04 to 0.11 mg/m^3, and no tridymite or cristobalite was found in any of the samples (McJilton, 1984). Since the 1990s, exposure levels to respirable dust and respirable silica have not been studied for epidemiological purposes in the taconite mining industry.

2. Current Taconite Workers Health Study

In the mid-1980s a cohort of approximately 72,000 Minnesota taconite miners was assembled. In 1997 this cohort was matched with cancer information in the Minnesota

Cancer Surveillance System (MCSS). A total of 80 Mesabi iron miners have been diagnosed with mesothelioma since 2003. The number of cases is expected to increase because the latency period is long. Exposure to asbestiform EMP is strongly associated with respiratory diseases including mesothelioma, lung cancer, and nonmalignant respiratory disease. The frequency, duration, and concentration of exposure to taconite dust components are important factors to consider when explaining the adverse health effects. MDH (2007) reported that the mesothelioma rate for males in northeastern Minnesota was twice as high as the expected rate in the general population.

Concerns about the potential excess rates of mesothelioma in this cohort led the Minnesota State Legislature to fund the Taconite Workers' Health Study (TWHS) to evaluate mesothelioma, lung cancer, and non-malignant respiratory disease. The TWHS forms the basis for the proposed research, which is the first large scale effort to study the potential health hazards of exposures in taconite mining industry. The TWHS consists of four main investigations, including an exposure assessment. The exposure assessment provides comprehensive exposure information to the other studies: 1) The Mortality Study compares the causes of death, including mesothelioma and lung cancer, of the taconite workers with those of the general population in Minnesota. The exposure to each taconite dust component is evaluated with particular focus on mortality, cancer incidence, and lung cancer outcomes; 2) The Incidence Study, which is the case-control study, compares the incidence of mesothelioma and lung cancer among the taconite workers to that among other groups in a cohort. The exposure data implements the employment history to calculate a cumulative exposure amount for each case and control; 3) The Respiratory Health Survey determines the lung conditions of taconite workers and their spouses. This survey included questionnaire, physical exam, lung function test, and chest x-ray. The exposures to the taconite dust assesse the quantitative relationship between respiratory symptoms, x-ray, and lung function test for taconite workers and their spouses.

3. Characteristics of the Mesabi Iron Range

The Mesabi Iron Range lies in northeastern Minnesota and extends 200 km from Babbit in the east to Grand Rapids in the west. The taconite mining sites, which can be up to 5 km in width and 150 m thick, are horizontally divided into four zones by distinctive geological characteristics. The three currently operating taconite-mining companies own five mines in the western zone (Zone 1) and one in the eastern zone (Zone 4).

The Biwabik Iron Formation layered the Mesabi Iron Range with iron-rich rock that had been metamorphosed by intrusions of the Duluth Complex (Jirsa *et al*., 2008). The mineralogy of the formation changes from east to west across the Mesabi Iron Range. The east is mainly comprised of amphiboles; these amphiboles are principally of the cummingtonite-grunerite series and include some actinolite (ferroactinolite). The west is dominated by phyllosilicates such as minnesotaite, greenalite, and stilpnomelane, which are not regulated as asbestiform or amphibole “fibers” (McSwiggen & Morey, 2008; Zanko *et al*., 2008).

Amphiboles and phyllosilicates form two distinct groups of minerals, differentiated by fundamental differences in their internal crystalline structures. The structure of phyllosilicates consists of sheets of linked silicon tetrahedra. Fibers of phyllosilicate minerals are created when these sheets curl to form tubes. The structure of amphiboles consists of chains of silicon tetrahedra. These silicate minerals that form EMP have different morphologies; however, the vast majority of the amphiboles are non-asbestiform EMP (Wilson, McConnell, Ross, Axten, & Nolan, 2008; Zanko *et al*., 2008). Due to the distinct metamorphic mineralogical characteristics of the eastern and western zones, workers in the two zones may be exposed to different types of EMP, but their exposures to respirable dust and respirable silica may be similar.

4. Taconite Mining and the Milling Process

The Mesabi Iron Range in northeastern Minnesota contains low-grade iron ore called "taconite," which is mined and milled to make iron pellets. The mining and milling processes generate a significant amount of taconite dust. The taconite process can be divided into four different sub-processes—mining, crushing, concentrating, and pelletizing. In mining, holes are drilled into the extremely hard taconite rock to break it up. Production trucks haul the taconite directly to the crushing plant, during which quite a large amount of dust is generated. The sizes of the crushed ore vary by type of crusher and by mine. The crushing process produces a significant amount of dust, by definition (the ore is crushed until it is small). The ore is crushed to about 10 cm in diameter in the primary crusher and about 2 cm in diameter in the secondary crusher.

In the next process, concentrating, the rock is mixed with water and ground in rotating mills until it is a fine powder. Then, the iron ore is separated from the taconite using a magnetic separator, which removes the waste rock (called "tailing") from the iron-bearing grains of taconite powder (called "concentrate"). The flotation method is used to separate floating particles such as waste and silica from sink particles such as iron. Unlike crushing, concentrating which encompasses the wet process from magnetic separation as well as the silica removal process from flotation significantly reduces the amount of taconite dust generated.

The last process, pelletizing, removes water from the concentrated iron slurry, which it then mixes with bentonite clay to adhere the particles. Large rotating cylinders make balls about 1 cm in diameter, which are then dried and fired up to 1300 degrees Celsius. After the balls of concentrated taconite are cooled, the resulting taconite pellets are hard enough and durable enough to be shipped. During the summer months, pellets are loaded directly into ore cars.

During the winter, when the Great Lakes are frozen over, the finished pellets are stored in a huge pile, containing as much as 4 million tons, and then loaded into ore ships. The ships sail to Gary, Indiana; Cleveland, Ohio; and other steel-making towns. In the steel mills, the taconite pellets are melted down into steel.

5. Characteristics of Taconite Dust Components

The components of taconite dust that may have relevant adverse health effects are asbestiform EMP; non-asbestiform EMP, including cleavage fragments; and respirable silica (quartz) with respirable dust. Therefore, our analysis is limited to these dust components.

• Asbestiform EMP

Asbestiform EMP consist of two mineral groups: serpentine (containing chrysotile) and amphibole, of which there are five types (Table 1-1). Amphibole EMP have a crystal structure (chain silicates), while chrysotile has a sheet structure with hollow tubes (Wagner & Pooley, 1986). Amphibole EMP can be asbestiform or non-asbestiform (NIOSH, 2011). Asbestiform EMP are likely to be thinner, longer, and more flexible than non-asbestiform EMP, with layers parallel to those from “native (unprocessed) samples” (Addison & McConnell, 2008). Although the chemical composition of asbestiform and non-asbestiform EMP amphiboles can be the same, they differ in their morphology

(Langer *et al*., 1979). Asbestiform EMP are “polyfilamentous” while non-asbestiform EMP have a “multidirectional” pattern (Bailey *et al*., 2003). For consistency and clarity, in the remainder of this dissertation we refer the terminology defined in Table 1-2.

• Non-asbestiform EMP including Cleavage Fragments

In the taconite mining industry, cleavage fragments refer to the fractured particles, or mineral EMP, created during the crushing process rather than to naturally occurring EMP (NIOSH, 2011). Since no standard definition exists, distinguishing cleavage fragments from asbestiform EMP is challenging (Axten & Foster, 2008). Even if a given EMP counting criterion is met (e.g., ≥5 µm in length and >3 in aspect ratio), standard methods such as phase contrast microscopy (NIOSH 7400 method) cannot distinguish between them and cleavage fragments. Researchers have characterized cleavage fragments using biological experiments, mineralogical properties, dimension analyses, and chemical compositions (Wylie, 1990). They have found that non-asbestiform cleavage fragments are inactive *in vitro* bioassays and have less strength and flexibility from a morphologic perspective (Mossman, 2008). In addition, a linear relationship exists between the width and length of cleavage fragments, while no such relation exists in asbestiform EMP (Bailey *et al*., 2003). A detailed dimension analysis has been conducted by Addison & McConnell (2008), who concluded that cleavage fragments are likely thicker than asbestiform EMP due to cleavage (crystal) planes that have a lozenge-shaped cross-section. The chemical compositions of asbestiform EMP and cleavage fragments, however, are similar (Berndt & Brice, 2008).

• Respirable Dust and Respirable Silica (Quartz)

Respirable dust and silica are the most common dust components observed during the taconite mining processes – drilling, crushing, feeding, and transferring. Respirable dust is the fraction of dust (50% of cut-size is 4 μm) that penetrates into the respiratory system (Hinds, 1999). Respirable silica, which is a subset of respirable dust, consists of two mineral forms: crystalline (free silica) and amorphous. The crystalline form has three subgroups: quartz, tridymite, and cristobalite (Steenland & Stayner, 1997), the most common of which is quartz. In our study, we focused on the crystalline form-quartz. No adverse health effects, including lung cancer, have been observed due to exposure to the amorphous form of silica (IARC, 1997). Therefore, this study focuses on crystalline form-quartz.

Exposure to the crystalline forms of silica most commonly observed in industrial settings have long been associated directly or indirectly with the development of silicosis, a fibrotic pulmonary disease seen among workers in industrial occupations (Hayumbu *et al.*, 2008; Pelucchi *et al.*, 2006; Steenland & Sanderson, 2001; Archer *et al.*, 2002; Chen *et al.*, 2001; Collins *et al.*, 2005). Cocco (2003) found that investigations of silica exposure and its effect on the dose-response relationship have consistently relied on cumulative estimates of silica concentration and exposure duration, estimates that have been considered to hold the same weight. de Klerk & Musk (1998) ranked exposure intensities and found that the duration and intensity of silica exposure strongly affects the incidence of silicosis. Specifically, the initial exposure to silica affects the incidence of silicosis more strongly than an exposure 10 years later.

6. EMP Dimensions and Risk of Lung Disease

EMP characteristics related to pathogenic and toxic health effects include durability, habit, harshness, chemical composition, morphological differences, surface charge, and activity, as well as dimensions (Wylie, 1990; Baron, 2003; Mossman, 2008). A vast number of studies have been published on the health effects of asbestiform EMP, which are strongly correlated to lung cancer and mesothelioma. Fewer studies have discussed the health effects of non-asbestiform EMP, which might have different risks. Because of the difficulty of distinguishing non-asbestiform EMP from other EMP, the relationship between non-asbestiform EMP and adverse health effects is still controversial (NIOSH, 2011). However, due to the characteristics of the taconite process, non-asbestiform EMP, including cleavage fragments, are a potentially major source of exposure. Therefore, the adverse health effects examined in epidemiology studies may be significantly linked to not only asbestiform but also non-asbestiform EMP. To date, no single study has conducted an extensive assessment of the relationship between non-asbestiform EMP dimensions and adverse health effects.

As mentioned earlier, the current regulations for asbestiform EMP are based on length (≥5 μm) and aspect ratio (>3:1). This counting protocol has been criticized as lacking a scientific basis (Wylie *et al*., 1993; Addison & McConnell, 2008). Nevertheless, dimension is relevant to health effects because different sizes of EMP reach and deposit in different regions of the lung. EMP dimension is an important factor because (1) the macrophage cannot remove particles from the lung when the EMP length is longer than the macrophage diameter and (2) the lung cannot function when the thinner EMP deposit in the deeper alveoli region of the lung (Baron, 2003). However, the question of how the dimensions of the asbestiform EMP are related to the development of carcinogenic lung disease has been controversial. Although various researchers have examined different sizes of EMP to determine the most health-relevant, no consensus exists. Suzuki (2005) concluded that shorter (≤5 μm), thin (≤0.25 μm) EMP were more strongly associated with malignant mesothelioma through an analysis of lung and mesothelial tissues in human patients. Chatfield (2009) proposed a protocol for differentiating between asbestiform and non-asbestiform amphibole EMP, characterizing asbestiform EMP as between 0.04 μm and 1.5 μm in width and between 20 and 1000 in aspect ratio.

The lack of consensus on an appropriate exposure metric might partially explain the different exposure-response relationships obtained in different epidemiological studies. The current study was carried out as part of an epidemiological study to investigate the relationship between exposures to EMP during the mining and processing of taconite ore and the development of diseases such as mesothelioma, lung cancer, and non-malignant respiratory disease. This research provides an opportunity to study the relationship between each of the exposure metrics for amphibole and other EMP and the health outcomes.

7. Research Objectives

This research takes a multi-faceted approach to assess present-day and historical exposures to EMP (both amphibole and non-amphibole), respirable dust, and respirable silica in the taconite mining industry in northeastern Minnesota. This effort is part of a larger epidemiological study to assess the respiratory health effects of exposure to components of taconite dust. The main goals of the occupational exposure assessment are to accurately classify workers into similar exposure groups (SEGs) and to assess the present-day and historical exposures to the taconite dust components that are health-relevant. The specific aims of this research are as follows.

Aim 1.

Since it is inefficient and expensive to sample all workers, workers who have similar exposure profiles and whose tasks involve similar procedures and materials are assigned to a single SEG and only some of the workers from each SEG are sampled for exposure assessment. SEGs are widely used in industrial hygiene studies to more efficiently assess exposures based on job title, location, task, and procedure (Bullock & Ignacio, 2006). To reduce exposure classification errors, it is important that each SEG be distinct and homogeneous. Present-day exposures are assessed by measuring personal exposure levels of workers to EMP (both amphibole and non-amphibole), respirable dust, and respirable silica. This assessment enables us to test for differences in the levels of contaminants between different processes, mines, companies, and geological zones. The extensive measurement of present-day exposure levels carries significant implications for the validation of the SEGs. The detailed exposure assessments discussed later provide a better understanding of exposure levels as well as the exposure-response relationships in the studies. Therefore, the first aim is to classify the workers into groups that are distinct from each other and homogeneous within and to assess the present-day exposure levels for each taconite dust component.

Aim 2.

Area monitoring data are used to determine the distribution of EMP sizes. Based on these data, relationships between various dimension-based EMP exposure metrics are derived. The assessment of cumulative exposure according to these metrics enable epidemiological testing of various hypotheses regarding the health-relevance of different sizes of EMP. Therefore, the second aim is to develop a methodology for assessing EMP exposure levels based on a variety of dimension-based metrics.

Aim 3.

The exposure histories for individual workers are used to compute cumulative doses for the contaminants of concern in the epidemiological analyses. However, historical exposure data are missing for many jobs and for many time periods between 1955 and 2010 in all of the mines. Imputing the missing data requires an understanding of temporal trends and workplace conditions, enabling the use of a variety of statistical techniques and mathematical models to predict exposures. These techniques are used to create an exposure matrix that provides an estimate of exposure for every job title, listed by year, task, mine, company, and type of process. Therefore, the third aim is to reconstruct past exposures of EMP according to various definitions, respirable dust, and respirable silica as a function of time and job category over the period 1955-2010.

8. Significance

This work is the first comprehensive exposure assessment in taconite industry to address long-standing concerns over exposures and health in the Iron Range communities. We have assessed exposure to three major components of taconite dust including EMP, respirable dust, and respirable silica.

Second, it helps address the controversy over the potential health effects of non-asbestiform EMP and cleavage fragments. In many industries including taconite mining and processing, EMPs are created during mechanical processing of the ore (e.g., crushing and fracturing of the mineral) that are referred to as cleavage fragments. These cleavage fragments could meet the regulatory definition of a “fiber” described above, even if they were not asbestiform in habit. Non-asbestiform EMPs are chemically no different from asbestiform EMPs (Berndt & Brice, 2008), but are morphologically different. Non-asbestiform and cleavage fragments have not been thought to have high potential for disease (Berry and Gibbs, 2008; Gamble and Gibbs, 2008; Mossman, 2008). This issue is relevant to NIOSH roadmap (NIOSH, 2011). NIOSH has explicitly included EMPs from the non-asbestiform analogs of asbestos in its recommended exposure limit (REL). Their rationale for this decision was three-fold (NIOSH, 2011): (1) the epidemiological evidence from studies where worker populations were exposed to non-asbestiform EMPs (New York talc miners and millers, Homestake gold miners, and taconite miners) was considered inconclusive due to inadequate EMP exposure characterization, not accounting for smoking status, poor reliability of death certificate information, and exposures associated with prior employment; (2) animal studies showed differential toxicity of asbestiform and non-asbestiform EMPs with lower effects of exposure to non-asbestiform EMPs and some evidence that EMP dimensions may be predictors of toxicity, (3) current analytical methods used for routine analysis of samples, i.e., the NIOSH 7400 phase contrast microscopy (PCM) and NIOSH 7402 transmission electron microscopy (TEM) methods cannot differentiate between asbestiform and non-asbestiform EMPs when present in a heterogeneous mixture.

Third, the use of ISO 13794 for area sampling complements the use of NIOSH 7400/7402 for personal sampling, comprising a methodology that may be useful to the scientific community. Using area sampling to derive the relationships between the various dimension-based EMP exposure metrics enables us to test the health-relevance of the alternative metrics.

Fourth, supplementing the sparse historical data with a present-day exposure assessment facilitates the creation of SEGs using precise exposure data and minimizes the likelihood of exposure misclassification. The missing historical data is reconstructed using several different statistical imputation methodologies. Although some of these imputation methodologies have been used in prior epidemiology studies, the specific combination of methods used in our study is novel.

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Table 1-1 Types of asbestiform and non-asbestiform EMP

Mineral group	**Asbestiform**	**Non-asbestiform†**
Serpentine	Chrysotile	Antigorite, Lizardite
Amphibole	Amosite (grunerite asbestiform)	Cummingtonite-grunerite
	Crocidolite	Galucophane-riebeckite
	Tremolite asbestiform	Tremolite
	Actinolite asbestiform	Actinolite
	Anthophyllite asbestiform	Anthophyllite

†These forms include cleavage fragments.

Table 1-2 Asbestiform and other EMP terminology

Term	Definition	References
Elongated Mineral Particles (EMP)	Any mineral particle with a minimum aspect ratio of 3:1 that is of inhalable, thoracic, or respirable size	(NIOSH, 2011)
Asbestiform EMP	Minerals with a macroscopic habit similar to that of asbestos	(NIOSH, 2011)
Non-asbestiform EMP	Chemically no different from asbestiform EMP formula, but morphologically different as needle-like (acicular) or prismatic crystal	(Ilgren, 2004), (NIOSH, 2011)
Cleavage fragment	Generated by crushing and fracturing minerals including the non-asbestiform analogs of the asbestiform minerals	(NIOSH, 2011)

Chapter 2 Comprehensive Assessment of Exposures to Elongate Mineral Particles (EMP) in the Taconite Mining Industry

INTRODUCTION

Since the 1970s, concerns about occupational health have intensified in both the taconite mining industry and the communities adjacent to the mines in the Mesabi Iron Range in northeastern Minnesota (Wilson *et al*., 2008; Axten and Foster, 2008). Minnesota counties in the vicinity of taconite mining operations have been found to have elevated age-adjusted rates for mesothelioma (Case *et al*., 2011). The elevated rates challenge conventional understanding because mineralogical data suggest that the ore body is comprised primarily of non-asbestiform cleavage fragments which have not been thought to have high potential for disease (Berry and Gibbs, 2008; Gamble and Gibbs, 2008; Mossman, 2008). For the last three decades, ongoing and unresolved concerns about health risks from taconite mining have been driven, in part, by limited epidemiological assessments and insufficient quantitative exposure data. Concerns about the elevated rates of mesothelioma in the Mesabi mining cohort led to epidemiological investigations evaluating the relationship between cumulative exposures to components of taconite dust and mesothelioma, lung cancer, and non-malignant respiratory disease. However, no research on exposure to taconite dust, which includes elongate mineral particles (EMP), has been conducted since 1990 (Sheehy and McJilton, 1990).

The results presented here are part of a larger epidemiological study assessing the respiratory health effects of exposure to components of taconite dust. This paper describes our approach to comprehensively assess present-day exposure levels to total and amphibole EMP in the taconite mining industry. The term "total EMP" refers to any mineral particle with a minimum aspect ratio of 3:1 that is of inhalable, thoracic, or respirable size, while the term "amphibole EMP" refers to a subset of double chain silicate minerals (crocidolite, amosite, anthophyllite, tremolite, and actinolite) that can be asbestiform or non-asbestiform (NIOSH, 2011). Asbestiform EMP are likely to be thinner, longer, and more flexible than non-asbestiform EMP, with layers parallel to those from “native (unprocessed) samples” (Addison and McConnell, 2008). Although the chemical composition of asbestiform and non-asbestiform EMP can be the same, they differ in their “habit” or morphology (Langer *et al*., 1979).

The first and most critical step of our exposure assessment involves the classification of workers into similar exposure groups (SEGs). SEGs can be used to efficiently assess exposure levels based on job titles, locations, tasks, and procedures rather than individual workers (Bullock and Ignacio, 2006). Workers who have similar exposure profiles and whose tasks involve similar procedures and materials are grouped together in a single SEG. The success of a grouping strategy depends on the between-group variability, between-worker variability, and within-worker variability. To reduce exposure misclassification errors in subsequent epidemiological studies, it is important that the exposure distributions of SEGs be distinct from each other and homogeneous within (Kromhout and Heederik, 1995). This requires a detailed characterization of between and within-SEG exposure variability. However, the sparseness of the available historical exposure data precludes such an analysis for taconite workers. A detailed assessment of present-day exposure levels was carried out to understand exposure variability, which enabled the development of better-formed SEGs.

The mineralogy of the Mesabi Iron Range changes from east to west, with the three taconite mining companies owning five operating mines in the western and one in the eastern zone. Amphiboles are mainly detected in the east. Phyllosilicates such as minnesotaite, greenalite, and stilpnomelane, which are not regulated as asbestiform or amphibole EMP, dominate the west (McSwiggen and Morey, 2008; Zanko *et al*., 2008). The amphiboles in the east are principally of the cummingtonite-grunerite series and include some actinolite (ferroactinolite). Amphiboles and phyllosilicates form two distinct groups of minerals, defined by fundamental differences in their internal crystalline structure. The structure of phyllosilicates is based on sheets of linked silicon tetrahedra. Fibers of phyllosilicate minerals are created when these sheets curl to form tubes. The crystalline structure of amphiboles is based on chains of silicon tetrahedra. The silicate minerals that form EMP have different morphologies in the east; however, the vast majority of the amphiboles are non-asbestiform EMP (Wilson *et al.*, 2008; Zanko *et al*., 2008). Due to the distinct metamorphic mineralogical characteristics of the eastern versus the western zones, workers in the two zones may potentially be exposed to different types of EMP.

The goals of this paper are (1) to assess the present-day levels of exposure to EMP in the taconite industry across the two mineralogical zones, (2) to estimate the between-SEG, between-worker, and within-worker components of variability in EMP exposures, (3) to use the components of variability to assess whether the SEG are distinct from each other and relatively homogeneous within, and (4) to evaluate the impact of variability on the exposure estimates for the SEGs that will be used in the epidemiological studies. We also examined whether SEGs developed for total EMP are valid for amphibole EMP, and if the same set of SEGs can be used for workers in the mineralogically distinct eastern and western zones of the Mesabi Iron Range.

METHODS

Formation of SEGs

For this study, we derived job titles from four sources: (1) records maintained by the Mine Safety and Health Administration (MSHA) that listed approximately 190 job titles; (2) information from a previous University of Minnesota study by Sheehy (1986) that listed 140 job titles; (3) industrial hygiene and human resources databases maintained by the three companies currently operating mines in the Mesabi Iron Range (U.S. Steel, Cliffs Natural Resources, Arcelor Mittal), which listed approximately 150 job titles; and (4) *Job Descriptions and Classifications* published by the Reserve Mining Company in 1974, which contained 142 job titles. Using information on the tasks and processes related to these job titles, we created a set of 60 SEGs. This list was further condensed to 28 SEGs using the subjective professional judgments of the lead industrial hygienists at the three mining companies. The number of job titles represented in each SEG ranged from 1 to 19. The final list contained 181 job titles, forming 28 SEGs that we further grouped into seven departments. Due to the distinct mineralogical characteristics of the eastern versus the western zones, the SEGs for the eastern and western zones were considered separately.

Sampling design and data handing

Personal exposure assessment was conducted across all operating mines in both zones of the Mesabi Iron Range beginning in January 2010 and ending in May 2011. The purpose of the personal sampling was to assess the present-day levels of worker exposures to EMP in the taconite mining industry. The researchers and representatives from each of the three mining companies discussed workers' schedules to identify potential participants prior to the day of sampling. At the beginning of the work shift on each sampling day, the researchers explained the purpose of the study to the potential participants and presented them with the consent form approved by the University of Minnesota Institutional Review Board (IRB code: 0901M58041).

To perform a baseline exposure profile for a job title, the American Industrial Hygiene Association (AIHA) sampling strategy by Bullock & Ignacio (2006) recommends a minimum of six data points per SEG and recommends eight to ten. Two workers per SEG were selected for personal EMP sampling in the eastern zone and each worker was sampled during three different shifts. In the western zone, approximately eight workers per SEG were chosen, with each worker being sampled on three different shifts. For the SEGs in the western zone, the eight workers were drawn from five different mines. This design allows the estimation of between- and within-SEG, between- and within-mine, between- and within-zone, and within worker variance components.

Each consenting participant wore a personal air-sampling pump (Apex Pro pump, Casella Inc., Amherst, NH) on his or her waist, with the sampler located in the breathing zone, for approximately six hours during the work shift. Six hours accounts for at least 70% of a daily work shift. Personal sampling for each worker was completed during three different work shifts, though not necessarily on consecutive days.

EMP sampling was conducted using a mixed cellulose ester (MCE) membrane filter 25 mm in diameter with 0.8 μm pores. The filter was placed in a polycarbonate membrane (PCM) cassette with a conductive extension cowl of 50 mm. The flow rate for the EMP sampling pump was set at the lowest available flow rate per pump to avoid overloading the filter (range 0.65-0.95 liter/min). As a further precaution against overloading, the PCM cassettes usually were changed at the end of about the first three hours of sampling. Overall, eighteen samples were excluded because they were either overloaded particles or damaged filter. Exceptions were made if the participants had a conflict in their work schedule or the researchers decided not to change the cassettes due to lower expected particle exposure levels for some samples (e.g., Warehouse technician, Office staff).

Analytical methods and limitations

The personal filter samples were analyzed by phase contrast microscopy (PCM) using NIOSH Method 7400 (NIOSH, 1994a), which identifies all EMP longer than 5 µm with an aspect ratio ≥ 3.0 (Counting Rules A). While this method can be used to count the number of EMP, it cannot differentiate between asbestiform and non-asbestiform EMP. While it is commonly stated that NIOSH 7400 cannot identify EMP with a width less than 0.25 µm (e.g., NIOSH, 1994a), this depends on the refractive index of the EMP (NIOSH, 2011). If the refractive index does not differ from the substrate material or the counting medium, the resolution is low, and vice versa (Kenny and Rood, 1987). Rooker *et al*. (1982) have shown that under proper calibration and use of appropriate mounting media, EMP with widths of 0.15 µm were measured using PCM. Kenny and Rood (1987) measured widths of 0.125 µm under PCM.

In contrast, the NIOSH Method 7402 by Transmission Electron Microscopy (TEM) (NIOSH, 1994b) is used to identify EMP that meet the PCM counting criteria. This method includes expanded characterization of elemental composition with energy dispersive X-ray analysis (EDXA) and crystalline structure by selected area electron diffraction (SAED). Therefore, it can identify EMP that are amphiboles or chrysotile. 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), especially in the heterogeneous mixture of particles found in the taconite industry in Minnesota.

As indicated previously, two samples per work shift were collected for most participants on three different days. The results from the two samples were combined to calculate a single time-weighted average (TWA) concentration for the shift for each participant. While all personal EMP samples were analyzed using NIOSH 7400, at least one sample per worker was randomly chosen to be analyzed using NIOSH 7402. Thus, while all of the filter samples underwent analysis using NIOSH 7400, ~18% of the samples underwent additional analysis using NIOSH 7402. For the NIOSH 7402 analysis, samples were analyzed by indirect preparation, which included suspension in solution, sonication, and re-filtration. For all personal samples, we used only ¼ or ½ of the filter depending on the analysis methods chosen and the remaining ¾ or ½ has been archived at the University of Minnesota.

Table 2-1lists the number of personal samples analyzed using both NIOSH 7400 and NIOSH 7402 for each mine and zone. In addition, one blank sample per sampling day was obtained for NIOSH 7400 quality control for a total of 243. Further, one blank sample per NIOSH 7402 sampling day was obtained for quality control for a total of 66. Table 2-1 also shows the percentage of samples with EMP levels that fell below the limit of detection (LOD) as measured by NIOSH 7400 and NIOSH 7402, respectively. Overall, many of the samples had levels less than the LOD, especially the NIOSH 7402 samples in the western zone. If all the measurements for a given SEG were below the LOD, summary statistics such as the arithmetic and geometric means and geometric standard deviations were not reported. If at least one sample for an SEG in a particular mine was above the LOD, then summary statistics were calculated by assuming that censored data were represented by one half of the LOD.

Only three chrysotile asbestiform EMP (0.24% of all EMP samples) were identified by the NIOSH 7402 analysis. These were excluded from our analyses, leaving only amphibole—specifically amosite—and non-amphibole EMP in our data set. Using the NIOSH 7400 and 7402 results, average concentrations of EMP identified as total and amphibole for each SEG in each mine were calculated. This estimate was then applied to all of the NIOSH 7400 samples for that SEG in that mine to obtain personal exposure levels to NIOSH 7402 amphibole EMP when the samples had at least one value above LOD for that SEG.

$$C_{ij} \text{ (NIOSH 7402, amphibole EMP)}$$

$$= C_{ij} \text{ (NIOSH 7400, total EMP)} \times \frac{\overline{C}_i\text{(NIOSH 7402, amphibole EMP)}}{\overline{C}_i\text{(NIOSH 7400, total EMP)}} \qquad \text{(Equation 2- 1)}$$

for C= Concentration (particles/cm^3), $\overline{C}$=Average concentration (particles/cm^3), i=SEG in a mine, j=observation

Statistical analysis methods

Of the 28 SEGs, 27 SEGs were monitored. We were not able to monitor the Janitor SEG because all janitors in the current taconite mining industry are independent contractors and not employed by the mining companies. A t-test was used to determine which SEGs differed between the two zones for each EMP classification (Table 2-2). Of the 27 SEGs, 21 were present in both zones for statistical evaluations. To ensure that at least one of the 27 SEGs is different from the others and that the exposures within each SEG are homogeneous, two different approaches were used to evaluate the variability of exposure between-SEGs, between-workers, and within-workers.

A one-way ANOVA: We used a simple one-way ANOVA model to compare the log-transformed estimated exposures Y_{ij} of each SEG.

$$Y_{ij} = \log (X_{ij}) = \mu_y + \alpha_i + \varepsilon_{ij} \text{ for } i = 1, 2, \ldots, 27, \text{ and } j = 1, 2, \ldots, 24 \qquad \text{(Equation 2- 2)}$$

where X_{ij} = exposure concentration of the ith SEG at the jth observation for each SEG, μ_y = overall mean of Y_{ij}, α_i = random deviation of the ith SEG's true exposure from μ_y, and ε_{ij} = random deviation of the jth observation from the ith SEG's true exposure. Equation.2-2 assumes that the ε_{ij} are independently and identically distributed with a normal distribution. This model was used to determine if the differences between the SEGs were statistically significant. A pairwise comparison of the SEGs was used to identify which SEGs were significantly different from each other.

Contrast and homogeneity: Kromhout and Heederik (1995) proposed a two-way nested random-effects ANOVA model for estimating between-SEG , between-worker, and within-worker variance components using the log-transformed exposure concentrations. The variance components were constructed using PROC NESTED with a nested structure of dataset as:

$Y_{ikn} = \log(X_{ikn}) = \mu_y + \alpha_i + \beta_{ik} + \varepsilon_{ikn}$ for the observations i =1, 2, ...27, k = 1, 2, …, 4, and n =1, 2,…6 (Equation 2- 3)

where X_{ikn} = nth observation of exposure concentration for the kth worker of the ith SEG, μ_y = overall mean of Y_{ikn}, α_i = random deviations of the ith SEG's true exposure from $\mu_{y,}$ β_{ik} = random deviations of the ith SEG's kth worker's true exposure from $\mu_{y,\,i}$ (mean exposure of the ith SEG), and ε_{ikn} = random deviations of the nth observation for the ith SEG's kth worker from $\mu_{y,\,ik}$ (mean exposure of the kth worker in the ith SEG). The random deviations (α_i, β_{ik}, and ε_{ikn}) are assumed to be normally distributed with zero means and variances (σ^2_α, σ^2_β, and σ^2_ε, respectively). These variances are mutually uncorrelated and estimated as variance components ($S_y{}^2{}_{BG}$, $S_y{}^2{}_{BW}$, and $S_y{}^2{}_{WW}$, respectively).

According to Kromhout and Heederik (1995), contrast (ε) is a ratio of between-SEG variability to the sum of between-SEG and between-worker (i.e., within-SEG) variability, and provides an overall measure of whether there are distinctions between the SEGs and is given by:

$$\text{Contrast } (\varepsilon) = \frac{S_y{}^2{}_{BG}}{S_y{}^2{}_{BG} + S_y{}^2{}_{BW}} \qquad \text{(Equation 2- 4)}$$

When the between-SEG variance component ($S_y{}^2{}_{BG}$) approaches 0, the contrast value approaches 0, indicating that the SEGs are similar and not distinct from each other. When the between-worker variance component within the SEG ($S_y{}^2{}_{BW}$) approaches 0, the contrast value approaches 1, indicating that between-SEG variability are dominant and implying that at least one SEG is distinct from the others.

Analogously, we can define homogeneity (η) to provide an overall measure of how similar the exposures are for workers within an SEG. It is defined as the ratio of the within-worker variance component to the sum of the between-worker and within-worker variance components, and is given by:

$$\text{Homogeneity } (\eta) = \frac{S_y^2{}_{WW}}{S_y^2{}_{BW} + S_y^2{}_{WW}} \qquad \text{(Equation 2- 5)}$$

When the within-worker variance component ($S_y^2{}_{WW}$) is small compared to the between-worker variability, homogeneity approaches 0, indicating that the exposures of the workers within each SEG are heterogeneous. When the between-worker variance component ($S_y^2{}_{BW}$) is small, homogeneity approaches 1.

The statistical analyses were conducted for total and amphibole EMP. Significance was defined by p-values of 0.05 or lower. All analyses reported here were conducted using SAS version 9.3 (SAS Institute, Cary, NC, USA).

RESULTS

The results of t-tests used to determine the differences between the zones by SEG are shown in Table 2-2. When a SEG was not present in both zones, the p-value could not be calculated. Sixty-two percent (13 of 21) of the SEGs were significantly different between the zones for total EMP. For the amphibole EMP exposures in the western zone, all the data were less than the LOD. Additionally, eight SEGs in the eastern zone contained all data less than the LOD. Therefore, we did not test for differences between two zones for amphibole EMP. Both the total and amphibole EMP classifications had substantially different arithmetic mean exposures between the two zones. Only four SEGs (Balling drum operator, Pelletizing maintenance, Warehouse technician, and Office staff) were found to have higher total EMP exposures in the western zone, but none of these four were significantly different between the two zones (p-value > 0.05).

Total and amphibole EMP concentrations

The box-plots in Figure 2-1show the total EMP concentrations by SEG across all mines. The concentration of total EMP in Northshore tended to be higher than in the mines in the western zone. For most of the SEGs in the various mines, the arithmetic mean (the X in the box plot) was greater than the median (the middle line in the box plot), indicating a non-normal, skewed distribution.

Table 2-3 shows the geometric means (GM) and geometric standard deviations (GSD) of total EMP concentration by SEG in all mines. Table 2-4 summarizes the amphibole EMP concentration by SEG in the eastern zone (Northshore). Since all amphibole EMP concentrations are less than the LOD in the western zone, we do not present the GM and GSD estimates. The GM for each SEG in Northshore was markedly less for amphibole EMP than for total EMP.

The measured amphibole EMP concentrations by SEG across all mines are illustrated using scatter-plots in Figure 2-2. Figure 2-2 shows that, with a few exceptions in Northshore, the concentrations of amphibole EMP were lower than the NIOSH recommended exposure limit (REL) of 0.1 particles/cm^3 for EMP by roughly an order of magnitude.

Comparison of EMP exposure differences

To explore the EMP exposure differences between the SEGs, a pairwise comparison of the SEGs within each mine was performed. The logarithms of the estimated EMP exposures were used in a simple one-way ANOVA model. In each mine, at least two of the SEG means were significantly different for total EMP (p-values: <0.0005).

Comparison of SEG variance components

Table 2-5 shows the between-SEG (S^2_{BG}), between-worker (S^2_{BW}), and within-worker (S^2_{WW}) variance components as absolute values and as percentage of total variance (sum of the three components), as well as the contrast (ε) and homogeneity (η) values for total EMP by mine in both geologic zones.

DISCUSSION

The available historical data on exposure of workers to taconite EMP are sparse and typically based on NIOSH 7400. They are insufficient for assessing exposure variability in any detail. Our detailed measurements allow for a study of the components of variance of exposure, that in turn, allows the creation of well-formed SEGs and reducing the likelihood of exposure misclassification (Nieuwenhuijsen, 1997; Ramachandran, 2005). Moreover, this analysis identifies notable heterogeneity of exposure to total EMP in the taconite mining industry.

Levels of total and amphibole EMP

This is the first study to report on the concentrations of total and amphibole EMP in the taconite mining industry. Overall, higher concentrations of total EMP were found in Northshore, including the highest exposure of 2.2 particles/cm^3, ~ 22 times greater than the PEL (0.1 particles/cm^3) for EMP. The lowest concentration of total EMP was found in Minorca, and the total EMP exposure concentrations for all SEGs in this mine were lower than the PEL. The concentrations of amphibole EMP were much less than the concentrations of total EMP, indicating that amphibole EMP are not major components of taconite EMP. In general, the amphibole EMP concentrations were lower than the PEL, except for a few SEGs in Northshore. Three individual measurements exceeded the PEL of the amphibole EMP.

Comparison of eastern and western zones

Overall, the exposure levels were higher in the eastern zone than in the western zone. The differences in the exposure levels support the idea of considering the SEGs in the eastern and western zones separately for the larger epidemiology study, and are consistent with the geological differences between the zones. For both total and amphibole EMP categories, the SEG with the highest exposure level in the eastern zone was Operating technician (Table 2-2). In the western zone, the Pelletizing operator was the SEG with the highest exposure levels for total EMP (Table 2-2). More than half of the SEGs had significantly different levels of total EMP exposures between the eastern and western zones. This analysis provides empirical evidence that the geological differences between the two zones are reflected in EMP exposures.

The highest concentration in each mine was observed not only in departments directly involved in the mining process (Mining, Crushing, Concentrating, and Pelletizing departments), but also in the Shop (mobile) department, suggesting that the non-mining process may be similarly affected. The employees in the Shop (mobile) department work at various places in the mine, rather than at specific workstations. Therefore, the characteristics of the exposure levels for this department can be similar to those found in the mining process, and these SEGs potentially can have high exposure levels.

When the amphibole EMP concentrations are subtracted from the total EMP concentrations in the eastern zone, there remains a substantial excess of non-amphibole EMP concentration. This is significantly higher than the non-amphibole EMP concentration in the western zone for most SEGs. It is possible that this difference in non-amphibole concentrations between the zones is related to the mineralogy. As described earlier, there are distinct metamorphic mineralogical differences between the zones. Phyllosilicates are prevalent in the western zone, while amphiboles are prevalent in the eastern zone. However, an analysis of how mineralogy affects the non-amphibole EMP concentration is beyond scope of this study.

Analysis of between-SEG and between-worker variability

The SEGs formed for this analysis identify workers with similar exposures; however the exposures to EMP do not vary across all SEGs an only certain SEGs contribute significantly to variance. The between-SEG variance component was higher than the between-worker variance component in the eastern zone. Therefore, at least one of the SEGs is significantly different from the other SEGs in the eastern zone. However, the others may still not be distinguishable. Within the western zone, the between-SEG variance component was highest in Keetac and the between-worker variance component was highest in Minorca for total EMP.

Much higher contrast was observed in the eastern zone (0.740) than in the western zone (0.130). Since the western zone included five different mines, each SEG included exposures from five different mines, leading to higher between-worker (or within-SEG) variability, which in turn led to lower contrast. In particular, the between-SEG variance component was low in the western zone except Keetac for total EMP. Across the five mines in the western zone, there was a wide range of contrast values (0.000-0.865 for total EMP). Contrast was zero in Minorca for total EMP (see Table 2-5). However, the smallest number of subjects was monitored and the fewest number of samples were taken at Minorca. The variability for each SEG in Minorca was also the least (GSD range: 1.10-2.81) for total EMP, as shown in Table 2-3. Interestingly, the percentage of the between-worker variance component was ~8% in Keetac in the western zone, which led to high contrast regardless of the value of the between-SEG variance component.

The between-worker variance is the only component that affects both contrast and homogeneity. A smaller value for the between-worker variance component leads to higher contrast and homogeneity of the SEG and thus increases the ability to identify exposure differences between the SEGs. The between-worker variance component was lower in the eastern than in the western zone, a finding consistent with the lower contrast in the western zone. For example, for total EMP, the pronounced fluctuation in the pattern of EMP concentrations between-SEGs in Keetac is reflected in the highest S^2_{BG} as shown in Figure 2-1 and Table 2-5, respectively. Likewise, the stable pattern of EMP concentrations between-SEGs found in the Minorca is reflected in the lowest S^2_{BG} for that mine.

Analysis of within-worker variability

Within-worker variability was higher in the eastern zone than the western. Although taconite processes are similar across all mines currently, the responsibilities for similar job classifications varied slightly between the mines due to the presence or absence of unionization, number of employees, and management. For instance, the workers at

Northshore, the sole mine in the eastern zone, are non-unionized, and the tasks performed by workers with the same job titles vary more depending on the work shift. Censored data, or values less than the LOD, also impact estimated within-worker variability. A higher percentage of values below the LOD were observed in the western zone, which led to the lower estimated within-worker variability.

The highest S^2_{WW} was observed in Keetac and the lowest in Hibbtac for total EMP. Overall, S^2_{WW} was the dominant variance component compared to S^2_{BG} and S^2_{BW}, for total EMP for all mines except Keetac and Minorca. This finding indicates that the workers' daily tasks are the main source of variability rather than environmental influences. Higher homogeneity was found in the eastern zone than in the western.

Optimality of SEGs

Our results suggest that, in the eastern zone, the SEGs that we defined are formed well-enough for total EMP. The pairwise comparison of SEGs between the two zones indicates that 62% of the SEGs had significantly different levels for total EMP. However, for the amphibole EMP, the p-value for each SEG was not comparable due to LOD presented in either one or both zones. Specifically, the western zone had lower values for contrast and homogeneity than the eastern zone. The primary reason we have low contrast between-SEGs in the western zone is that all amphibole EMP exposure levels in the western zone were below the LOD.

As described earlier, Department is a grouping variable that can be used as an alternative to SEG. Therefore, we also evaluated the variance components at the departmental level. However, the contrast and homogeneity values were lower than those calculated for the original SEGs. This finding reconfirmed that the original SEGs were as good as; if not better than, other possible grouping schemes that we considered and represent an appropriate level of analysis.

CONCLUSIONS

For many SEGs in several mines, the exposure levels of total EMP were higher than the PEL for EMP. 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. The concentrations of amphibole EMP were well controlled across all mines and were much lower than the concentrations of total EMP, indicating that amphibole EMP are not major components of taconite EMP. Overall, we found that the variability of each SEG across mines was small for both total and amphibole EMP. Theoretically, the variability in the eastern zone should have been lower than the western, as it consists of only one mine as opposed to five. However, due to the low concentration of EMP (often below LOD), we found lower variability in the western zone. When we compared zones, higher values for contrast and homogeneity were observed in the eastern zone. While low contrast and homogeneity was observed for the western zone taken as a whole, higher values were observed when these parameters were calculated for each mine. We conclude that the SEGs that we defined are appropriate for use in an epidemiological study when grouped by mine for total EMP.

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Table 2-1 Number of personal samples and percent of samples < limit of detection (LOD) by mine and mineralogical zone

Zone	**Mine**	**Workers**	**Samples analyzed by PCM**[a]	**%<LOD by PCM**	**Samples analyzed by TEM**[b]	**%<LOD by TEM**
Eastern	Northshore	56	266	7.1	102	68.6
Western	Hibbtac	34	197	68.5	34	100.0
	Utac	38	218	53.2	36	100.0
	Keetac	34	203	37.0	34	100.0
	Minntac	48	267	20.6	47	100.0
	Minorca	22	129	48.8	22	100.0
Total		232	1298		275	

[a] Personal samples analyzed by NIOSH 7400 phase contrast microscopy (PCM), counting all EMP with length > 5 µm and aspect ratio > 3.0.

[b] Personal samples analyzed by NIOSH 7402 transmission electron microscopy (TEM), counting only amosite, non-amosite, and chrysotile EMP with length > 5 µm and aspect ratio > 3.0.

Table 2-2 Arithmetic mean (particles/cm^3) in each zone and t-test results (p-value) by EMP classification for each SEG

Department	**SEG**	**Total EMP (particles/cm^3)**			**Amphibole EMP (particles/cm^3)**		
		East	**West**	**p-value**	**East**	**West**	**p-value**
Mining	Basin operator	.	0.053	.		<LOD	.
	Mining operator 1	**0.065**	**0.015**	**<0.0001**	<LOD	<LOD	NA
	Mining operator 2	**0.097**	**0.031**	**0.0016**	0.004	<LOD	NA
	Rail road	0.072	.	.	<LOD		.
Crushing	Crusher maintenance	**0.194**	**0.044**	**<0.0001**	0.026	<LOD	NA
	Crusher operator	**0.193**	**0.038**	**<0.0001**	0.030	<LOD	NA
	Operating technician	**0.341**	**0.014**	**<0.0001**	0.110	<LOD	NA
Concentrating	Concentrator maintenance	**0.207**	**0.058**	**<0.0001**	0.030	<LOD	NA
	Concentrator operator	**0.176**	**0.023**	**<0.0001**	0.024	<LOD	NA
Pelletizing	Balling drum operator	0.050	0.077	0.9371	0.010	<LOD	NA
	Dock man	**0.206**	**0.085**	**0.0014**	0.020	<LOD	NA
	Furnace operator	**0.066**	**0.040**	**0.0141**	0.015	<LOD	NA
	Pelletizing maintenance	0.067	0.073	0.0852	<LOD	<LOD	NA
	Pelletizing operator	0.109	0.095	0.1739	0.014	<LOD	NA
Shop (mobile)[a]	Boiler technician	.	0.041	.	.	<LOD	.
	Carpenter	.	0.064	.	.	<LOD	.
	Electrician	**0.309**	**0.077**	**<0.0001**	0.063	<LOD	NA
	Lubricate technician	**0.145**	**0.033**	**0.0006**	0.016	<LOD	NA
	Maintenance technician	0.043	0.031	0.0919	<LOD	<LOD	NA
	Pipefitter/Plumber	.	0.048	.	.	<LOD	.
	Repairman	.	0.064	.	.	<LOD	.
	Supervisor	0.064	0.045	0.3246	0.012	<LOD	NA
Shop (stationary) [b]	Auto mechanic	**0.118**	**0.023**	**<0.0001**	<LOD	<LOD	NA
	Lab analyst	**0.114**	**0.030**	**<0.0001**	<LOD	<LOD	NA
	Warehouse technician	0.018	0.041	0.3243	0.004	<LOD	NA
Office/ Control room	Control room operator	0.021	0.017	0.5269	<LOD	<LOD	NA
	Office staff	0.009	0.016	0.0546	<LOD	<LOD	NA

Numbers in **boldface** indicate statistically significant differences between eastern and western zone ($P< 0.05$), <LOD indicates all samples containing LOD, and NA indicates the data containing LOD in either one of two zones.

[a] Shop (mobile) refers to those SEGs whose work is more likely done in multiple places in the plants. [b] Shop (stationary) refers to those SEGs whose work is more likely done in a single workplace.

Table 2-3 Summary statistics of total EMP for each SEG measured in all mines (GM unit: particles/cm^3)

Department	SEG	Northshore		Hibbtac		Utac		Keetac		Minntac		Minorca	
		GM	GSD	GM	GSD	GM	GSD	GM	GSD	GM	GSD	GM	GSD
Mining	Basin operator	.	.	0.017	1.96	0.089	2.40	0.014	1.94	0.028	1.88	.	.
	Mining operator 1	0.054	1.96	0.010	1.25	0.013	1.80	0.012	1.81	0.019	1.78	0.012	1.76
	Mining operator 2	0.057	3.14	0.011	1.27	0.030	2.04	0.025	2.73	0.018	1.95	0.010	1.14
	Rail road	0.054	2.53	.	.	.	.	.	.	.	.	.	.
Crushing	Crusher maintenance	0.131	2.70	0.013	1.95	0.026	2.68	0.025	1.85	0.068	2.29	0.027	2.13
	Crusher operator	0.157	1.95	0.018	2.66	0.015	2.43	0.012	1.91	0.071	1.51	0.027	2.13
	Operating technician	0.140	3.53	.	.	0.012	1.67	.	.	.	.	.	.
Concentrating	Concentrator maintenance	0.180	1.71	0.013	1.77	0.060	2.01	0.093	1.42	0.048	2.16	0.042	1.57
	Concentrator operator	0.116	3.06	0.009	1.08	0.013	2.27	0.016	2.07	0.029	1.69	0.035	1.50
Pelletizing	Balling drum operator	0.042	1.90	.	.	0.015	2.79	0.119	1.77	0.063	2.23	0.015	1.86
	Dock man	0.155	2.12	0.010	1.48	0.024	3.28	0.049	4.48	0.187	1.88	0.014	2.04
	Furnace operator	0.056	1.94	.	.	0.010	1.45	0.091	1.65	0.028	1.64	0.012	1.49
	Pelletizing maintenance	0.061	1.56	0.012	1.45	0.057	2.14	0.077	2.00	0.103	2.57	0.016	1.98
	Pelletizing operator	0.094	1.77	0.050	2.32	0.009	1.39	0.140	1.72	0.104	2.21	0.024	1.52
Shop(mobile)[a]	Boiler technician	.	.	.	.	0.020	2.63	.	.	0.034	2.39	.	.
	Carpenter	.	.	0.023	2.50	.	.	0.100	1.26	0.054	1.65	.	.
	Electrician	0.279	1.62	0.036	1.71	0.057	3.06	0.029	2.04	0.104	2.51	0.017	2.06
	Lubricate technician	0.104	2.43	.	.	0.025	2.40	.	.	.	.	0.026	1.72
	Maintenance technician	0.036	2.04	0.012	1.80	0.054	2.66	0.041	2.15	0.019	1.85	0.025	2.32
	Pipefitter/Plumber	.	.	.	.	0.042	2.28	.	.	0.039	1.91	.	.
	Repairman	.	.	0.050	1.85	.	.	.	.	0.070	2.07	0.023	2.81
	Supervisor	0.034	3.70	0.010	1.17	0.011	1.29	0.041	1.64	0.073	2.22	0.011	1.39
Shop (stationary)[b]	Auto mechanic	0.086	2.34	0.015	2.85	0.009	1.48	0.020	2.12	0.019	2.49	0.015	1.49
	Lab analyst	0.093	2.23	0.009	1.14	0.033	1.28	0.026	2.41	0.025	2.06	0.018	1.91
	Warehouse technician	0.015	1.82	0.011	1.46	0.027	2.48	0.011	1.51	0.053	2.86	0.013	1.71
Office/ Control room	Control room operator	0.008	1.65	0.011	1.34	0.010	1.13	0.011	1.42	0.018	2.15	0.010	1.10
	Office staff	0.016	2.18	0.012	1.37	0.011	1.29	0.012	1.36	0.012	1.45	0.021	2.35

[a] Shop (mobile) refers to those SEGs whose work is more likely done in multiple places in the plants.

[b] Shop (stationary) refers to those SEGs whose work is more likely done in a single workplace.

Table 2-4 Summary statistics of amphibole EMP for each SEG measured in eastern zone (GM unit: particles/cm^3)

Department	SEG	GM	GSD
Mining	Basin operator	.	.
	Mining operator 1	<LOD	<LOD
	Mining operator 2	0.003	2.62
	Rail road	<LOD	<LOD
Crushing	Crusher maintenance	0.019	2.11
	Crusher operator	0.023	2.07
	Operating technician	0.037	4.02
Concentrating	Concentrator maintenance	0.025	1.96
	Concentrator operator	0.015	3.11
Pelletizing	Balling drum operator	0.009	1.71
	Dock man	0.014	2.18
	Furnace operator	0.013	2.01
	Pelletizing maintenance	<LOD	<LOD
	Pelletizing operator	0.012	1.66
Shop(mobile)[a]	Boiler technician	.	.
	Carpenter	.	.
	Electrician	0.041	2.95
	Lubricate technician	0.012	2.27
	Maintenance technician	<LOD	<LOD
	Pipefitter/Plumber	.	.
	Repairman	.	.
	Supervisor	0.007	3.26
Shop (stationary)[b]	Auto mechanic	<LOD	<LOD
	Lab analyst	<LOD	<LOD
	Warehouse technician	0.004	1.60
Office/ Control room	Control room operator	<LOD	<LOD
	Office staff	<LOD	<LOD

[a] Shop (mobile) refers to those SEGs whose work is more likely done in multiple places in the plants.

[b] Shop (stationary) refers to those SEGs whose work is more likely done in a single workplace.

Table 2-5 Between-SEGs, between-worker, and within-worker variance components by mine and zone for total EMP

Zone	**Mine**	**Subject**	**Sample**	**BG**		**BW**		**WW**		**ε** [b]	**η** [c]
				S^2_{BG}	**%**	S^2_{BW}	**%**	S^2_{WW}	**%**		
East	Northshore	56	266	0.097	39.65	0.034	13.91	0.113	46.44	0.740	0.77
West	All	176	1014	0.021	8.69	0.142	58.24	0.081	33.07	0.130	0.36
	Hibbtac	34	197	0.041	33.85	0.020	16.70	0.060	49.45	0.670	0.75
	Utac	38	218	0.038	19.17	0.076	37.76	0.086	43.07	0.337	0.53
	Keetac	34	203	0.120	53.24	0.019	8.30	0.087	38.46	0.865	0.82
	Minntac	48	267	0.054	28.85	0.069	36.80	0.065	34.36	0.439	0.48
	Minorca	22	129	0.000 [a]	0.00	0.204	76.39	0.063	23.61	0.000 [a]	0.24

[a] Assuming that the use of the PROC NESTED model is appropriate, the negative variance components were treated as zero.

[b] ε: contrast

[c] η: homogeneity

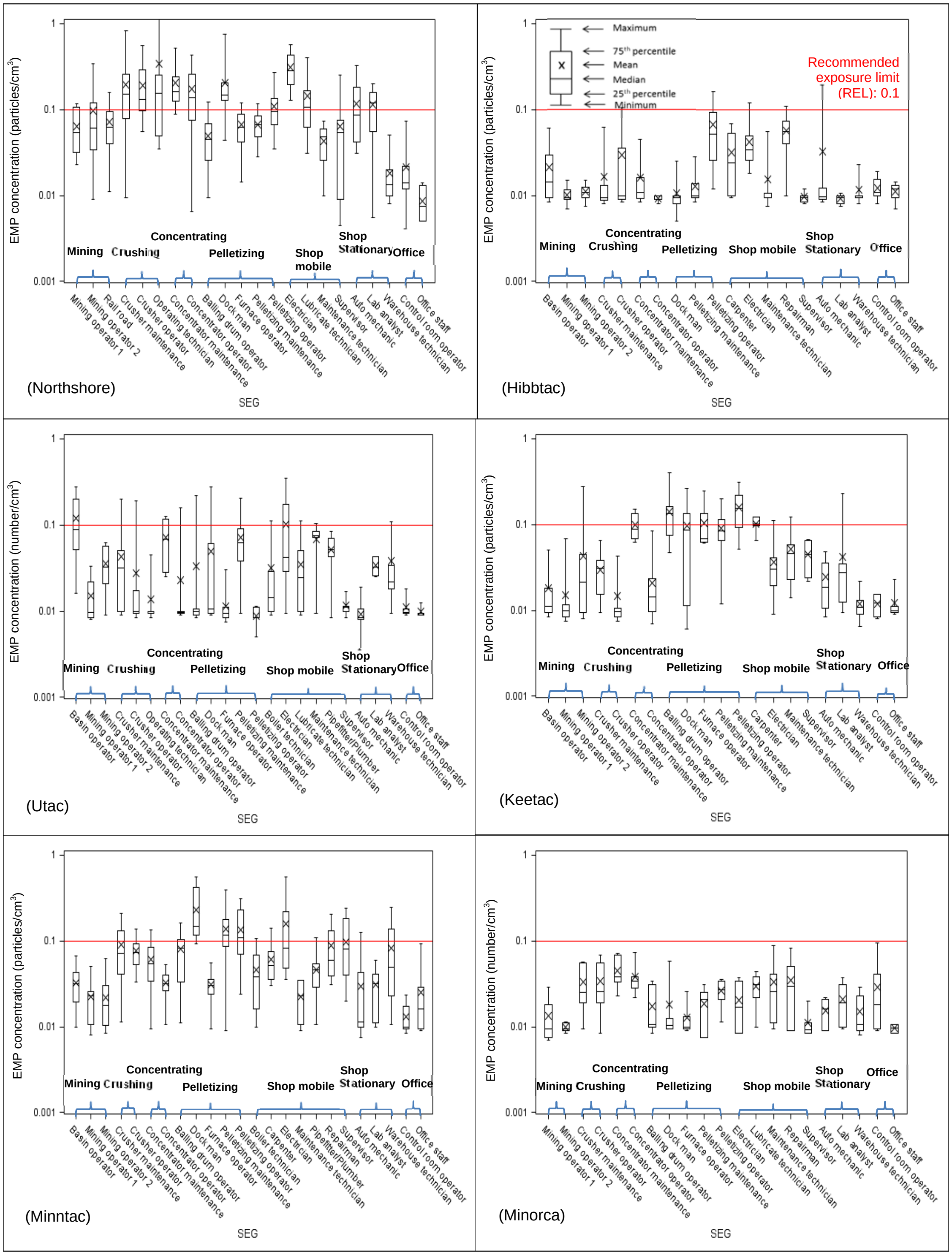

Figure 2-1 Box plot of total EMP for each SEG in all mines (the horizontal line indicates the NIOSH REL for EMP = 0.1 particles/cm^3)

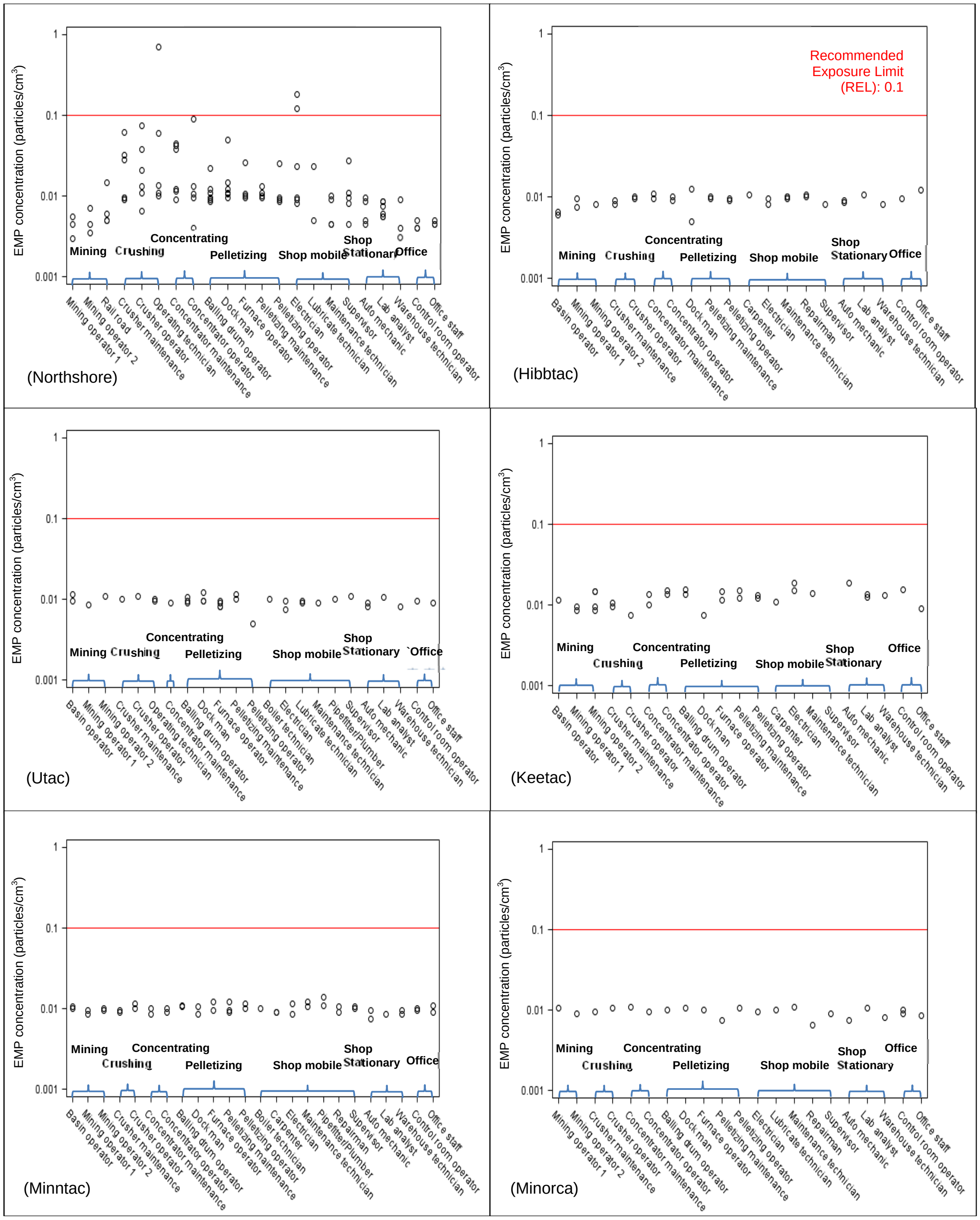

Figure 2-2 Scatter plot of amphibole EMP for each SEG in all mines (the horizontal line indicates the NIOSH REL for EMP = 0.1 particles/cm^3)

Chapter 3 Relationship Between Various Exposure Definitions For Elongate Mineral Particles (EMP)

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 (e.g. Eastern Research Group, 2003), 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 any mineral particle with a minimum aspect ratio of 3:1 that is of inhalable, thoracic, or respirable size (NIOSH, 2011).

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 (Wylie *et al.*, 1993; Addison & McConnell, 2008). EMP dimensions are important for several reasons: (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 (Baron, 2003). Other EMP characteristics related to pathogenic and toxic health effects include durability, habit, strength, chemical composition, morphology, surface charge, and activity (Wylie, 1990; Baron, 2003; Mossman, 2008).

Minnesota counties in the vicinity of taconite mining operations have been found to have elevated age-adjusted rates for mesothelioma (Minnesota taconite workers health study, 2013; Case *et al*., 2011), a disease thought to be associated with exposure to asbestiform EMP. Studies measuring EMP dimensions (length and width) have been relatively scarce (Wilson *et al*., 2008). However, due to the characteristics of the ore body and the taconite process, non-asbestiform EMP is a potentially major source of exposure. Therefore, adverse health effects may be linked not only to asbestiform but also to non-asbestiform EMP. To date, no single study has conducted an extensive assessment of the relationship between non-asbestiform EMP dimensions and adverse health effects in taconite mining industry. In general, non-asbestiform cleavage fragments have not been thought to have high potential for disease (Berry and Gibbs, 2008; Gamble and Gibbs, 2008; Mossman, 2008).

In the taconite mining industry, cleavage fragments refer to the fractured mineral EMP created during the crushing and fracturing process rather than to naturally occurring EMP (NIOSH, 2011). Because no standard definition exists, distinguishing cleavage fragments from asbestiform EMP is challenging (Axten & Foster, 2008). Even if a given EMP counting criterion is met, standard methods such as phase contrast microscopy (NIOSH method 7400, NIOSH, 1994a) cannot distinguish between non-asbestiform cleavage fragments and asbestiform EMP. Researchers have characterized cleavage fragments using biological experiments, mineralogical properties, and chemical compositions, as well as EMP dimension analysis (Wylie, 1990). They have found that non-asbestiform cleavage fragments are inactive in *in vitro* bioassays and that they have less strength and flexibility in morphologic analyses (Mossman, 2008). A linear relationship has been found to exist between the width and length of cleavage fragments, while no such relation exists in asbestiform EMP (Bailey *et al*., 2003). The term "amphibole EMP" refers to a subset of double chain silicate minerals that can be asbestiform (crocidolite, amosite, anthophyllite asbestiform, tremolite asbestiform, and actinolite asbestiform) or non-asbestiform (riebeckite, cummingtonite-grunerite, anthophyllite, tremolite, and actinolite) (Bailey *et al*., 2003; (NIOSH, 2011). Addison & McConnell (2008) concluded that cleavage fragments are likely thicker than asbestiform EMP and have a diamond-shaped cross-section, while asbestiform EMP are likely to be longer and more flexible.

Because of the difficulty of distinguishing between asbestiform and non-asbestiform EMP, the relationship between non-asbestiform EMP and adverse health effects is still not well understood (NIOSH, 2011). Although the chemical composition of asbestiform and non-asbestiform EMP can be the same, they differ in their habit or morphology (Langer *et al*., 1979; Berndt & Brice, 2008). Asbestiform EMP are "polyfilamentous" whereas non-asbestiform EMP display a "multidirectional" pattern (Bailey *et al*., 2003). The relationship between the size of these EMP and the development of carcinogenic lung disease has been difficult to establish despite many animal and human studies.

No consensus exists regarding the most health-relevant, dimension-based exposure metric for EMP. Stanton *et al*. (1981) hypothesized that the carcinogenicity of EMP is related to dimension and durability and less with mineral type. Carcinogenicity was ascribed to EMP with a length greater than 8 µm and a diameter less than 0.25 µm. Subsequently, Lippmann (1990) suggested that lung cancer was associated with asbestos EMP longer than 10 µm with a diameter greater than 0.15 µm, while mesothelioma was associated with asbestos EMP longer than 5 µm with a diameter less than 0.1µm. An analysis by Berman *et al*. (1995) 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 (Eastern Research Group, 2003) concluded that there is a weight of evidence that asbestos and synthetic vitreous fibers shorter than 5 µm are unlikely to cause cancer in humans. Chatfield (2009) 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 (OSHA, 1998) 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. Dodson et al. (2003) concluded that asbestos EMP of all lengths induce pathological responses and cautioned against ignoring EMP shorter than 5 µm since they constitute most of the EMP exposure. Suzuki et al. (2005) 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. Using a TEM analysis of chrysotile fibers, Dement et al. (2008) showed that exposures to 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 reinforced a previous conclusion that even though the traditional counting method includes only EMP longer than 5 μm, shorter EMP that contribute to health-relevant work exposures may be important (Dement *et al.*,1983).

Table 3-1 summarizes the dimension-based EMP definitions that will be used in this paper. In Figure 3-1, the same four size-based EMP definitions are compared using a typical sample collected for this study. Each graph shows 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 (Quinn *et al*., 2000).

The aims of this study were to: 1) determine the size distribution by length and width of EMP as measured in conjunction with the MOUDI collection method 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 from the standard NIOSH Methods 7400/7402-based EMP definition 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 diseases such as mesothelioma, lung cancer, and non-malignant respiratory disease.

METHODS

Sampling sites

The Mesabi Iron Range lies in northeastern Minnesota and extends 200 km from Babbitt in the east to Grand Rapids in the west. The mineralogy of the range changes from east to west with distinct metamorphic mineralogical zones. In the eastern zone, the iron ore contains amphibole (cummingtonite-grunerite series and ferroactinolite-tremolite series), whereas the ore in the western zone contains predominantly phyllosilicates such as minnesotaite, greenalite, and stilpnomelane that are not regulated as asbestiform or amphibole EMP (McSwiggen and Morey, 2008; Zanko *et al*., 2008).

Based on the distinct 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 (Northshore) and five mines in the western zone (Hibbtac, Utac, Keetac, Minntac, and Minorca). The first criterion for the sampling design was to determine locations for area sampling representing the eastern and western regions of the Mesabi Iron Range that generally corresponded to the similar exposure groups (SEGs), the basis for personal sampling (Hwang *et al.*, 2013). The term location is used to refer to physical places where the area measurements were obtained for each SEG. Table 3-2 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 rotating cascade impactor (Model 125R MOUDI-II, MSP Co., Shoreview, MN, USA). 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 approximately 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, morphology, and mineral type for each EMP regardless of the EMP dimension (ISO, 1999). We chose ISO 13794 because it immerses the whole filter in water and re-filters the particles suspended in the water through a secondary filter. Thus, the particles are evenly distributed across the surface of the secondary filter. 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 a laboratory (EMSL Analytical Inc., Minneapolis, MN, USA) accredited by the American Industrial Hygiene Association.

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 (NIOSH, 1994b), 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.

Data management

The impactor data were used to determine the distribution of EMP sizes. Based on these data, relationships between various dimension-based EMP exposure metrics were derived. 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.

Chrysotile, also known as a common commercial asbestiform EMP (Pigg, 1994), was found at locations corresponding to the *Maintenance technician, Lab analyst*, and *Crusher maintenance* SEGs in Northshore, *Balling drum operator* SEG in Keetac, and *Pelletizing operator* SEG in Minntac. Since only five chrysotile 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). Both total and amphibole EMP ≥ 0.3 μm in length and ≥ 3 in aspect ratio were counted.

The number of grid openings observed for each stage sample was recorded to allow for later scaling for the fraction of the filter analyzed. For each sample, grid openings were analyzed for EMP until the 100th particle was counted or the required analytical sensitivity was achieved, whichever occurred first (ISO, 1999). 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 (e.g., *Concentrator maintenance*/*Repairman* in Hibbtac and *Crusher operator/ Maintenance technician* in Keetac).

Because we analyzed only selected stages (3, 5, 7, 9, 11), the non-analyzed stages (4, 6, 8, 10) 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.

ANOVA test for loading on stages of the MOUDI impactor

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, the Tukey's studentized range was used for pair-wise comparisons of the log of the number concentration of EMP collected by each stage.

Relationship between NIOSH 7400 and other size-based definitions

The associations between NIOSH 7400 and various total EMP concentration metrics such as Suzuki, Chatfield, and Cleavage fragment were assessed (a) using a simple linear regression based on the log-transformed exposure concentrations across all locations in each zone (Equation 3-1) 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 3-2). 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:

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

(Equation 3- 1)

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

(Equation 3- 2)

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

$C_{\text{NIOSH EMP}}$, 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.

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

$$C_{\text{Definition}} = a_i C_{\text{NIOSH EMP}} \text{ of the } i^{\text{th}} \text{ location} \qquad \text{(Equation 3- 3)}$$

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-values: 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 3-2 shows the combined EMP concentration of stages 3 through 11 by location in the two zones, with a reference line indicating the NIOSH recommended exposure limit (REL) of 0.1 particles/cm^3 for EMP. In the case where more than one sample was available for a location, the EMP concentration was averaged. 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. As shown in Figure 3-2, the concentrations of total and amphibole EMP in the eastern zone are markedly higher than that in the western zone. 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). The amphibole EMP concentration levels for all locations in the western zone were also close to zero.

The differences between total and amphibole EMP by location are also compared in Figure 3-2. The upper limit of the y-axis scale (concentration) for amphibole EMP is half that for total EMP, as the concentration of amphibole EMP is generally smaller than that of total EMP. Most interestingly, the concentration patterns by location for total and amphibole EMP were similar, except for a few locations. 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 3-3 and Figure 3-4, respectively. 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 across 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. In particular, amphibole EMP in the western zone were only detected in the locations corresponding to the *Concentrator operator* (0.0002 particles/cm^3) and the *Maintenance technician* (0.0008 particles/cm^3) SEGs for the NIOSH and Chatfield definitions, respectively. Figure 3-3 and Figure 3-4 indicate that the relative magnitudes of the concentrations of total and amphibole EMP at different locations were similar using the various 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 3-5 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 3-3 and Figure 3-4. 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 3-6. 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 was derived to relate EMP log-concentrations based on NIOSH 7400 to the log-concentrations based on each of the size-based EMP definitions for total and amphibole EMP (Table 3-3). 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 regression model 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 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 3-4 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 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 3-7 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 length/width categories (5 length categories x 5 width categories) 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

Similar exposure patterns were observed based on the different EMP size definitions, consistent with the high degree of correlation between the exposure levels. Therefore, dimensional definitions probably do not result in different dose-response relationships in epidemiological analyses. 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.

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 (Figure 3-2), 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 of Cleavage fragment concentrations are clearly higher in the mining processes, consistent with the generating of cleavage fragments in the mining and processing of ore (Figure 3-3 and Figure 3-4). 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

In this study, we found that relative magnitudes of concentrations across locations were similar for the different dimension-based EMP definitions in the eastern zone (Figure 3-3 and Figure 3-4), 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 (Figure 3-5). The high correlation among these EMP concentration metrics will limit the ability of epidemiological analyses in determining relative differences of effect by different EMP metrics. In other words, the effects of correlated relationships are not identifiable. Quinn *et al*. (2000) 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 would be a loss of power using another EMP definition. Dement *et al*. (2008) 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*. (2008) 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. 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. Therefore, the area measurements obtained using the MOUDI provide a unique opportunity for investigating the relationship between the various EMP metrics.

Quinn *et al*. (2000) and Dement *et al*. (2008) 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. For total EMP, the ratios in the western zone had a wider range than those in the eastern zone because of lower EMP concentration by the NIOSH definition in the western zone. The ratios for total EMP using the Cleavage fragment definition in the western zone had a wide range. The high ratios and the wide range indicate that cleavage fragments are 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. The small ratios and narrow range indicate that relatively few Chatfield EMP exist in the eastern 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 much 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 in Figure 3-7. This size metric referred to short EMP and is a subset of Suzuki and Cleavage fragment definitions. Dodson *et al*. (2003) 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, as shown in Figure 3-7.

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. While it would have been preferable to have analyzed the personal samples to estimate exposures based on the various EMP definitions, this was not feasible in this project. Furthermore, the area samples from stages 1, 2, 12, 13 were not analyzed, 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 (EDXA) and crystalline structure by selected area electron diffraction (SAED). 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 analog). 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 other analytical methods (e.g., scanning electron microscopy). However, these methods have not been extensively validated. While we expect that most of amphibole EMP are likely non-asbestiform in the Iron Range, this is based on past studies (e.g., Wilson *et al.*, 2008; Zanko *et al.*, 2008).

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 the 1-3 µm in length and 0.2 - 0.5 µm in width size, 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 (i.e., relative levels across SEGs) 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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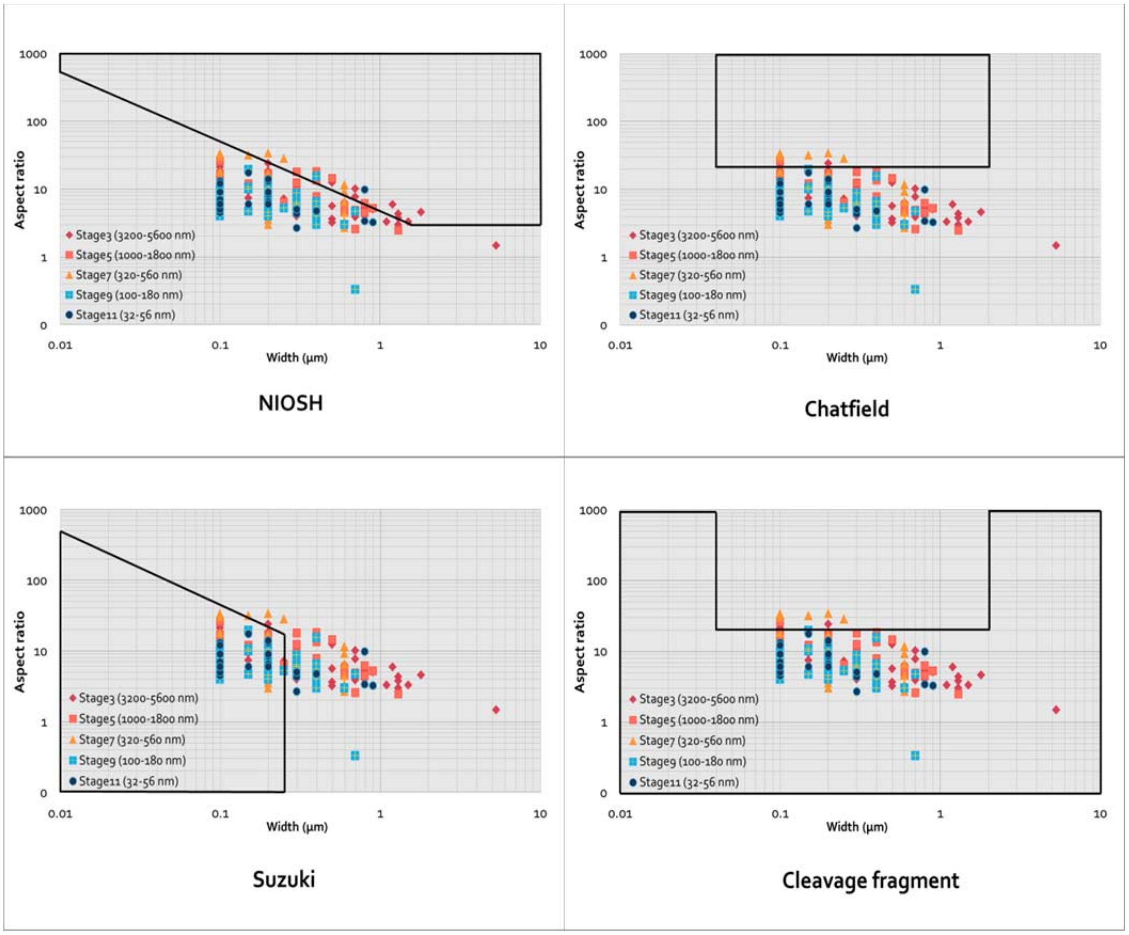
Table 3-1 Characteristics of four dimension-based EMP metrics

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

[a] PCM: Phase contrast microscopy
TEM: Transmission electron microscopy

Table 3-2 Number of MOUDI samples and locations by mine

Mine	# MOUDI samples	# Locations
Northshore	23	17
Hibbtac	14	14
Utac	20	16
Keetac	12	11
Minntac	15	15
Minorca	8	8



NIOSH

Chatfield

Suzuki

Cleavage fragment

Figure 3-1 Comparison of total EMP count for four dimension-based EMP exposure metrics by MOUDI impactor stage for the Crusher Maintenance location in Northshore. The black boxes indicates the dimension-based EMP exposure metrics based on each definitions.

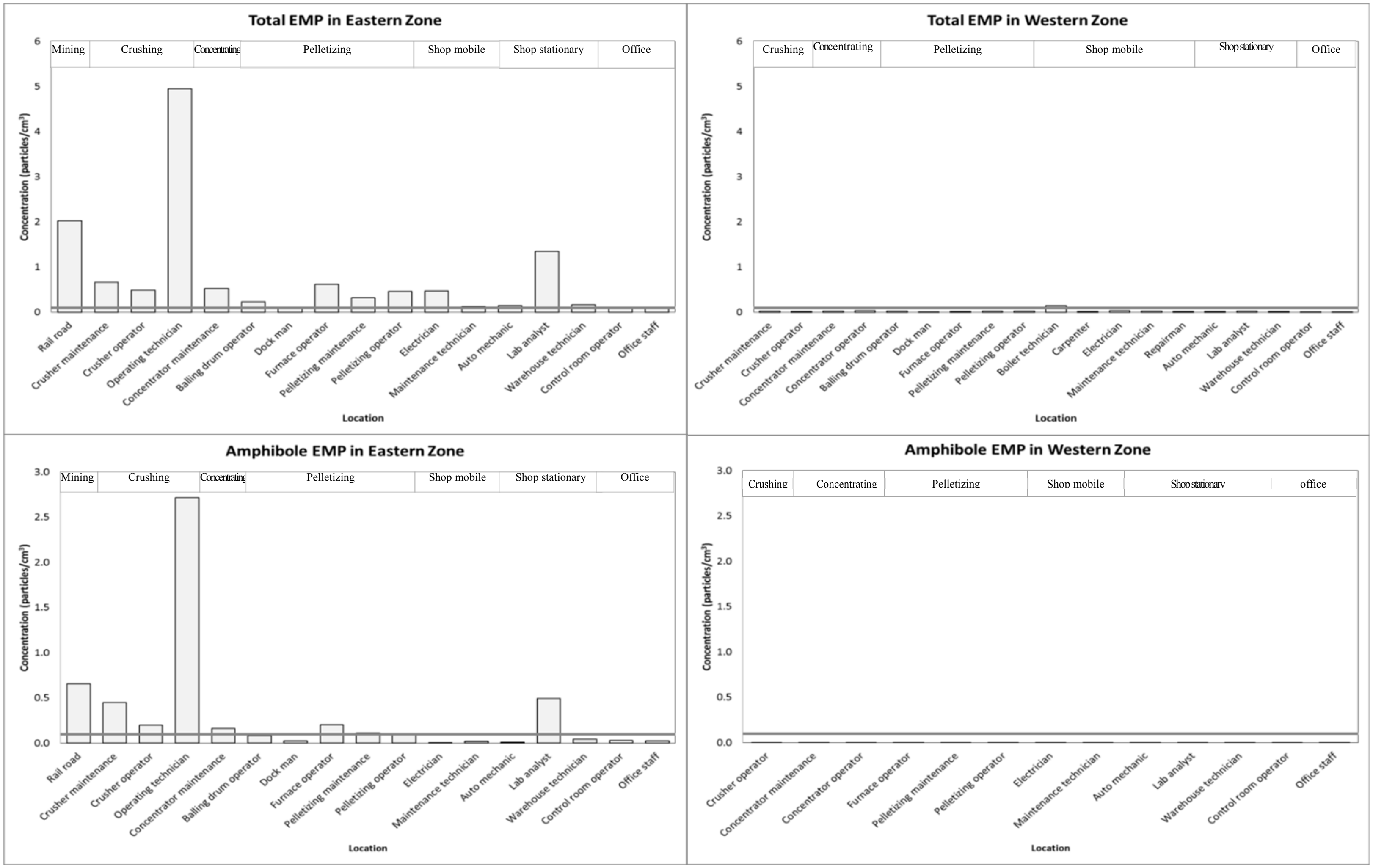


Figure 3-2 Total and amphibole EMP concentration by location in two zones (line indicates the NIOSH REL for NIOSH 7400/7402 EMP= 0.1 particles/cm^3)

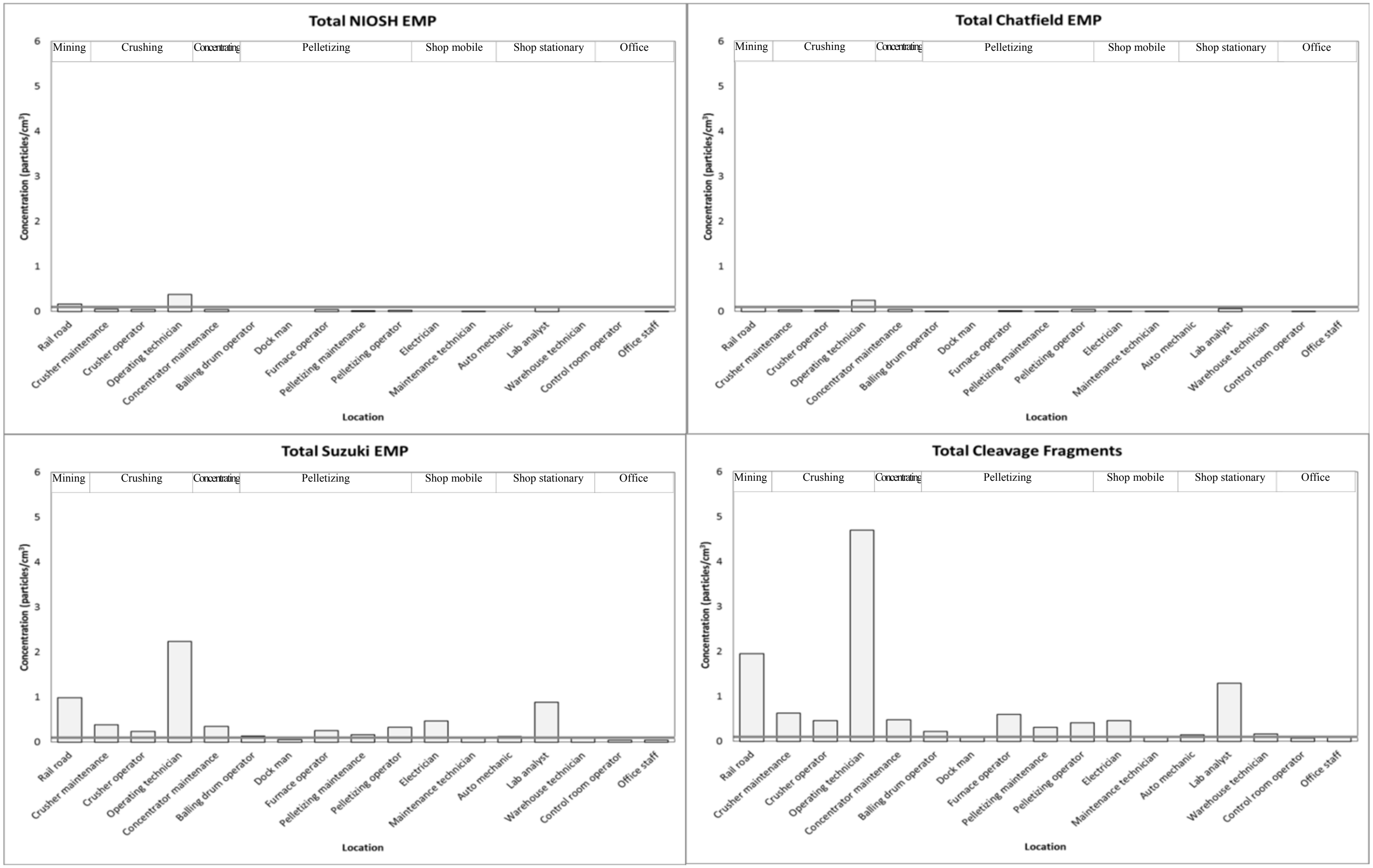
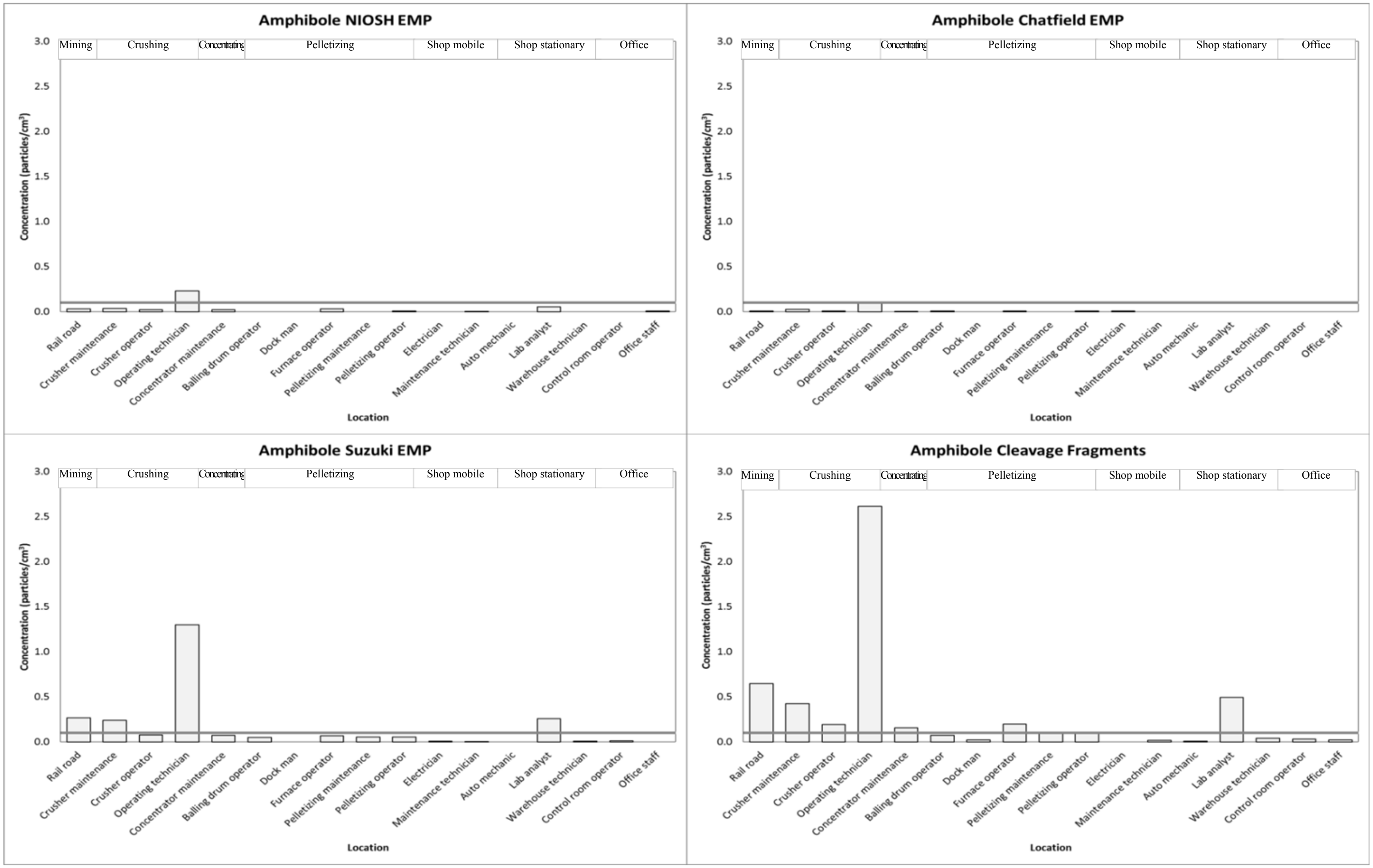


Figure 3-3 Total EMP concentration by size-based definitions in eastern zone (line indicates the NIOSH REL for NIOSH 7400 EMP = 0.1 particles/cm^3)


Amphibole NIOSH EMP

Amphibole Chatfield EMP

Amphibole Suzuki EMP

Amphibole Cleavage Fragments

Figure 3-4 Amphibole EMP concentration by size-based definitions in eastern zone (line indicates the NIOSH REL for NIOSH 7400/7402 EMP = 0.1 particles/cm^3)

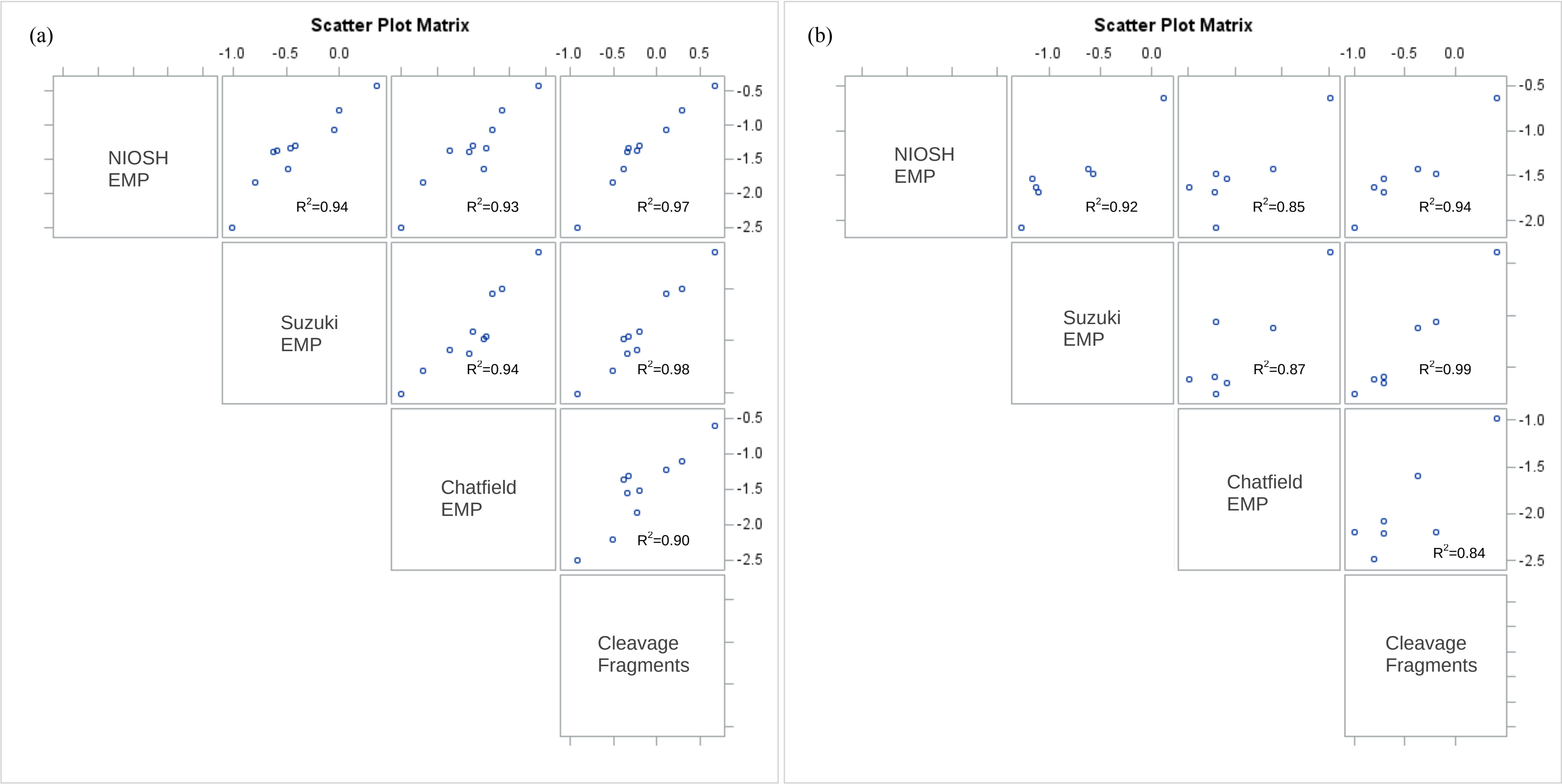


Figure 3-5 Coefficients of determination between (a) total and (b) amphibole EMP definitions in the eastern zone

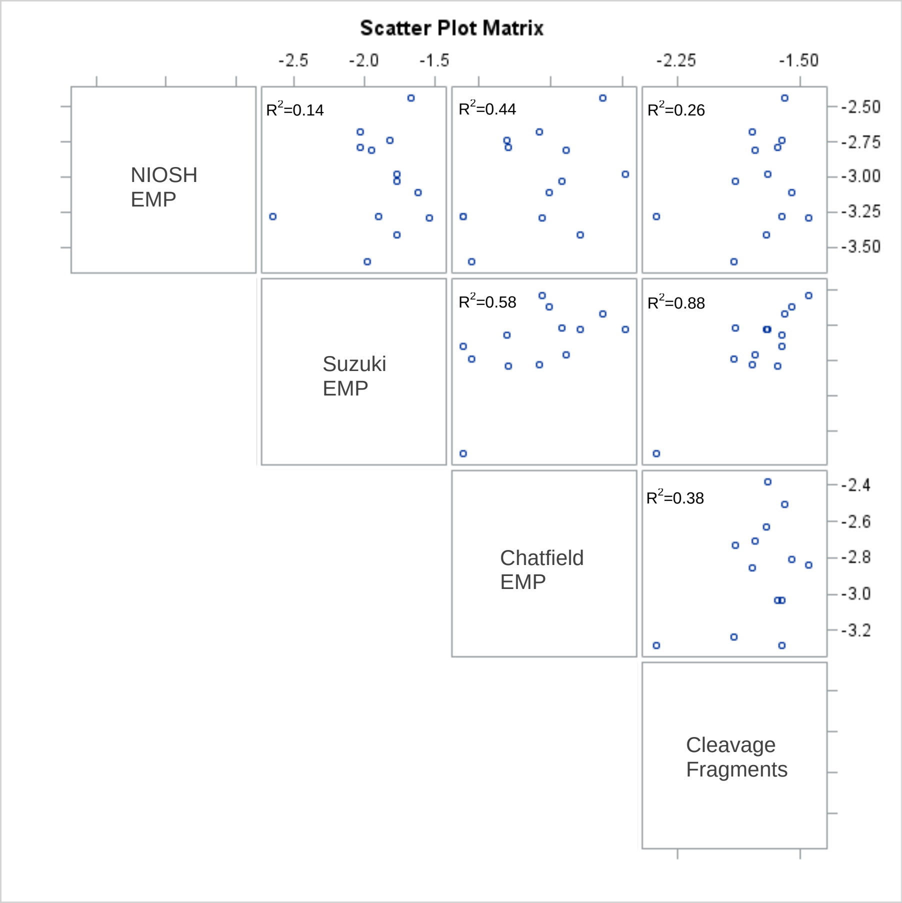


Figure 3-6 Coefficients of determination between total EMP definitions in the western zone

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

$C_{\mathbf{Definition}}$[a]	**Total EMP**		**Amphibole EMP**	
	East	**West**[b]	**East**	**West**[c]
$C_{\text{Suzuki EMP}}$	$3.83C_{\text{NIOSH EMP}}^{0.744}$	$13.0C_{\text{NIOSH EMP}}$	$6.40C_{\text{NIOSH EMP}}^{1.089}$	NA[d]
$C_{\text{Chatfield EMP}}$	$0.501C_{\text{NIOSH EMP}}^{0.894}$	$1.46C_{\text{NIOSH EMP}}$	$0.328C_{\text{NIOSH EMP}}^{1.011}$	NA
$C_{\text{Cleavage fragment}}$	$8.28C_{\text{NIOSH EMP}}^{0.819}$	$14.9C_{\text{NIOSH EMP}}$	$11.9C_{\text{NIOSH EMP}}^{1.062}$	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] Concentration of amphibole by NIOSH are zero in the western zone except at the *Concentrator operator* location.
[d] Not applicable (NA) due to no regression parameters are estimated for amphibole EMP in the western zone.

Table 3-4 Ratios of EMP concentrations based Suzuki, Chatfield, and Cleavage Fragment definitions to EMP concentration

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	—	—	—
	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.

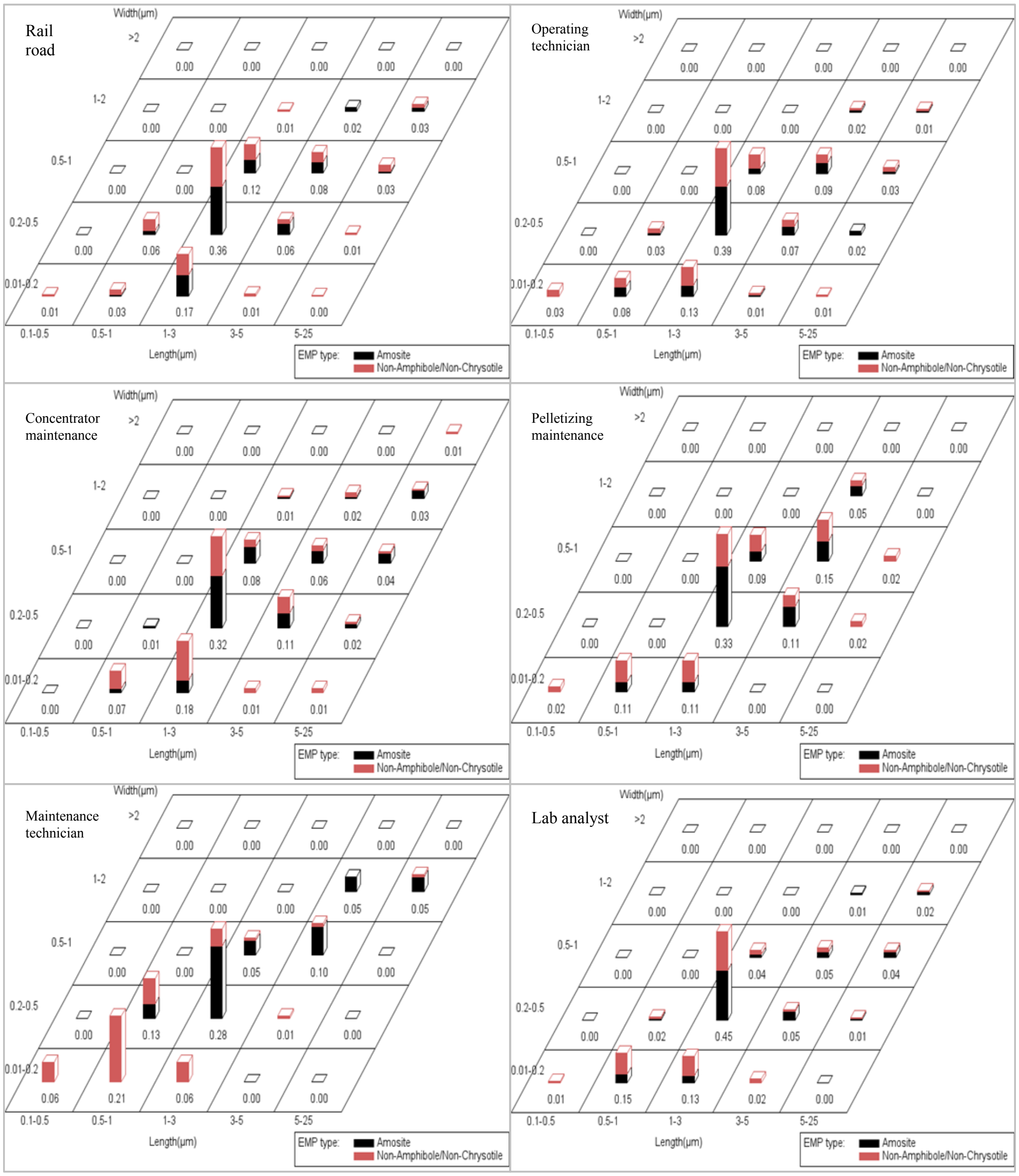


Figure 3-7 Fraction of EMP size distribution by location in eastern zone

Chapter 4 Reconstructing Historical Exposures to Elongate Mineral Particles (EMP) in the Taconite Mining Industry for 1955 – 2010

INTRODUCTION

The Mesabi Iron Range in northeastern Minnesota contains low grade iron ore called taconite, which is mined and milled to make iron pellets. Reports of potential health issues related to taconite mining, especially to airborne taconite dust, began to appear in the 1980s. Initial reports concerned with pneumoconiosis (Clark *et al*., 1980; Higgins *et al*., 1983) were followed by mortality assessments in specific mining companies (Higgins *et al*., 1981; Cooper *et al*., 1988; Cooper *et al*., 1992). These studies were inconclusive, exhibiting limited statistical power and providing minimal information regarding worker exposures. Sheehy (1986) collected earlier EMP data spanning 1973-1977 in his study. However, these data were collected using area sampling and were not included in our analysis. Subsequently, the Minnesota Department of Health (MDH) conducted studies to understand the taconite workers' health in response to the rise in cases of mesothelioma between 1988 and 1996 under the Minnesota Cancer Surveillance System (MCSS, 1999). They found 17 workers who were diagnosed with mesothelioma during this period (MDH, 2003). Since 2003, a total of 80 additional miners in the Mesabi Iron Range have been diagnosed with mesothelioma. Minnesota Department of Health (MDH, 2007) reported that the mesothelioma rate for males in northeastern Minnesota was twice as high as the expected rate in the general population of the rest of Minnesota. This fact supports the possibility that exposure to taconite elongate mineral particles (EMP) could be a cause of the mesothelioma; however, it is still unclear that the excess of mesothelioma is caused by the taconite ore itself.

Measuring “asbestos” in EMP collected by air sampling is difficult due to the complex mineralogy and variations in EMP size, as well as different analytical techniques, sample collection strategies, sample preparations, counting rules (ASTDR, 2008; Meeker *et al*., 2003), and definitions of what constitutes a health-relevant EMP (Lippmann, 1988). Some researchers have attributed carcinogenicity to asbestiform EMP with a length greater than 8 μm and a diameter less than 0.25 μm (Stanton *et al*., 1981), while others have specified shorter EMP (Suzuki, 2005). Because the traditional counting method only detects EMP greater than 5 μm in length (NIOSH, 1994a; NIOSH 1994b), shorter EMP, which may contribute substantially to work exposure, have not been included (Dement *et al*., 1983). Recent but controversial hypotheses speculate that non-asbestiform EMP based on a variety of size-based (length and width) definitions may also be contributing causes of mesothelioma and lung cancer (Wilson *et al*., 2008). Chatfield (2009) proposed a protocol that defined EMP with widths of 0.04-1.5 μm and aspect ratios of 20-1000 as asbestiform and EMP with aspect ratios ≤ 20 as non-asbestiform, including cleavage fragments. OSHA (1998) also defines cleavage fragments as having aspect ratios smaller than 20:1. In many industries including taconite mining and milling, cleavage fragments refer to the fractured mineral EMP created during the crushing and fracturing process rather than to naturally occurring EMP (NIOSH, 2011). Comprehensive EMP samples from the Mesabi Iron Range have never been fully investigated based on dimensions. In fact, no previous studies have examined the exposures to EMP in the taconite industry.

Several epidemiological studies have looked for associations between non-asbestiform EMP and mesothelioma. Ludwig *et al*. (1981) studied the Homestake gold mine in South Dakota where exposures to longer (>5 μm) non-asbestiform EMP were cummingtonite-grunerite, which is also the primary type of EMP in taconite mining operations in the Iron Range. However, the majority of EMP at Homestake gold mine were short (<5 μm) which is also similar to findings in this study. McDonald *et al*. (1978) observed that the excess of mesothelioma in this population of Homestake was small. Several research groups investigated the exposure to talc in upstate of New York (Enterline & Henderson, 1987; Hull *et al*., 2002; Vianna *et al*., 1981). The biopsy samples from the talc workers did not yield asbestos in these studies. However, an excess of mesothelioma was observed.

The current research was carried out as part of a larger epidemiological study investigating the relationship between exposures to EMP during the mining and processing of taconite ore in the Mesabi Iron Range in northeastern Minnesota and the development of diseases such as mesothelioma, lung cancer, and non-malignant respiratory disease. Specifically, to understand the relationship between various size-based EMP exposure and mesothelioma, we developed a methodology based on area measurements, advanced instrumentation and analytical methods that classify EMP by size, and statistical analyses. This methodology can be used to calculate concentrations of

EMP based on different dimensions, data that can be used in epidemiological studies. An ongoing mesothelioma case-control study is comparing the incidence of mesothelioma among taconite workers to that among other groups in the cohort. Each mesothelioma case is matched to four worker controls in this cohort.

This study is the first to reconstruct exposure levels to EMP for an epidemiological mesothelioma case-control study in the taconite processing industry. In particular, no previous studies have characterized the exposures to either total/amphibole EMP based on the NIOSH definition or other size-based EMP definitions including cleavage fragments in this industry. Here "total EMP" refers to EMP irrespective of mineralogical composition, while "amphibole EMP" refers to EMP that belong to a subset of double chain silicate minerals that can be asbestiform (crocidolite, amosite, anthophyllite asbestiform, tremolite asbestiform, and actinolite asbestiform) or non-asbestiform (riebeckite, cummingtonite-grunerite, anthophyllite, tremolite, and actinolite) (Bailey *et al.*, 2003; (NIOSH, 2011). Our study is the first major effort to systematically understand these relationships using a unified, large dataset. Thus, our research on exposure assessment addresses several unanswered questions regarding the health effects of taconite mining on workers. The methodology for obtaining relationships between various size-based exposure metrics for EMP may be useful to other researchers in environmental and occupational health studies.

This paper has the following specific aims: (1) develop a matrix of exposure levels as a function of SEG, mine, and year using various size-based EMP definitions for an epidemiological study; (2) to compute cumulative EMP exposure levels for each worker.

METHODS

Description of database

Construction of historical exposure database

Retrospective exposure data were extracted from two sources: (a) the Mine Data Retrieval System maintained by the Mine Safety and Health Administration (MSHA, 2013), (b) the internal databases of two of the currently operating taconite mining companies in the Mesabi Iron Range (U.S. Steel, and Cliffs Natural Resources). No personal EMP monitoring results were available in the Arcelor Mittal database (the third currently operating company). The MSHA-mine data retrieval system is a web-based database that includes personal exposure-sampling results including EMP with length > 5µm and aspect ratio greater than 3.0. The EMP were measured using NIOSH Method 7400 (NIOSH, 1994a), and are hereafter referred to as NIOSH 7400 EMP. For the purposes of this study, only the personal exposure data for NIOSH 7400 EMP were integrated into the retrospective exposure assessment database. In total, 568 data points for NIOSH 7400 EMP exposure were collected between 1978 and 2007. Data from the companies' internal industrial hygiene monitoring databases were included in the historical database. A total of 96 measurements collected between 1990 and 2009 were obtained from two of the mine databases (N=65 from Minntac and N=31 from Northshore). Forty-six of the data points from Cliffs Natural Resources were area monitoring samples and were, therefore, excluded from our analysis. Table 4-1 shows the number of historical and present-day data points in each SEG by mine.

In total, the historical dataset for taconite EMP consists of approximately 682 data points, and exposure data are missing for many time periods. If the data are broken down by the 28 SEGs, it is clear that most of the data relate to only a few SEGs. For example, the largest number of observations used to reconstruct exposures was 81 for the *Crusher operator* SEG at the Northshore mine.

The database provides data for seven mines - six currently active mines (Northshore, Hibbtac, Utac, Keetac, Minntac, and Minorca) and one inactive mine (LTV). The retrospective EMP data from the two source databases were combined into a master database using Microsoft Office Access 2010.

Present-day data measurements

To assess present-day exposure levels and supplement the sparse historical data, we conducted an assessment of personal exposures across six mines in the Mesabi Iron Range from January 2010 to May 2011. The LTV mine in Hoyt Lakes closed in 2001;

therefore, no present-day measurements are available. We established 28 similar exposure groups (SEGs) and collected personal samples from a subset of workers in each SEG to assess the present-day levels of exposure to EMP (Hwang *et a*l., 2013a). Total numbers of personal samples from present-day exposure assessment dataset were 1280. Thus, including 682 historical data points, a total of nearly 2000 data points were used to reconstruct exposures to NIOSH 7400 EMP.

A substantially higher percentage (65%) of NIOSH 7400 EMP measurements came from the present-day measurements collected during 2010-2011 than from the historical data (35%) which covered the years 1978-2009. Eighteen percent of the present-day NIOSH 7400 EMP values were randomly chosen and analyzed for amphibole EMP using NIOSH 7402; no measurements for amphibole EMP existed in the historical database.

Data treatment

Handling data below the limit of detection

In total, 17 data points were below the limit of detection (LOD) in the historical database, all from Northshore's internal industrial hygiene data, while 463 data points were below the LOD in the present-day measurements. The censored data values were represented by one half of the LOD. The lowest present-day LOD value was 0.006 EMP/cm^3, while the lowest historical LOD value was 0.0005 EMP/cm^3, 12 times lower. The lower LOD in the historical dataset may have been related to longer sampling times. Furthermore, many of the EMP exposure data values in the historical database were entered as zero (N=159). In these cases in which the historical data value was lower than the average of present-day LOD or was recorded as zero, we substituted the average of the LODs from the present-day measurements.

Relationship between total EMP (NIOSH 7400) and amphibole EMP (NIOSH 7402)

Total EMP refers to both amphibole and non-amphibole/non-chrysotile EMP, whereas amphibole EMP refers to cummingtonite-grunerite and actinolite EMP. While present-day total EMP were analyzed using NIOSH 7400, amphibole EMP were analyzed using NIOSH Method 7402 (NIOSH, 1994b). The available historical data are based on NIOSH 7400 analysis. These are consistent with the present-day data which also were analyzed using NIOSH 7400. The combined data set was used for historical exposure reconstruction for total EMP. However, while some of the present-day EMP samples were also analyzed for amphibole EMP using NIOSH 7402, there are no available historical measurements of amphibole EMP. To address this issue, we developed conversion factors (CFs) between total EMP and amphibole EMP concentrations using the present-day data. These CFs were then to be applied to the historical NIOSH 7400 EMP measurements to develop estimates of past exposures to amphibole EMP.

All present-day samples obtained in the western zone had concentrations of amphibole EMP below the LOD. Therefore, CFs could be developed only for SEGs in the eastern zone when at least one sample in a data set for an SEG was above the LOD. Using the NIOSH 7400 and 7402 measurements for each SEG in the eastern zone, we calculated the ratio of the average amphibole EMP concentration and average total EMP concentration (Hwang *et al*., 2013b). Each NIOSH 7400 measurement for that SEG was then multiplied by this ratio to obtain exposure levels to NIOSH 7402 amphibole EMP.

Relationship between NIOSH 7400 EMP exposures and exposures based on other size-based definitions

In addition to the NIOSH 7400 definition of an EMP, many other size-based definitions have been proposed for assessing concentrations of EMP. We chose three additional EMP dimensional definitions: Suzuki, Chatfield, and Cleavage fragment (Hwang *et al*., 2013b). Suzuki *et al*. (2005) 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. Chatfield (2009) 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. He defined everything else with aspect ratio ≤ 20 as non-asbestiform EMP cleavage fragments. The associations between various EMP exposure metrics and the NIOSH 7400 metric for total EMP were assessed using area measurements of all EMP using a MOUDI (Micro Orifice Uniform Deposit Impactor, Model 125R MOUDI-II, MSP Co., Shoreview, MN, USA). The MOUDI is a rotating cascade impactor that was used to obtain the area samples at locations representative of each SEG. The EMP sampled at each location in each mine were counted for each impactor stage using the different dimension-based definitions. The ISO Method 13794, used to analyze the impactor samples, provides details of EMP dimension, structure type, EMP type, and mineral type for each EMP (ISO, 1999). The resolution limit of ISO 13794 is 0.3 µm in length and 0.1 µm in width, so we counted all fibers > 0.3 µm in length with an aspect ratio > 3.

The associations between various EMP exposure metrics and the NIOSH 7400 metric for total EMP were assessed using a simple linear regression of the exposure according to each alternative metric and the NIOSH 7400. A high degree of correlation between these metrics was found in the eastern zone. The regression results are presented in a previous study (Hwang *et al*., 2013b). For total EMP, Cleavage fragments had a markedly higher concentration, followed by Suzuki, in both zones. Chatfield had a slightly lower concentration in the eastern zone, but a higher concentration in the western zone. To relate total EMP concentrations using the Suzuki, Chatfield, or Cleavage fragment definitions ($C_{\mathrm{Definition}}$) to concentrations from NIOSH 7400 (C_{NIOSH}) in the eastern zone, equations of the form

$$C_{\mathrm{Definition}} = a_1 \, C_{\mathrm{NIOSH}}^{\,b} \qquad \text{(Equation 4- 1)}$$

were developed with a_1 and b derived via regression analyses of logged concentration values. In the western zone, equations of the form

$$C_{Definition} = a_2\, C_{NIOSH} \qquad \text{(Equation 4- 2)}$$

were sufficient with a_2 being determined statistically from logged concentration ratios. Because so few amphibole EMP were present in the eastern zone and none in the west, we decided not to calculate amphibole EMP concentrations for the Suzuki, Chatfield, and Cleavage fragment size definitions.

Data analysis

Historical exposure data are unavailable or sparse for many SEGs even though data have been collected over the past 40 years. This sparseness is mainly due to the fact that most of the historical sampling data was not generated for epidemiological purposes. Imputing the missing data requires a variety of statistical techniques as well as knowledge of temporal trends. By combining comprehensive present-day exposure levels with the minimal historical data, we generated exposure matrices that provide estimates of exposure levels. Thus, these exposure matrices provide an estimate of exposure for every SEG as a function of year and mine.

Estimating the retrospective exposure data

After combining the historical and present-day data into a master database, we used two different models to impute the missing EMP exposures for any combination of SEG, year, or mine. The first approach was to use a time-varying quantile and linear regression model (time-varying exposure model) based on the observation that the data fit neither a normal nor lognormal distribution. The second approach assumed a constant exposure over time that was equal to the mean of all historical and present-day EMP data taken together (constant exposure model). All statistical analyses reported here were conducted using SAS Version 9.3 (SAS Institute, Cary, NC, USA).

a. Time-varying exposure model

We estimated exposures to EMP for each year between 1955 and 2010, treating 1955 as year 0. Present-day exposure levels were higher than those expected from the trends in the historical data. Adjusting for whether the measurements were collected presently or historically helped to ensure that the trends over the years were not a byproduct of the more sensitive techniques employed in present-day measurements. Therefore, the quantile regression model included a term for whether the data represented present-day or historical measurements.

In this paper, the quantile regression model was estimated with covariates for present-day data, 28 SEGs (a *Janitor* SEG from the historical database was included) and a linear term for year. The model below was estimated separately for each mine at the 25th, 50th, and 75th percentiles of the estimated EMP exposures (Equation 4- 3)

$Y_{kij} = \log(X_{kij}) = \alpha_0^{(p)} + \sum_{k=1}^{k=28} \alpha_k^{(p)}(k) + \beta_1^{(p)}(i) + \beta_2^{(p)}(j)$ for k= 1, 2, ..., 28th SEG, i = 0, 1, …, 55th year , and j = 0, 1 for adjusted present-day in 2010 (Equation 4- 3)

where log (X_{kij}) = log concentration of NIOSH 7400 definition for the ith year of the kth SEG with the adjusted present-day in 2010 (j = 1), $\alpha_0^{(p)}$= intercept of pth quantile based on Y_{ijk}, $\sum_{k=1}^{k=28} \alpha_k^{(p)}(k)$ = slope of pth quantile at the kth SEG, $\beta_1^{(p)}(i)$= slope of pth quantile at the ith year, and $\beta_2^{(p)}(j)$ = slope of pth quantile at the adjusted present-day in 2010 (j=1), so that the exposure estimates are not affected by the discrepancy between present-day and historical measurement.

We also reconstructed the EMP exposure matrix with a linear regression model that used the arithmetic mean of the concentration of NIOSH 7400 to estimate exposures for all combination of SEG, mine, and year between 1955 and 2010 (Equation 4- 4).

$X_{kij} = \alpha_0 + \sum_{k=1}^{k=28} \alpha_k (k) + \beta_1 (i) + \beta_2 (j)$ for k= 1, 2, ..., 28th SEG, i = 0, 1, …, 55th year , and j = 0, 1 for adjusted present-day in 2010 (Equation 4- 4)

where X_{kij} = concentration of NIOSH 7400 definition for the ith year of the kth SEG with the adjusted present-day in 2010 (j=1), α_0 = intercept based on X_{kij}, $\sum_{k=1}^{k=28} \alpha_k$ (k) = slope of the kth SEG, β_1 (i)= slope of the ith year, and β_2 (j) = slope of the adjusted present-day in 2010.

b. Constant exposure model

This model assumed that EMP exposures between 1955 and 2010 for each SEG in each mine were constant over time. The constant value was obtained as the median of all the historical data and present-day measurements taken together. The quantile regression model mainly used the 50th percentile to calculate the cumulative exposure; therefore, in the constant exposure model we used the median exposure value instead of the mean of the available data as the constant value.

$\widetilde{Y}_{kl} = \log (X_{kl})$ for k = 1, 2, …, 28th SEG, and l = 1, 2, …, 7th mine (Equation 4- 5)

where $\log (X_{kl})$ = log concentration of NIOSH 7400 definition for the kth SEG in lth mine.

Developing the SEG exposure matrix

Our SEG exposure matrix includes 7 mines: 2 in the east, 5 in the west, and 28 SEGs, covering 56 years, at 1-year intervals between 1955 and 2010. Exposure data were not available for many of the cells of the matrix. For each SEG in a mine that did not have any exposure data available, the exposure was imputed as the average of all SEGs within the department in that mine to which the SEG belonged. For example, there were no measurements for the *Basin operator* SEG in the Northshore mine. Therefore, we averaged the total EMP exposures in the *Mining* department, which included the *Mining operator 1, Mining operator 2*, and *Rail Road* SEGs, to impute the missing value. As there were no present-day measurements for LTV, we used the same data for each SEG as in Northshore, which, according to the Natural Resources Research Institute (French, 1968; McSwiggen & Morey, 2008), has similar geological characteristics. Based on these imputation methods, the exposure matrix consists of 10,976 cells (7 mines x 28 SEGs x 56 years) for the time-varying exposure model and 196 cells (7 mines x 28 SEGs) for the constant exposure model for each of the EMP exposure metrics. We used separate models to estimate the 25th, 50th, and 75th percentiles for each EMP exposure metric.

Calculating cumulative exposures

The resulting exposure matrix was combined with the employment history of each worker who is either a case or control in the epidemiological study to estimate a cumulative exposure. All cases (n=57) and controls (n=184) were nested within the Mineral Resources Health Assessment Program (MRHAP) cohort (n=68,737), and included employment history in the taconite mining industry. These cumulative exposures will be used for the dose-response assessment in the epidemiological analysis. Assigning equal weight to each time period over each individual worker's work history, the cumulative exposure for a worker is given by

$$\text{Cumulative exposure (EMP-year/cm}^3\text{)} = \sum_{i=1}^{n} C_i t_i \qquad \text{(Equation 4- 6)}$$

where C_i is the estimate of exposure for the i^{th} time period and is obtained from the time-varying and constant exposure models and t_i is the time period over which exposure occurred. The average work-life exposure for a worker is given by:

$$\text{Average exposure (EMP/cm}^3\text{)} = \frac{\sum_{i=1}^{n} C_i t_i}{\sum_{i=1}^{n} t_i} \qquad \text{(Equation 4- 7)}$$

RESULTS

Conversion factors from total NIOSH 7400 EMP to amphibole NIOSH 7402 EMP

The CFs from total to amphibole EMP were calculated for SEGs in the eastern zone only. However, all of the amphibole EMP measurements in eight SEGs in the eastern zone were below the LOD, preventing us from calculating CFs. As seen in Table 4-2, all the calculated CFs were less than 1, indicating that all the SEGs in the eastern zone had small fractions of amphibole EMP. The highest CF (0.47) was found in the *Operating technician* SEG, which is part of the *Crushing* department. The range of CFs for the remaining SEGs was smaller, 0.05 – 0.29.

SEG exposure matrix

Estimating exposures using two regression models

The exposures were reconstructed using a time-varying and a constant exposure model. The only mine that did not exhibit any variation with time in the time-varying model was Hibbtac with a zero slope for the temporal trend obtained from the quantile regression. The other mines exhibited different temporal trends: increasing exposures with time at Keetac and Minntac and decreasing exposures with time at Northshore, Utac, Minorca, and LTV. Second, we used a constant exposure model to obtain the median of exposure value based on the actual data. A median value by SEG by mine was calculated to estimate exposure across all years (1955-2010), so the slope for this model is zero. A comparison of the time-varying and constant exposure models for the Northshore mine is provided in Figure 4-1 for each year between 1955-2010, along with available present-day measurements and historical data. The two models provide similar estimates for the present-day, from where most of the data come. However, the exposure estimates of the two models differed by a factor of ~10 during the early historical periods (1 EMP/cm^3 for the time-varying exposure model versus 0.1 EMP/ cm^3 for the constant exposure model).

Adjustment of present-day measurements

The present-day exposure measurement values were slightly higher than the historical measurement across SEG and mine. To prevent yearly trends from being affected by more sensitive present-day measurements, we added a term in the model for whether the data was collected from present-day measurements and adjusted the historical data. An example plot for the *Crusher operator* SEG at Northshore mine using the NIOSH 7400 data is shown in Figure 4-2. In a comparison of the two models, one with and one without the present-day measurement adjustment, our results showed that present-day measurements were 0.026 EMP/cm^3 higher than the trend suggested by the historical data alone. Account for this difference attenuated the time trend.

Size-based total EMP definition with NIOSH 7400

Using Equation 4- 1 and Equation 4- 2, we converted the exposure estimates based on the NIOSH definition to the other size-based EMP definitions, and created exposure matrices for each of the definitions for each SEG by mine between 1955 and 2010. The estimates for Northshore are shown in Figure 4-3. The reconstructions for each definition can be compared with the NIOSH 7400 method estimate. In addition, the amphibole EMP using NIOSH 7402 method are shown to compare with the total EMP using NIOSH 7400 method. Across each mine, we found that the exposure levels were higher for total Suzuki and Cleavage fragment EMP than for total NIOSH 7400 and Chatfield EMP. In general, the Suzuki and Cleavage fragment concentrations were similar to one another as were the NIOSH 7400 and Chatfield concentrations. This observation is consistent with our measurements from the area sampling, in which the total EMP were found to be primarily comprising short EMP with a small aspect ratio, most commonly 1–3 µm in length and 0.2–0.5 µm in width (Hwang *et al*., 2013b).

Comparison between departments by mine

In Figure 4-4, we selected one representative SEG from each department across all mines so that we could compare the NIOSH 7400 estimates across departments and mines. Since the exposure estimates for the *Office staff/Control room operator* department were low and did not substantially differ across mines, we excluded this department from the panel. Because almost all data from the Keetac mine were drawn from present-day data (87%, 203/233), we were unable to predict changes with time for many of the processes there. Overall, Northshore, one of the two mines in the eastern zone, had the highest EMP concentrations. Utac, one of the mines in the west, overall exhibited the highest concentration in the earlier years. The exposures for the Utac mine dramatically decreased over the years, a trend driven by the large number of high EMP exposure concentrations in the historical data.

Cumulative exposure distribution

The resulting exposure matrix was combined with employment history of each worker to estimate an average exposure in units of EMP/cm^3 and a cumulative exposure in units of EMP-year/cm^3 for each worker. As shown in Figure 4-5(a), the average total EMP exposure concentration for the entire cohort population was calculated using both the time-varying and constant exposure models. Using the time-varying exposure model, ~60% of the workers in the mesothelioma case control study had lifetime average total EMP exposure levels greater than 0.1 EMP/cm^3. However, the difference between the control group and all workers was not large, as the number of workers in the control group was much larger. With the constant exposure model, ~10% of the workers in the mesothelioma case control study had lifetime average total EMP exposure levels greater than 0.1 EMP/cm^3. The cumulative EMP exposure concentration is shown for both the case (n=57) and control (n=184) populations in Figure 4-5(b) using the time-varying

EMP concentration model. The cumulative distributions were not markedly different for the two groups.

DISCUSSION

In this study, we reconstructed historical exposures for a mesothelioma case-control study using four different EMP size definitions, including NIOSH 7400. Each of the four exposure matrices provides estimates of exposure levels for every SEG, listed by year and mine.

Linear vs. Quantile Regression Model

While the linear-regression model specifies the change in the conditional mean of the response variables, the quantile regression (time-varying) model specifies changes in the conditional quantile. Under the conditions of interest, the quantile regression analysis estimates the relationship between the dependent and the independent variables (Hao and Naiman, 2007; Zou and Yuan, 2008). Right-skewed distribution, commonly occur in industrial hygiene measurements, resulting in neither normal nor lognormal distributions (Bullock and Ignacio, 2006). In such distributions, the conditional mean can become an inappropriate and misleading indicator of central location because it is heavily influenced by outliers. Therefore, quantile regression analysis, which is not sensitive with respect to assumptions about normality or lognormality distribution of the data, was used to reconstruct the historical exposure for each SEG by mine for each year of operation between 1955 and 2010. The panels in Figure 4-6 are shown to compare the linear and quantile regression model estimates for NIOSH 7400 EMP in Northshore mine. The linear regression model with the arithmetic mean is what we will be using in the mesothelioma case-control study. The cumulative exposures are calculated under the assumption that the dose-response relationships in epidemiology studies are linear even if the exposure data are not log-normally distributed (Rappaport, 1991; Seixas *et al.*, 1988). The exposure estimates using the linear regression model were similar to those using the constant exposure model (Figure 4-1). As mentioned previously, a great deal of information was incomplete, and time-varying exposure model allowed us to estimate missing measurements and determine exposures.

The model is the best that can be done given our sparse data, as it is based on year and SEG only. Our model choice and its simplicity were dictated by the sparseness of the data and the fact that workplace processes have not changed significantly over the years of the study. The estimated concentrations were determined by mine, so the estimate for one mine does not affect another. The slope within a mine is the same across all SEGs because the model is run by year and by SEG separately with no interaction term. With our sparse database, a model including interaction terms was problematic because too few measurements are available to evaluate those terms. However, other studies have used interaction term (random effects) and/or quadratic term in the model (Koh *et al.*, 2012). Furthermore, as shown in Figure 4-4, both actual present-day measurements and available historical data fell within the range of our reconstructed exposure estimates. It was tempting to consider using a constant exposure model as no significant taconite processes changes have occurred. However, we observed significant slopes, indicating that the constant exposure model is not justifiable. Thus, we felt confident using the time-varying regression exposure model instead of the constant exposure model.

We observed that the large negative slope of concentration versus time for the Utac mine might be influenced by 12 historical data values for the *Auto mechanic* SEG in this mine that were significantly higher than any other data value. To evaluate the sensitivity of the slope to these 12 data points, we excluded them and reconstructed the exposure matrix. Although the slope was dramatically reduced so that the earliest concentrations were reduced by a factor of almost 10, the cumulative distribution for the cumulative EMP exposure concentration was not noticeably altered.

Present-Day vs. Historical Measurements

The present-day measurements were slightly higher than the historical measurements although the engineering control system (e.g., enclosures, ventilation, and dust collector), administrative control, and personal protective equipment (PPE) have been improved in recent years. One possible explanation is that the sampling times were different for the present-day and the historical data. In addition, the purpose of the sampling in present-day data was for the on-going epidemiologic studies.

The inclusion of an adjustment for present-day measurement fit the data better. The LTV mine was the sole mine for which only historical data was available, as the mine was closed in 2001. Therefore, the exposure matrix for that mine was the same for both the adjusted and non-adjusted present-day data. Department is a higher-level variable than SEG, so the average of department estimates was better than the average of all data in the LTV mine. In contrast to the other mines, department was not used to substitute the missing exposures for LTV. For instance, neither the *Office staff* nor the *Control room* SEGs was available in this mine; the department level (*office staff/control room operator*) was also missing. Therefore, we filled in the missing information for these two SEGs with corresponding data from the Northshore mine, which has similar geological characteristics.

Classification by EMP Size Definition

The workers were classified into tertiles according to their cumulative exposure using each of the EMP definitions. Across EMP definitions, a very limited number of workers (average 5 out of 241 workers in cohort) were differently classified. In comparison to the exposures according to the NIOSH 7400 definition, only 2, 3, and 4% of worker exposures were underestimated using the Suzuki, Chatfield, and Cleavage fragments definitions, respectively (See Table 4-3). This small range of differential classification makes sense when compared to the high correlations among the EMP definitions reported in Hwang *et al*. (2013b). The findings indicate that using different EMP size definitions will not affect subsequent epidemiological analyses substantially.

The chief limitation of this study is that our reconstructed historical exposure matrix may be biased towards present-day data, which comprise the majority of the data. Since many SEGs have no historical data, our regression model has the same slope across all SEGs within a mine. However, this was the best statistical model given the sparseness of the historical record.

CONCLUSIONS

Our SEG exposure matrices are based on present-day measurements as well as historical data, while other studies often depend on only historical data. Although no significant taconite processes changes have occurred, we observed sizeable slopes in the time-varying exposure model, indicating significant time dependency. There were minor differential classifications of EMP across the size-based definitions, which is consistent with the finding of high correlations among the definitions. Our findings indicate that different EMP size definitions will not affect subsequent epidemiological analyses substantially. The average total EMP exposure concentration for the entire cohort population was calculated using both time-varying and constant exposure models. The difference between the control group and all (both control and case) workers was not large, as the number of workers in the control group was much larger. Similarly, the cumulative distributions were not markedly different for the case and control groups.

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Table 4-1 Number of available historical (H) and present-day (P) EMP observations by mine

SEG	Northshore		Hibbtac		Utac		Keetac		Minntac		Minorca		LTV [a]
	H [b]	P [c]	H	P	H	P	H	P	H	P	H	P	H
Basin operator	. [d]	.	0 [e]	12	0	12	0	6	0	12	.	.	0
Mining operator 1	31	6	6	12	14	6	7	12	22	12	14	6	3
Mining operator 2	29	9	8	6	6	6	6	24	25	12	14	5	14
Rail road	0	6	.	.	.	.	.	.	.	.	.	.	2
Crusher maintenance	6	22	0	12	3	12	0	12	0	12	1	5	32
Crusher operator	81	22	6	11	24	11	4	6	19	10	33	6	.
Operating technician	2	10	.	.	4	12	1	0	.	.	.	.	0
Concentrator maintenance	0	10	0	12	0	6	0	12	0	12	0	6	.
Concentrator operator	13	22	4	12	4	11	4	12	29	12	6	6	3
Balling drum operator	0	18	.	.	0	9	0	12	3	12	0	6	0
Dock man	2	10	0	11	1	12	0	6	0	11	1	6	0
Furnace operator	3	9	3	0	0	18	0	12	10	9	1	6	0
Pelletizing maintenance	0	12	0	12	0	12	0	12	0	12	0	5	.
Pelletizing operator	1	11	7	11	3	5	3	12	22	11	3	6	2
Boiler technician	.	.	.	.	0	6	.	.	1	12	.	.	.
Carpenter	4	0	0	6	.	.	0	6	0	12	.	.	0
Electrician	5	12	0	12	0	12	0	12	0	12	0	6	.
Janitor	6	0	3	0	4	0	.	.	12	0	.	.	0
Lubricate technician	0	6	1	0	0	18	.	.	.	.	0	6	0
Maintenance technician	25	9	2	17	14	5	0	6	8	7	2	6	1
Pipefitter/Plumber	11	0	.	.	0	6	.	.	3	12	.	.	2
Repairman	.	.	0	12	.	.	5	0	1	10	0	6	.
Supervisor	1	15	0	5	0	5	0	6	0	11	1	6	0
Auto mechanic	0	10	0	11	12	11	0	5	0	12	1	6	8
Lab analyst	0	24	0	6	0	6	0	12	0	6	0	6	0
Warehouse technician	1	8	0	6	0	5	0	6	0	12	0	6	0
Control room operator	0	9	0	6	0	6	0	6	0	12	0	12	3
Office staff	0	6	0	5	0	6	0	6	0	12	0	6	0
Total N=	221	266	40	197	89	218	30	203	155	267	77	129	70

[a] LTV does not have present-day measurement, [b] Historical data, [c] Present-day measurement, [d] SEGs are not present, [e] SEGs do not have measurement

Table 4-2 Conversion factors (CFs) between NIOSH 7400 and NIOSH 7402 in the eastern zone

Department	**SEG**	**CFs**
Mining	Basin operator	.[a]
	Mining operator 1	-[b]
	Mining operator 2	0.05
	Rail road	-
Crushing	Crusher maintenance	0.13
	Crusher operator	0.14
	Operating technician	0.47
Concentrating	Concentrator maintenance	0.13
	Concentrator operator	0.14
Pelletizing	Balling drum operator	0.24
	Dock man	0.09
	Furnace operator	0.21
	Pelletizing maintenance	-
	Pelletizing operator	0.12
Shop (mobile)	Boiler technician	.
	Carpenter	.
	Electrician	0.19
	Janitor	0.16
	Lubricate technician	0.10
	Maintenance technician	-
	Pipefitter/Plumber	.
	Repairman	.
	Supervisor	0.19
Shop (stationary)	Auto mechanic	-
	Lab analyst	-
	Warehouse technician	0.29
Office/control room	Control room operator	-
	Office staff	-

[a] No present-day measurement analyzed by NIOSH 7402.

[b] All samples using NIOSH 7402 are below LOD.

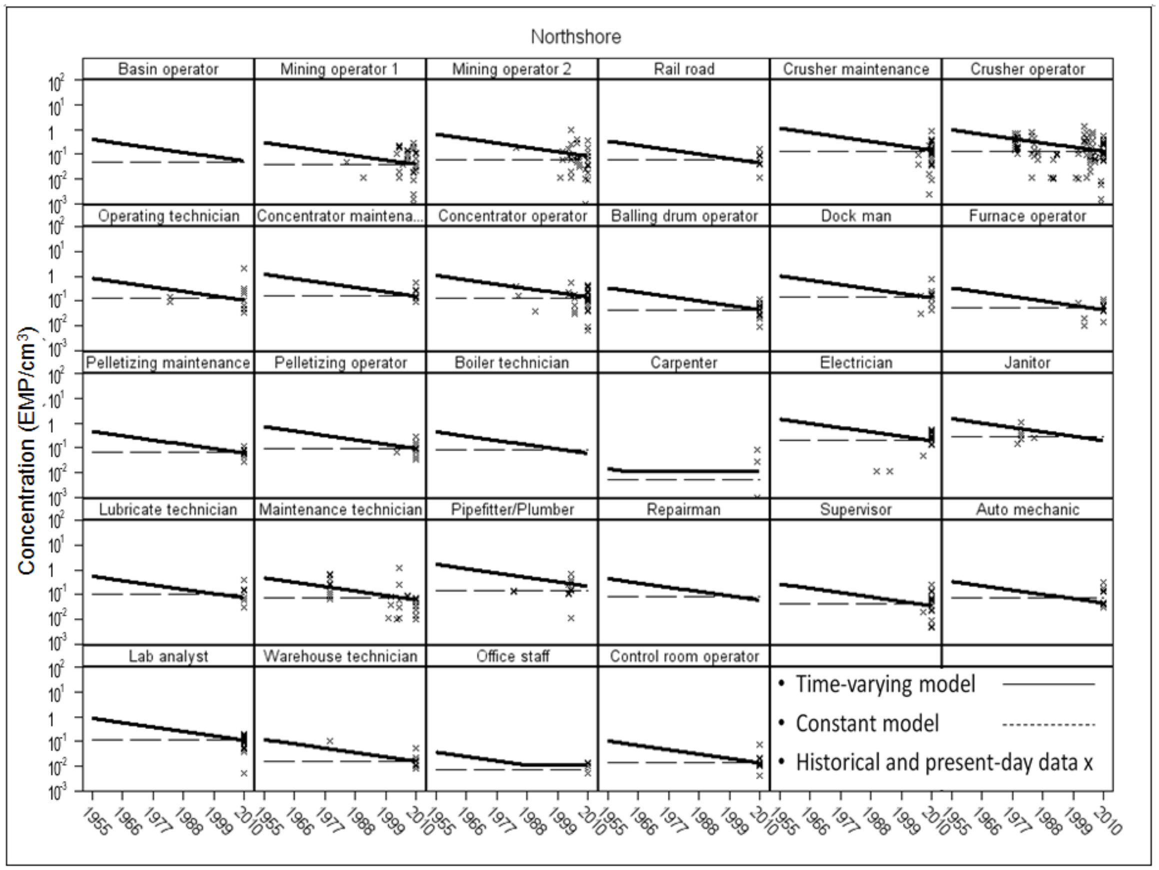

Figure 4-1 Exposure panel matrix comparing time-varying and constant exposure models by SEG for the Northshore mine.

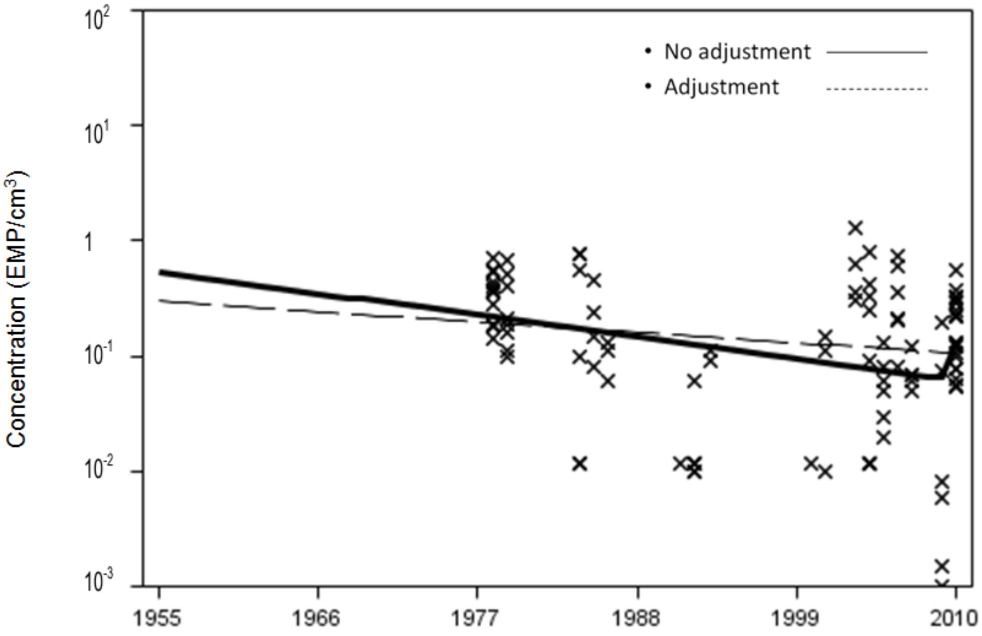

Figure 4-2 Comparison of non-adjusted and adjusted present-day data by year

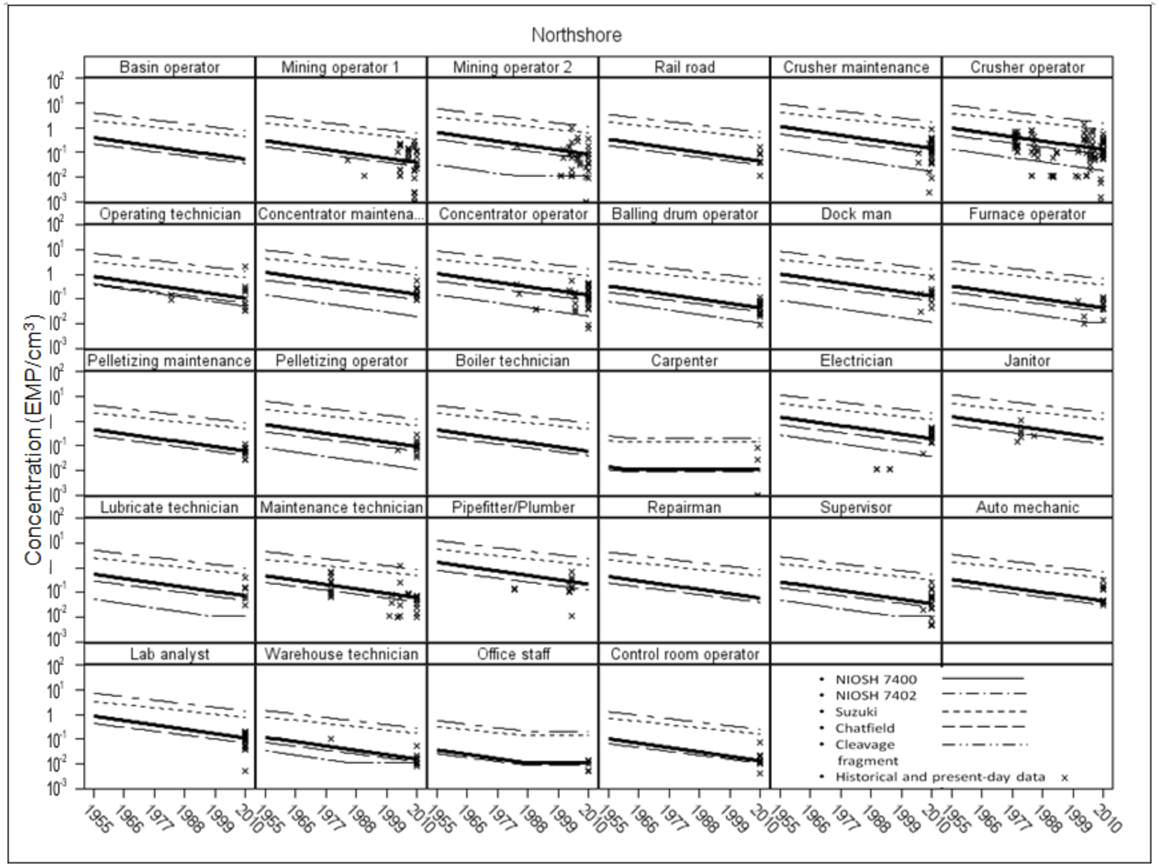

Figure 4-3 Exposure reconstruction matrix for different EMP definitions (NIOSH 7400, Suzuki, Chatfield, and Cleavage fragments) and amphibole EMP (NIOSH 7402) by SEG for Northshore (Scatterplot displays actual NIOSH 7400 measurements)

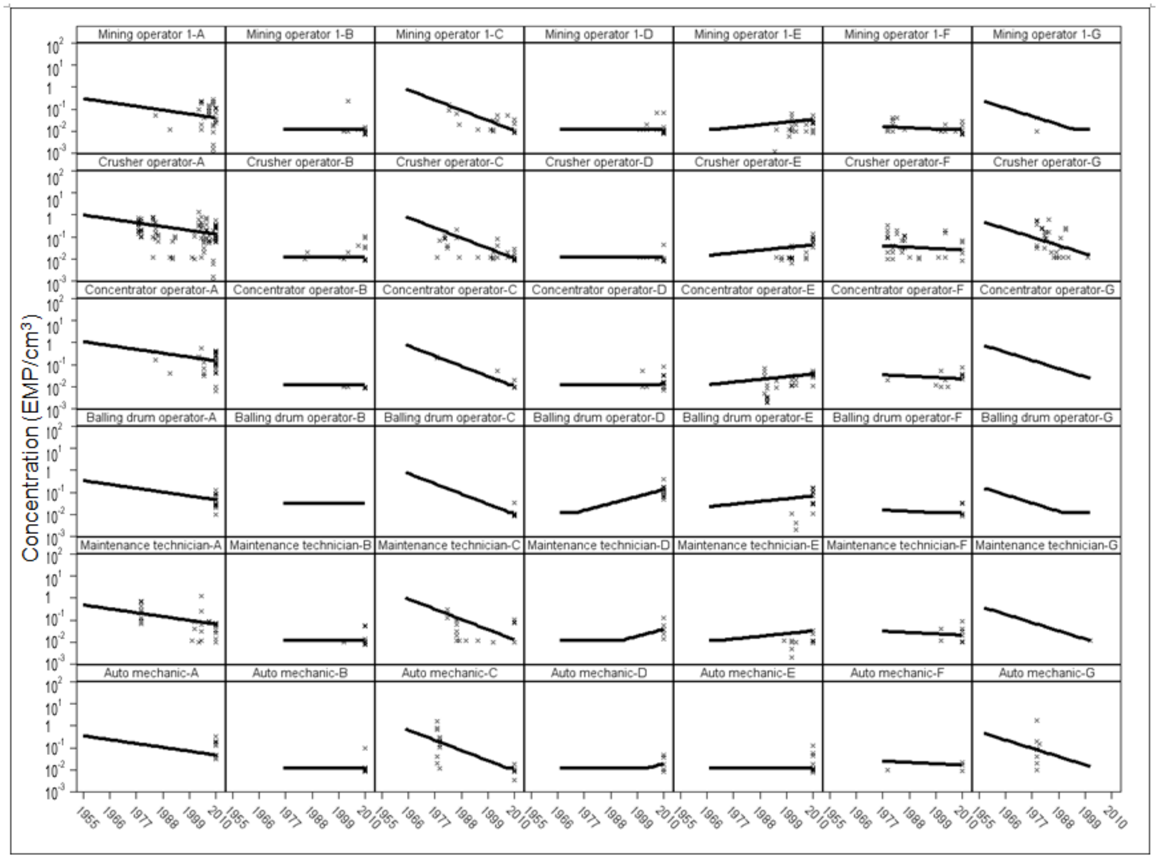

Figure 4-4 NIOSH 7400 exposure estimates for one representative SEG in each of the six departments across all mines (A: Northshore, B:Hibbtac, C:Utac, D:Keetac, E: Minntac, F: Minorca, G: LTV).

Table 4-3 Misclassification by size-based definitions

Classification	#of worker (%)	#of worker (%)	#of worker (%)
Suzuki	ST1	ST2	ST3
NT1	77 (0.32)	3 (0.01)	0 (0.00)
NT2	3 (0.01)	75 (0.31)	2 (0.01)
NT3	0 (0.00)	2 (0.01)	79 (0.33)
Chatfield	CT1	CT2	CT3
NT1	77 (0.32)	3 (0.01)	0 (0.00)
NT2	4 (0.02)	74 (0.31)	2 (0.01)
NT3	0 (0.00)	2 (0.01)	79 (0.33)
Cleavage fragment	CFT1	CFT2	CFT3
NT1	77 (0.32)	3 (0.01)	0 (0.00)
NT2	3 (0.01)	76 (0.32)	1 (0.00)
NT3	0 (0.00)	1 (0.03)	80 (0.33)

T1: tertile 1, T2: tertile 2, T3:tertile 3
N: NIOSH, S: Suzuki, C: Chatfield, CF: Cleavage fragment

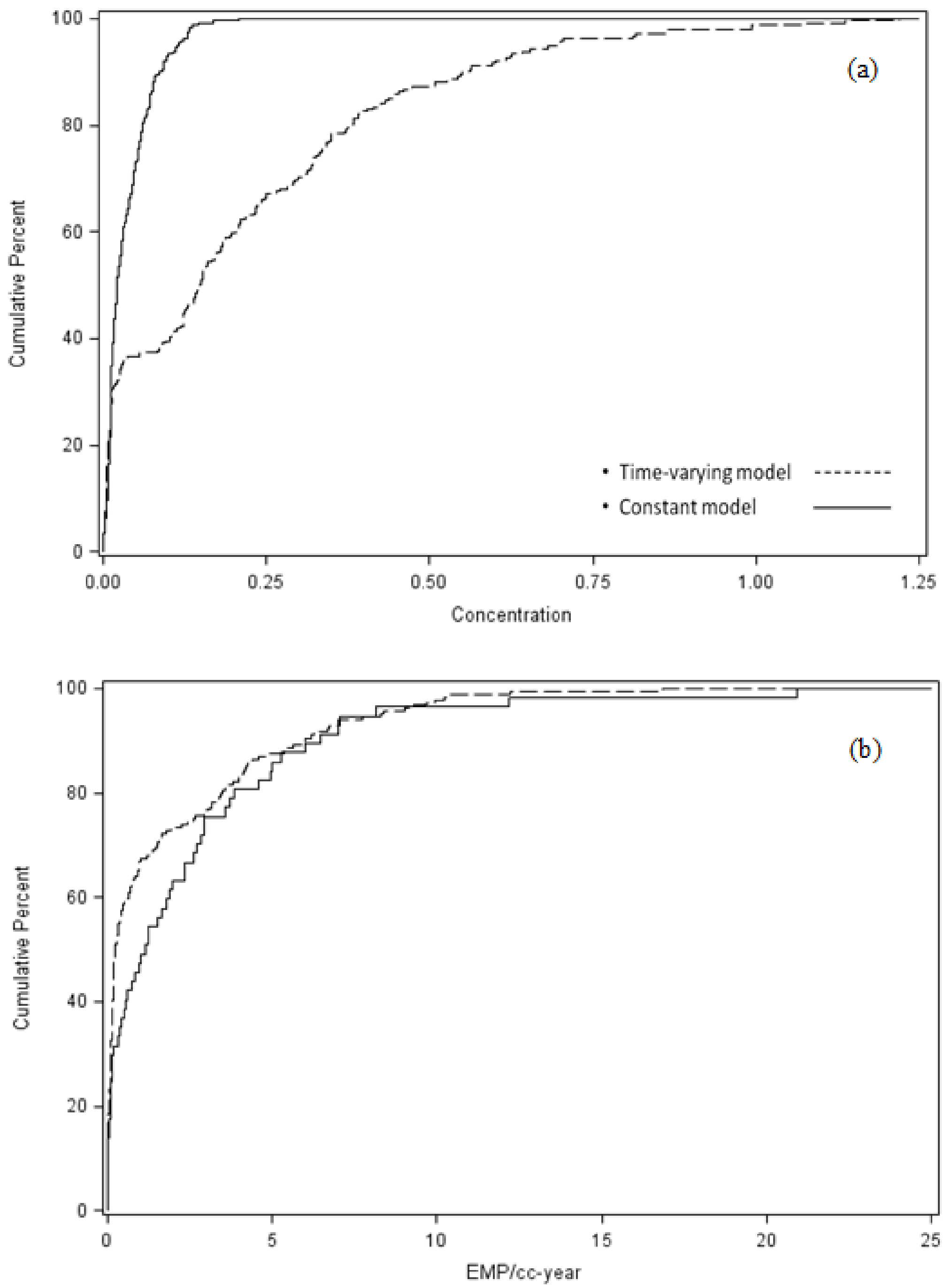

Figure 4-5 Cumulative distribution for (a) the average NIOSH 7400 EMP exposure concentration using both time-varying and constant exposure models for cases and controls (n=241) for NIOSH 7400 and (b) the cumulative NIOSH 7400 EMP exposure distributions using time-varying exposure model

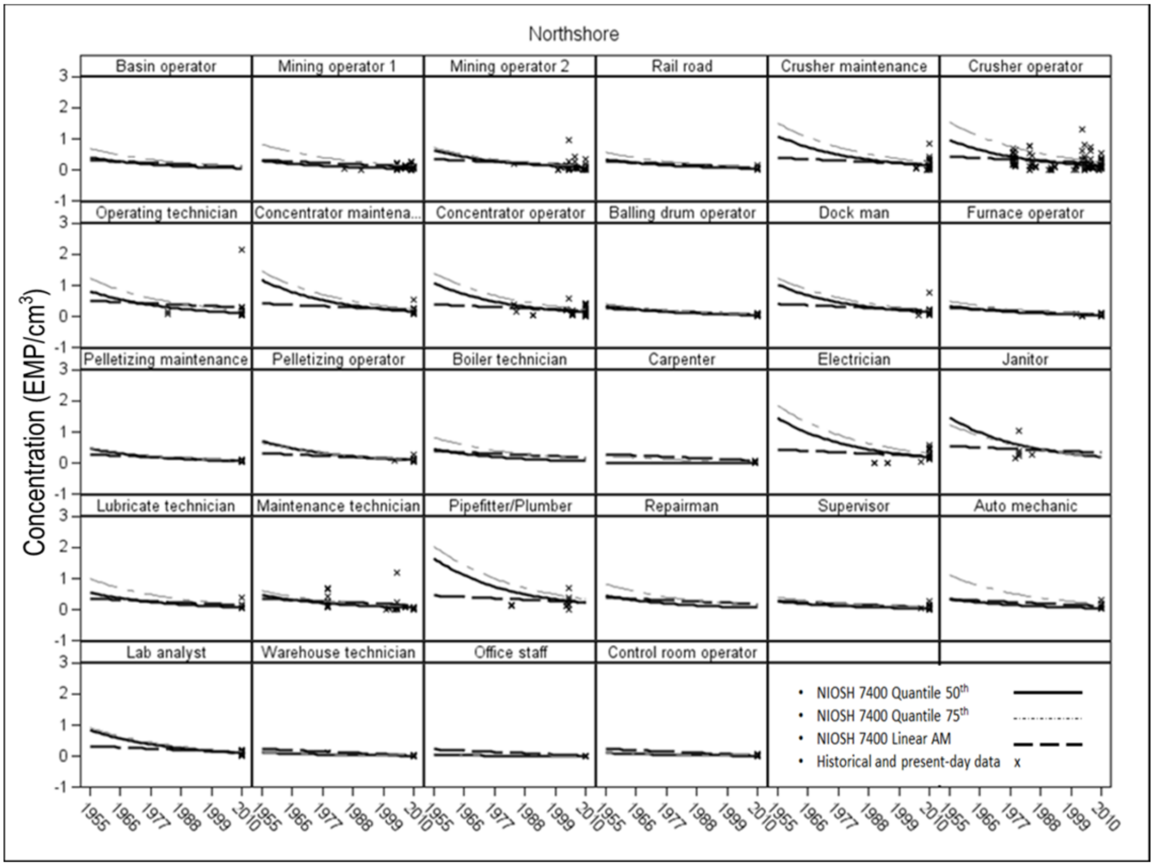

Figure 4-6 Exposure reconstruction matrix for comparison models between quantile regression vs. linear regression by SEG for Northshore

Chapter 5 Comprehensive Assessment of Exposures to Respirable Dust (RD) And Respirable Silica (RS) in the Taconite Mining Industry

INTRODUCTION

The potential relationship between taconite dust and health risks in northeastern Minnesota's Mesabi Iron Range has raised concerns in the taconite mining industry and surrounding communities. Therefore, we conducted a study to measure exposure levels to various components of taconite dust. Respirable dust and silica are the most common dust components observed during the taconite mining processes – drilling, crushing, feeding, and transferring. Respirable dust is the fraction of dust (50% of cut-size is 4 µm) that penetrates into the respiratory system (Hinds, 1999). Respirable silica, which is a subset of respirable dust, consists of two mineral forms: crystalline (free silica) and amorphous. The crystalline form has three subgroups: quartz, tridymite, and cristobalite (Steenland & Stayner, 1997), the most common of which is quartz. In our study, we focused on the crystalline form-quartz.

Exposure to the crystalline forms of silica in industrial settings has long been associated with the development of silicosis, a fibrotic pulmonary disease (Hayumbu *et al*., 2008; Pelucchi *et al*., 2006; Steenland & Sanderson, 2001; Archer *et al*., 2002; Chen *et al*., 2001; Collins *et al*., 2005). Although the United States Geological Survey (USGS, 2013) reported that quartz production in the U.S. dramatically increased between 1968 and 1988, from 30.4 to 389 tons, between 1989 and 2000, production dropped from 464 to 189 tons (data were not available after 2001). Correspondingly, the National Institute for Occupational Safety and Health (NIOSH, 2012) found that the number of deaths from silicosis has decreased by approximately 10 times from 1968 to 2007. Similarly, in our taconite mining mortality study (Minnesota Taconite Workers Health Study, 2013), we found that the number of deaths by silicosis had not increased. A radiographic study of taconite miners in 1980 found several cases of silicosis but no other significant abnormalities or diseases (Clark *et al*., 1980). Higgins *et al*. (1981) studied the taconite workers in the Reserve mining company and found no association between mortality and lifetime exposure to either respirable dust or respirable silica, concluding that the taconite miners had no elevated risk of mortality.

Only a few studies have investigated exposure to respirable dust and respirable silica in the taconite mining industry. Sheehy (1986) conducted respirable quartz exposure sampling and found that respirable silica exposures in the taconite industry have often exceeded 0.1 mg/m^3, which was the standard NIOSH recommendation at that time. Sheehy and McJilton (1990) reported that the concentrations of silica-containing dust were above acceptable limits in mines, crushers, and concentrators, but not in pellet plants. The limits referenced included the Recommended Exposure Limit (REL), set by the NIOSH, and the Permissible Exposure Limit (PEL), set by the Mine Safety and Health Administration (MSHA). Quartz silica concentrations ranged from less than 0.04 to 0.11 mg/m^3, and no tridymite or cristobalite was found in any of the samples (McJilton, 1984). Since the 1990s, exposure levels to respirable dust and respirable silica have not been studied for epidemiological purposes in the taconite mining industry. To assess exposure levels to these dust components, we conducted personal air sampling across all currently operating mines in the Mesabi Iron Range.

Exposure levels to respirable dust and respirable silica can vary across taconite processes. In this article, we mainly focus on the taconite milling process, sub-grouped by Department – Mining, Crushing, Concentrating, and Pelletizing. In Mining, holes are drilled into the extremely hard taconite rock to break it up. The production trucks haul the taconite directly to the Crushing plant, during which quite a large amount of dust is generated. In Crushing, the ore is crushed to about 10 cm in diameter in the primary crusher and the desired smaller sizes in the secondary or tertiary crusher. The sizes of the crushed ore vary by type of crusher and by mine. The crushing process produces a significant amount of dust by definition (the ore is crushed until it is small). In the next department, Concentrating, the rock is mixed with water and ground in rotating mills until it is as fine as powder. Then, the iron ore is separated from the taconite using a magnetic separator, which removes the waste rock (called "tailings") from the iron-bearing grains of taconite powder (called "concentrate"). Flotation processes separate floating silica-containing particles from settling iron-containing particles. Unlike the previous processes, Concentrating, which consists of the wet process from magnetism as well as the silica removal process from flotation, significantly reduces the amount of respirable dust and silica. The last milling process, which occurs in the Pelletizing department, removes water from the concentrated iron slurry and then mixes the slurry with bentonite and/or limestone to adhere the particles. Large rotating cylinders make balls about 1 cm in diameter, which are then dried and fired at up to 1300 degrees Celsius. Although this last process is a dry process, silica containing dust is less likely to be generated after the Concentrating process.

The mineralogy of the Mesabi Iron Range changes from east to west, exhibiting distinct metamorphic mineralogical zones. In the eastern zone, the iron ore contains amphibole from the cummingtonite-grunerite series and the ferroactinolite-tremolite series. In contrast, the ore in the western zone predominantly contains phyllosilicates such as minnesotaite, greenalite, and stilpnomelane, which are not regulated like asbestiform or amphibole elongated mineral particles (EMP) (McSwiggen & Morey, 2008; Zanko *et al.*, 2008).

The goals of this paper are (1) to assess the present-day levels of exposure to respirable dust (RD) and respirable silica (RS) in the taconite mining industry; (2) to estimate the between-mine, between- SEG (similar exposure group), and within-SEG components of variability for RD and RS exposures; and (3) to evaluate how the taconite mining process influences the exposures to RD and RS.

METHODS

Formation of SEGs

SEGs can be used to assess exposures more efficiently using job titles, locations, tasks, and procedures (Bullock & Ignacio, 2006).We created a historical exposure assessment database, which listed workers (or areas) and job titles according to tasks and processes. The job titles were mainly derived from (i) records maintained by the Mine Safety and Health Administration (MSHA); (ii) information from a previous study by Sheehy (1986); (iii) industrial hygiene and human resources databases maintained by the three companies currently operating mines in the Mesabi Iron Range (U.S. Steel, Cliffs Natural Resources, Arcelor Mittal); and (iv) *Job Descriptions and Classifications*, published by the Reserve Mining Company in 1974. The final list contained 181 job titles, forming 28 SEGs that we further grouped into seven departments.

Sampling design and data handling

A personal exposure assessment was conducted across six mines, all currently operational, in the Mesabi Iron Range beginning in January 2010 and ending in May 2011. The purpose of the sampling was to assess present-day levels of worker exposure to RD and RS in the taconite mining industry. Prior to the day of sampling, the researchers and representatives from the mining companies discussed workers' schedules and identified potential participants. At the beginning of the work shift on each sampling day, the researchers explained the purpose of the study and presented the potential participants with the consent form approved by the University of Minnesota Institutional Review Board (IRB code: 0901M58041).

To perform a baseline exposure profile for a job title, the American Industrial Hygiene Association (AIHA) sampling strategy (Bullock & Ignacio, 2006) recommends that a minimum of 6, but preferably 8 to 10, data points per SEG be used. Two workers per SEG were selected for personal sampling in the eastern zone; in the western zone, approximately eight workers per SEG were chosen. Each consenting participant wore a personal air-sampling pump (Apex Pro pump, Casella Inc., Amherst, NH, USA) on his or her waist, with the sampler located in the breathing zone, for approximately six hours, which accounts for at least 70% of a daily work shift. Personal sampling for each worker was completed during three different work shifts, though not necessarily on consecutive days. One blank sample per sampling day (approximately 14% of the samples) was collected for quality control.

Each sample for RD and RS was taken using a single filter cassette. We used a 5-micron pore size, 37mm in diameter polyvinyl chloride (PVC) membrane filter and 3-piece filter cassettes for sampling. The flow rate for the sampling pump was calibrated and operated at 2.5 liter/min using a 37mm aluminum cyclone (SKC Inc., Eighty Four, PA, USA).

Analytical methods and limitations

The RD concentration, based on the mass of the respirable dust fraction, was calculated using NIOSH 0600 *Respirable particulates not otherwise regulated gravimetric*. The RS analysis was performed using NIOSH 7500 *Crystalline silica X-ray diffraction*. These NIOSH methods stipulate that particles collected by the filter not exceed a mass of 2 mg because the collected particles could then block the filter, resulting in a lower airflow rate than that in the calibrated air sample. We excluded three samples that were overloaded with particles and six that exhibited a low sampling volume from further data analysis.

Table 5-1 lists the number of personal samples analyzed using both NIOSH 0600 and NIOSH 7500 for each mine. The total number of samples and the number of blanks were 679 and 132, respectively. The blanks did not show any dust or silica above limits of detection, so they were not used further in our analyses. If all of the measurements for a given SEG fell below the LOD, summary statistics such as the arithmetic and geometric means and geometric standard deviations were reported as “<LOD”. If at least one sample for an SEG in a particular mine had measurements above the LOD, then summary statistics were calculated under the assumption that censored data were represented by one half of the LOD.

Statistical analysis methods

Of the 28 SEGs, 27 were monitored. We were not able to monitor the *Janitor* SEG because all of the janitors in the current taconite mining industry are independent contractors. Furthermore, not all 27 SEGs were represented in each mine. For instance, some mines have detailed job titles that correspond to an SEG (e.g., *Boiler technician*), but others have no corresponding jobs or tasks.

To compare the log-transformed estimated exposures Y_{ij} of each mine by SEG, we used a simple one-way ANOVA model (Equation 5- 1). Tukey's Studentized Range (HSD) test was also used for pair-wise comparison of exposures within each mine by SEG to determine homogeneity. The dependent variables were RD and RS, respectively.

$$Y_{ij} = \log (X_{ij}) = \mu_y + \alpha_i + \varepsilon_{ij} \text{ for } i = 1, 2, \ldots, 6, \text{ and } j = 1, 2, \ldots, 12 \qquad \text{(Equation 5- 1)}$$

where X_{ij} = exposure concentration of the i^{th} mine at the j^{th} observation for each SEG, μ_y = overall mean of Y_{ij}, α_i = random deviation of the i^{th} mine's true exposure from μ_y, and ε_{ij} = random deviation of the j^{th} observation from the i^{th} mine's true exposure.
(Equation 5- 1 assumes that the ε_{ij} are independently and identically distributed with a normal distribution.

A one-way nested random-effects ANOVA model was used for estimating between-SEG (BG) and within-SEG (WG) variance components using the log-transformed exposure concentrations (Equation 5- 2).

$Y_{kj} = \log (X_{kj}) = \mu_y + \alpha_k + \varepsilon_{kj}$ for the observations $k = 1, 2, \ldots, 27$, and $j = 1, 2, \ldots, 12$

(Equation 5- 2)

where $X_{kj} = j^{th}$ observation of exposure concentration for the k^{th} SEG, μ_y = overall mean of Y_{kj}, α_k = random deviations of the k^{th} SEG's true exposure from μ_y, and ε_{kj} = random deviations of the j^{th} observation for k^{th} SEG from $\mu_{y,\,k}$ (mean exposure of the k^{th} SEG). The random deviations (α_k and ε_{kj}) are assumed to be normally distributed with zero means and variances (σ^2_α and σ^2_ε, respectively). These variances are mutually uncorrelated and estimated as variance components ($S_y{}^2_{BG}$ and $S_y{}^2_{WG}$, respectively).

A two-way nested random-effects ANOVA model was used for estimating between-mine (BM), between-SEG, and within-SEG variance components (Equation 5- 3).

$Y_{ikj} = \log (X_{ikj}) = \mu_y + \alpha_i + \beta_{ik} + \varepsilon_{ikj}$ for the observations $i = 1, 2, \ldots 6$, $k = 1, 2, \ldots, 27$, and $j = 1, 2, \ldots, 12$

(Equation 5- 3)

where X_{ikj} = j^{th} observation of exposure concentration for the k^{th} SEG of the i^{th} mine, μ_y = overall mean of Y_{ikj}, α_i = random deviations of the i^{th} mine's true exposure from $\mu_{y,}$ β_{ik} = random deviations of the i^{th} mine's k^{th} SEG's true exposure from $\mu_{y,\,i}$ (mean exposure of the i^{th} mine), and ε_{ikj} = random deviations of the j^{th} observation for the i^{th} mine's k^{th} SEG from $\mu_{y,\,ik}$ (mean exposure of the k^{th} SEG in the i^{th} mine). The random deviations (α_i, β_{ik}, and ε_{ikj}) are assumed to be normally distributed with zero means and variances (σ^2_α, σ^2_β, and σ^2_ε, respectively). These variances are mutually uncorrelated and estimated as variance components ($S_y{}^2{}_{BM}$, $S_y{}^2{}_{BG}$, and $S_y{}^2{}_{WG}$, respectively).

The variance components for each model (Equation 5- 2 and Equation 5-3) were constructed using PROC NESTED. The statistical analyses were conducted for respirable dust and respirable silica. Significance was defined by p-values of 0.05 or lower. All analyses reported here were conducted using SAS version 9.3 (SAS Institute, Cary, NC, USA).

RESULTS

Respirable dust (RD) and respirable silica (RS) concentrations

Table 5-1 shows the percentage of samples that had RD and RS levels below the limit of detection (LOD), as measured by NIOSH 0600 and NIOSH 7500, respectively. Overall, many of the RD samples had levels lower than the LOD across all mines (ranges: 39-69%), and most of the RS samples had levels lower than the LOD (ranges: 50-79%).

Table 5-2 shows the geometric means (GM) and geometric standard deviations (GSD) of RD concentrations by SEG in all mines. The highest and lowest GM were found in the Northshore mine (0.608 mg/m^3 for *Balling drum operator*, 0.059 mg/m^3 for *Supervisor*). Except for a few SEGs, most of the GSDs were less than 3, indicating moderate variability. The highest GSD was found for *Dock man* in the Keetac mine, which only had two values that differed widely (< 0.1 and 1.1 mg/m^3). In the Northshore mine, *Office staff* was the only SEG in which all samples had RD concentrations less than the LOD. The other five mines had multiple SEGs (ranging from 6–12 SEGs) in which all measurements fell below the LOD.

Similarly, Table 5-3 summarizes the RS concentration by SEG in all mines. The highest GM (0.133 mg/m^3) was found for *Operating technician* in the Utac mine, and the highest GSD (5.22) was found for *Mining operator 1* in Northshore. Even if the RD concentration was less than the LOD for a given SEG, the RS concentration did not necessarily fall below the LOD for that same SEG, and vice versa. For instance, all samples for the *Crusher maintenance* SEG in Keetac had RD concentrations less than the LOD, but not all RS concentrations were less than the LOD. For the *Pelletizing operator* SEG in Northshore, all samples had RS concentrations less than the LOD, but not RD.

While mineralogical differences lead to different exposure levels for EMP, respirable dust and respirable silica do not differ between the two zones. Although our sampling strategy took into account the two different mineralogical zones, we found no differences between them for most of the SEGs. Therefore, distinguishing between the zones was not necessary when measuring exposure levels to respirable dust and respirable silica. To explore the differences in RD and RS exposure levels across mines, the p-values by SEG indicated differences in at least one mine across all mines (see Table 5-2 and Table 5-3, respectively). The logarithms of the estimated RD exposure levels were used in a simple one-way ANOVA model. Thirteen SEGs had at least two mines with significantly different means for RD (p-values: <0.0005). However, only seven SEGs had at least two mines with significantly different means for RS. Almost all values fell below the LOD in the *Mining* and *Office/control room operator* departments; thus, we were not able to calculate p-values for these departments. When we examined the SEGs by mine, the p-values were significantly different in many SEGs for RD, but not for RS, for which at least one mine was different from the rest of the mines (see Table 5-2 and Table 5-3).

The distribution of each dust component's concentration is illustrated by box-plots in Figure 5-1 and Figure 5-2. The RD concentrations by mine across all SEGs are displayed in Figure 5-1. The RD concentrations in all mines were below the ACGIH TLV for RD, 3 mg/m^3. The RD concentrations in the milling processes (crushing, concentration, and pelletizing) tended to be higher than those in the mining and shop processes. For many of the SEGs, the range of the box-plot is almost non-existent (e.g., *Basin operator* SEG in the Hibbtac mine), indicating that the maximum and minimum values for the SEG are similar (or equal) because most of the samples had concentrations less than the LOD. The RS concentrations by mine across all SEGs are displayed in Figure 5-2. Except for a few exceptions in the Hibbtac and Keetac mines, the concentrations of RS in the crushing and/or concentration processes were higher than the ACGIH TLV for RS, 0.025 mg/m^3, as well as higher than those measured in the rest of the processes.

Comparison of SEG variance components

A one- or two-way nested ANOVA was used to determine the distribution of variance for both RD and RS. Table 5-4 shows the between-mine (S^2_{BM}), between-SEG (S^2_{BG}), and within-SEG (S^2_{WG}) variance components for RD and RS by mine, as well as the percentage of the total variance (the sum of the components) represented by each single component. To determine the variability of the between-mine components, we compared the total variance calculated as the sum of all three components with the total variance calculated as the sum of only the between- and within-SEG components. The between-mine variability was very small for both RD and RS (3.4% and 2.1 %, respectively). In other words, there was little to no variability between the six mines. Overall, the Northshore mine had the highest S^2_{TOTAL} (0.199) for RD, a finding that is consistent with the number of large GSD (>3) for several SEGs (see Table 5-2). Hibbtac had the highest S^2_{TOTAL} (0.202) for RS, a similarly consistent finding given the number of large GSD (see Table 5-3). The S^2_{TOTAL} confirms that the GSD ($=10^{S^2_{TOTAL}}$) by SEG fall within the ranges shown in Table 5-2 and Table 5-3. The S^2_{WG} values tended to be higher than the S^2_{BG} values in most of the mines, particularly at Minntac for RD and Keetac for RS.

Comparison of percent of quartz by department

After excluding all samples with values less than the LOD for both RD and RS, we calculated coefficient of determination (R^2) between RD and RS concentrations by department using log scale data (see Figure 5-3). The value of R^2 decreased substantially from 0.51 in *Crushing* department to 0.02 in *Concentrating* department. With the exception of the *Concentrating* department, the RD and RS concentration in all departments have a significant positive relationship. The low correlation found in the *Concentrating* department may indicate that the RS concentrations are more independent from the RD as the concentrating processes remove the silica including quartz. Considerable evidence suggested that the percentage of quartz in each sample was needed for a better understanding of the taconite mining process. Therefore, we divided the RS concentration (mg/m^3) by RD concentration (mg/m^3) to calculate the percentage of quartz, which provides direct evidence for research interested in adverse health effects such as silicosis. Our results indicate that the mean percentage of quartz was approximately 14% across all departments except for the Pelletizing department, where the percentage dramatically dropped to 5% (see Figure 5-4). As mentioned earlier, the magnetic separator and flotation processes in the Concentrating department remove the silica including quartz. Therefore, the respirable dust in the Pelletizing department has a lower percentage of quartz.

DISCUSSION

Levels of RD and RS

Overall, the highest average concentrations of RD were found in the Northshore mine. The *Balling drum operator* SEG had the highest exposure level of 0.608 mg/m^3, a value much lower than the ACGIH TLV for RD (3 mg/m^3) (see Table 5-2). The Minorca mine had the lowest average concentration of RD, 0.116 mg/m^3, which is approximately 2 times lower than the average in Northshore, 0.252 mg/m^3. No single RD exposure concentration was higher than the TLV in any of the mines. Overall, between-mine differences for the RD across all SEGs were almost nonexistent (S^2_{BM}=0.005).

The *Operating technician* SEG in the Utac mine had the highest average concentration of RS, 0.133 mg/m^3, which is approximately 5 times greater than the ACGIH TLV for RS (0.025 mg/m^3). The highest average concentration of RS (0.025 mg/m^3) was found in Hibbtac and the lowest (0.010 mg/m^3) in Keetac. This pattern was not consistent with that of the RD concentrations. The exceptions were in the mines Hibbtac and Keetac, in which the highest concentrations of RS in all mines were found in the crushing departments. For instance, the RS concentration in the crushing department ranged from 0.036 mg/m^3 for the *Crusher maintenance* SEG in Minntac to 0.133 mg/m^3 for the *Operating technician* SEG in Utac (Table 5-3). More specifically, the magnetic separator and flotation processes in the concentrating department may have reduced the levels of RS significantly. The highest concentration of RS in Keetac was found in the *Shop* (*mobile*) SEG; the *Crusher maintenance* SEG had a similar concentration. The concentrations of RS were often greater than the ACGIH TLV for RS. In addition, the RS concentrations were higher at the beginning of the taconite process, or in the mining and crushing departments. This pattern was consistent across all mines. Sheehy (1986) found that the greatest potential for overexposure to respirable silica occurred in the mining (drill operator), crushing (crusher helper), and concentrating (primary attendant, concentrator laborer) departments. However, it was unclear whether the specific job tasks performed by the concentrator laborer occurred before or after the magnetic separator process.

Between-mine variability by process

The taconite mining and milling processes are very similar in all mines; thus, the levels of exposure to RD and RS during these processes display no significant differences across mines. In the pit, the processes of drilling and loading taconite rock into the crusher are also very similar across mines. Given that many mining jobs are performed in a cab (e.g., driller, truck driver, shovel operator), the level of exposure to RD and RS completely depends upon the ventilation system in the operating equipment. There are no major differences between mines for these jobs; however, there are differences between the equipment used for each job. For instance, the ventilation filters must be checked more often in some vehicles (e.g., bulldozer or grader) than others (e.g., truck or loader). Also, some small vehicles do not have a ventilation system.

Inside the taconite plants, the conveyance of rocks through the primary crusher is, in general, a dry process that generates significant RD and RS across mines. A small fraction of the crushed rock is mixed with water and ground to a powder-like substance in the rod and ball mills. After this milling process, the amount of RD and RS is significantly reduced because of the wet materials. The amount of solid recovered depends on the type of magnetic separator. A cobber recovers 55-60% of solids containing iron, the rougher recovers 75%, and the finisher recovers 90%. All taconite mines use these three types of magnetic separators in sequence. The magnetic separating process is slightly different between mines, as it depends on the size of the particles and the content of the ore across the mines. For instance, dry cobbers are used in the Northshore mine, while the rest of the mines use a wet magnetic separator, which generates less dust in the *Concentrating* departments.

Between-SEG vs. within-SEG variability

When only the between-SEG (S^2_{BG}) and within-SEG (S^2_{WG}) components were considered, the S^2_{BG} variance was slightly larger because it encompassed the between-mine (S^2_{BM}) variance as well. The S^2_{BG} for RD did not differ across mines; however, a significantly smaller S^2_{BG} for RS was observed in the Keetac mine (see Table 5-4). The S^2_{TOTAL} for Keetac was 0.063, more than three times smaller than that for other mines (for example, 0.202 in Hibbtac). Many of the SEGs had a relatively consistent level of RS exposure, especially between the departments in Keetac (see box-plot in Figure 5-2). However, the RS levels in the *Crushing* department varied within a minimum range in Keetac, while the levels in the *Crushing* department in the other mines varied much more. In contrast, the levels of RS in all other departments varied less across mines. Interestingly, all of the RD concentrations for the *Crushing* department in Keetac fell below the LOD because the crusher operator works in a crusher control operating room at that mine. The work environment of the crusher operator in Northshore is totally different. In Northshore, the crusher operators are more exposed to the work done in the nearby crusher building, while the crusher operator in Keetac works in a cleaner environment.

The sampling was conducted in Keetac in June 2010, which was 21 out of 30 rainy days (Minnesota Department of Natural Resources, 2013), a factor that also contributed to the smaller S^2_{TOTAL}. The between-SEG variability for the Minntac mine was much smaller (0.028) than that of Keetac. However, Minntac is the largest mine, employing approximately 1400 workers and having a capacity of 14.7 million tons of ore, so the job tasks are much more specific than in other mines. For instance, the shop mobile in Minntac has two different shops: the “heavy equipment shop” assembles, maintains, and repairs all machines, while the “machine shop” fixes and customizes the machines after the heavy equipment shop.

The Keetac mine is located at the end of the western zone of the Mesabi Iron Range and is the farthest mine from Northshore. Therefore, the characteristics of the taconite rock in Keetac and Northshore are totally different. Moving from Keetac to Northshore across the iron range, the taconite rock changes from soft to hard. Thus, Keetac only needs a primary crusher, because the taconite rock is much softer, while Northshore has a primary crusher in Babbit and a secondary crusher in Silver Bay. These taconite characteristics also might contribute to the lower RS concentrations in the mining process in Keetac.

CONCLUSIONS

Respirable dust (RD) and respirable silica (RS) levels do not significant between the two zones, although the geological differences between the zones are reflected in the EMP exposures. No single RD exposure concentration was higher than the ACGIH TLV in any of the mines, whereas the concentrations of RS were often greater than the TLV for RS. The highest concentrations of RS were found in the *Crushing* departments for all mines, with few exceptions. The processes in the *Concentrating* department may have reduced the levels of RS significantly, as well as lowered the percentage of quartz in RD in the *Pelletizing* department. There was little to no variability between the six mines for either RD or RS exposures because the taconite mining and milling processes are very similar across all mines. The between-SEG variability for RD did not differ across mines; however, a significantly smaller between-SEG variability for RS was observed in the

Keetac mine. This finding could result from the characteristics of the taconite rock, seasonal effects during sampling, or the tasks assigned for each job.

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Table 5-1 Number of personal samples and percent of samples < limit of detection (LOD) by mine and mineralogical zone

Zone	**Mine**	**Workers**	**Number of RD/RS Samples**	**% <LOD of RD** [a]	**% <LOD of RS** [b]
Eastern	Northshore	56	161	39	50
Western	Hibbtac	34	101	48	50
	Utac	38	113	47	56
	Keetac	34	100	69	79
	Minntac	48	139	68	65
	Minorca	22	65	74	72
Total		232	679		

[a] Personal respirable dust (RD) samples analyzed by NIOSH 0600.

[b] Personal respirable silica (RS) samples analyzed by NIOSH 7500.

Table 5-2 Summary statistics of respirable dust for each SEG measured in all mines (GM unit: mg/m3)

Department	**SEG**	**Northshore**		**Hibbtac**		**Utac**		**Keetac**		**Minntac**		**Minorca**		**P-values**[e]
		GM	**GSD**	**GM**	**GSD**	**GM**	**GSD**	**GM**	**GSD**	**GM**	**GSD**	**GM**	**GSD**	
Mining	Basin operator	.[c]	.	<LOD[d]	<LOD	0.088	2.10	<LOD	<LOD	<LOD	<LOD	.	.	NA
	Mining operator 1	0.090	3.23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	NA
	Mining operator 2	0.083	1.96	0.184	3.00	<LOD	<LOD	0.065	1.49	0.092	1.98	<LOD	<LOD	0.1380
	Rail road	0.095	4.14	.	.	.	.	.	.	.	.	.	.	NA
Crushing	Crusher maintenance	0.280	2.39	0.135	1.57	0.279	2.11	<LOD	<LOD	0.175	2.50	0.164	1.14	**0.0095**
	Crusher operator	0.225	2.53	0.355	2.42	0.099	1.66	<LOD	<LOD	0.186	2.78	0.201	1.19	**0.0387**
	Operating technician	0.225	1.21	.	.	0.563	1.73	.	.	.	.	.	.	**0.0064**
Concentrating	Concentrator maintenance	0.252	3.75	0.157	1.76	0.252	1.36	0.078	1.84	0.087	1.78	0.190	3.24	0.1206
	Concentrator operator	0.093	1.70	0.172	1.83	0.136	1.68	<LOD	<LOD	0.122	2.47	<LOD	<LOD	**0.0140**
Pelletizing	Balling drum operator	0.608	1.86			0.196	1.63	0.284	1.56	0.246	1.53	0.161	2.62	**0.0028**
	Dock man	0.258	2.39	0.172	2.81	<LOD	<LOD	0.235	8.90	0.092	2.07	<LOD	<LOD	**0.0377**
	Furnace operator	0.513	1.67			0.420	2.22	0.126	1.94	0.171	2.54	0.095	2.02	**0.0029**
	Pelletizing maintenance	0.593	1.83	0.155	1.82	0.243	1.36	0.186	2.81	0.134	2.04	0.086	1.54	**0.0031**
	Pelletizing operator	0.323	1.49	0.330	1.91	0.201	1.34	0.285	1.35	0.169	1.87	0.346	1.65	0.1678
Shop(mobile)[a]	Boiler technician	.	.	.	.	<LOD	<LOD	.	.	<LOD	<LOD	.	.	NA
	Carpenter	.	.	0.091	2.23	.	.	<LOD	<LOD	0.139	2.62	.	.	0.6130
	Electrician	0.097	1.92	<LOD	<LOD	0.102	2.17	0.157	2.12	0.069	1.41	<LOD	<LOD	**0.0469**
	Lubricate technician	0.068	1.57	.	.	0.163	1.89	.	.	.	.	<LOD	<LOD	**0.0065**
	Maintenance technician	0.152	2.52	0.125	2.84	<LOD	<LOD	0.159	2.34	0.180	5.26	0.212	2.99	0.7348
	Pipefitter/Plumber	.	.	.	.	0.103	1.72	.	.	<LOD	<LOD	.	.	**0.0316**
	Repairman	.	.	0.175	1.79	.	.	.	.	0.077	1.99	<LOD	<LOD	**0.0290**
	Supervisor	0.059	1.70	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.061	1.60	<LOD	<LOD	0.8962
Shop (stationary)[b]	Auto mechanic	0.113	2.24	0.111	2.12	0.075	1.62	0.103	1.79	0.062	1.40	0.075	1.72	0.4767
	Lab analyst	0.106	1.70	0.259	1.23	<LOD	<LOD	0.069	1.31	<LOD	<LOD	<LOD	<LOD	**0.0001**
	Warehouse technician	0.064	1.72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.070	1.33	<LOD	<LOD	0.5491
Office/ Control room	Control room operator	0.080	1.99	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	NA
	Office staff	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.076	2.05	<LOD	<LOD	NA

[a] Shop (mobile) refers to those SEGs whose work is more likely done in multiple places in the plants.

[b] Shop (stationary) refers to those SEGs whose work is more likely done in a single workplace.

[c] . indicates no measurement.

[d] <LOD indicates all samples containing below LOD.

[e] Numbers in **boldface** indicate statistically significant differences among mines (P<0.05).

Table 5-3 Summary statistics of respirable silica for each SEG measured in all mines (GM unit: mg/m3)

Department	SEG	Northshore		Hibbtac		Utac		Keetac		Minntac		Minorca		P-values[e]
		GM	GSD	GM	GSD	GM	GSD	GM	GSD	GM	GSD	GM	GSD	
Mining	Basin operator	.[c]	.	<LOD[d]	<LOD	0.012	3.19	<LOD	<LOD	0.007	1.45	.	.	0.2853
	Mining operator 1	0.017	5.22	<LOD	<LOD	0.008	1.62	<LOD	<LOD	0.006	1.37	0.012	2.51	0.2005
	Mining operator 2	0.010	1.61	0.067	2.76	0.013	2.47	0.007	2.01	0.008	1.60	<LOD	<LOD	**0.0004**
	Rail road	0.010	4.84	.	.	.	.	.	.	.	.	.	.	NA
Crushing	Crusher maintenance	0.032	3.57	0.014	2.02	0.038	1.36	0.012	2.54	0.036	1.30	0.027	1.20	0.1014
	Crusher operator	0.061	2.40	0.066	2.21	0.028	1.44	<LOD	<LOD	0.033	2.02	0.052	1.09	**0.0003**
	Operating technician	0.045	1.22	.	.	0.133	1.56	.	.	.	.	.	.	**0.0007**
Concentrating	Concentrator maintenance	0.016	1.80	0.034	2.59	0.024	1.27	0.011	2.13	0.015	1.55	0.018	3.17	0.1979
	Concentrator operator	0.009	1.83	0.057	1.48	0.018	1.71	0.010	1.79	0.034	2.23	0.016	1.25	**<0.0001**
Pelletizing	Balling drum operator	0.009	1.76	.	.	0.008	1.57	0.009	1.65	0.007	1.47	<LOD	<LOD	0.6009
	Dock man	0.008	1.59	0.011	1.73	<LOD	<LOD	0.011	3.21	<LOD	<LOD	<LOD	<LOD	0.1260
	Furnace operator	0.007	1.59	.	.	0.010	2.50	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.4598
	Pelletizing maintenance	0.007	1.73	0.009	1.56	0.011	1.76	0.009	1.86	<LOD	<LOD	<LOD	<LOD	0.2500
	Pelletizing operator	<LOD	<LOD	0.010	1.69	<LOD	<LOD	0.008	1.65	0.008	1.72	<LOD	<LOD	0.2044
Shop(mobile)[a]	Boiler technician	.	.	.	.	<LOD	<LOD	.	.	0.006	1.39	.	.	NA
	Carpenter	.	.	<LOD	<LOD	.	.	<LOD	<LOD	0.019	3.48	.	.	NA
	Electrician	0.012	2.25	0.008	1.62	0.009	1.80	0.013	2.50	0.011	1.87	<LOD	<LOD	0.5163
	Lubricate technician	0.010	1.94	.	.	0.020	1.83	.	.	.	.	0.008	1.78	0.0534
	Maintenance technician	<LOD	<LOD	0.010	1.94	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.014	2.31	**0.0446**
	Pipefitter/Plumber	.	.	.	.	0.009	2.17	.	.	0.011	2.18	.	.	0.7187
	Repairman	.	.	0.048	1.36	.	.	.	.	0.010	1.88	0.010	1.61	**0.0002**
	Supervisor	0.011	2.59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.008	2.05	<LOD	<LOD	0.5169
Shop (stationary)[b]	Auto mechanic	0.006	1.52	0.016	2.94	<LOD	<LOD	0.011	1.88	0.009	2.48	<LOD	<LOD	0.1216
	Lab analyst	0.012	1.75	0.032	2.42	<LOD	<LOD	0.007	1.46	<LOD	<LOD	<LOD	<LOD	**0.0006**
	Warehouse technician	0.008	1.80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.008	1.56	<LOD	<LOD	0.4179
Office/ Control room	Control room operator	0.008	1.76	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.007	1.42	0.6488
	Office staff	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.008	2.37	<LOD	<LOD	NA

[a] Shop (mobile) refers to those SEGs whose work is more likely done in multiple places in the plants.

[b] Shop (stationary) refers to those SEGs whose work is more likely done in a single workplace.

[c] . indicates no measurement.

[d] <LOD indicates all samples containing below LOD.

[e] Numbers in **boldface** indicate statistically significant differences among mines (P<0.05).

Table 5-4 Between-mine, between-SEG, and within-SEG variance components across mine and by mine for respirable dust and respirable silica

Classification	**Mine**	**Subject**	**Sample**	**Total** [b]	**BM**		**BG**		**WG**	
				S^2_{TOTAL}	S^2_{BM}	**%**	S^2_{BG}	**%**	S^2_{WG}	**%**
RD	All [a]	232	679	0.144	0.005	3.4	0.063	43.5	0.077	53.1
	All	232	679	0.144			0.067	46.6	0.077	53.4
	Northshore	56	161	0.199			0.093	46.7	0.106	53.3
	Hibbtac	34	101	0.133			0.054	40.6	0.079	59.5
	Utac	38	113	0.149			0.097	65.0	0.052	35.0
	Keetac	34	100	0.105			0.049	47.3	0.055	52.7
	Minntac	48	139	0.115			0.028	24.5	0.087	75.5
	Minorca	22	65	0.093			0.051	54.7	0.042	45.3
RS	All [a]	232	679	0.147	0.003	2.1	0.071	48.4	0.073	49.5
	All	232	679	0.147			0.074	50.3	0.073	49.7
	Northshore	56	161	0.184			0.072	39.4	0.111	60.6
	Hibbtac	34	101	0.202			0.135	66.7	0.067	33.3
	Utac	38	113	0.177			0.119	67.0	0.058	33.0
	Keetac	34	100	0.063			0.001	1.4	0.062	98.6
	Minntac	48	139	0.114			0.050	43.6	0.064	56.4
	Minorca	22	65	0.090			0.056	61.5	0.035	38.5

[a] This variance components include between-mine.

[b] Total variance components (S^2_{TOTAL}) are sum of partitioned BM, BG, and WG variance components.

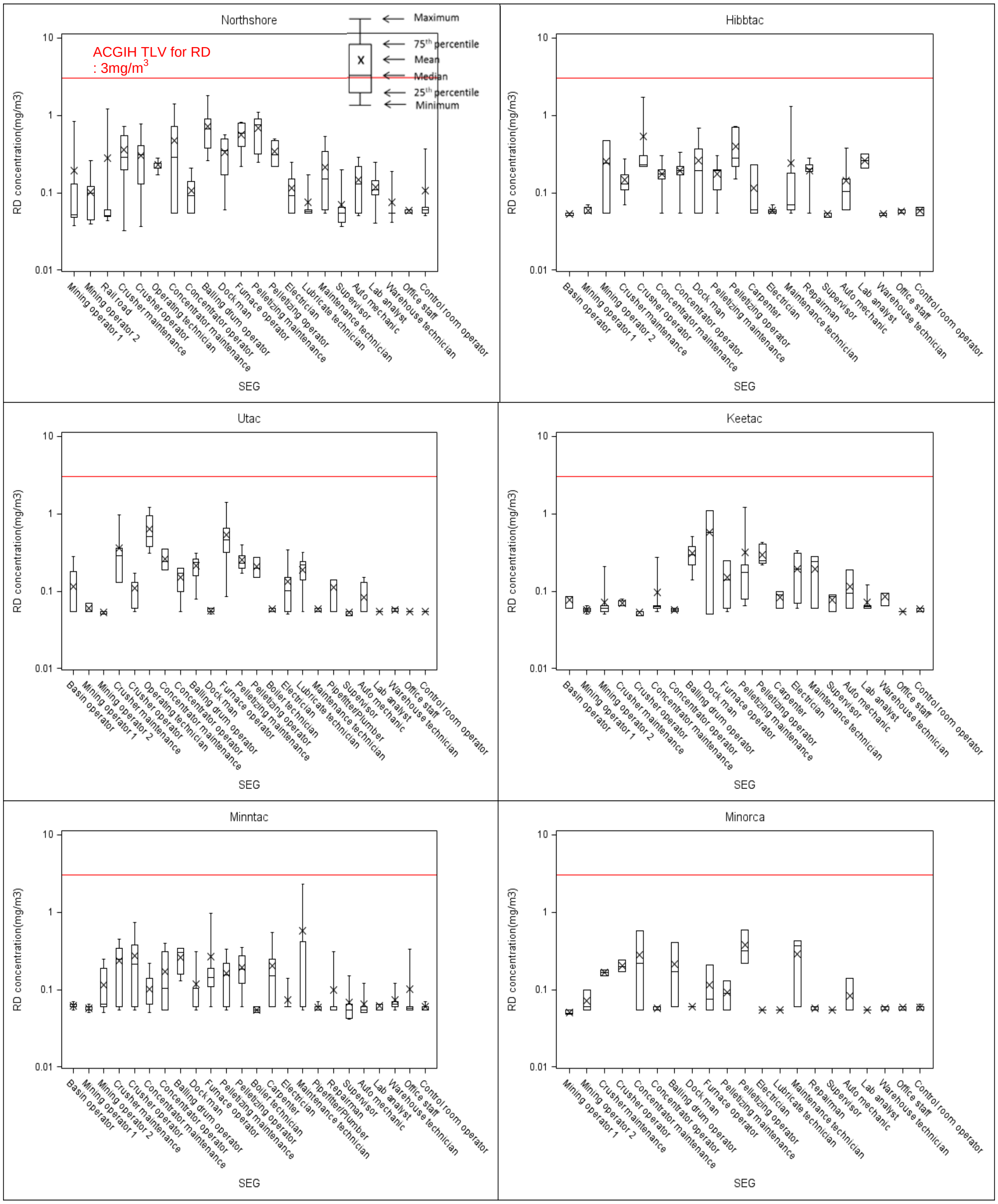

Figure 5-1 Box plots for respirable dust for each SEG in all mines (the horizontal line indicates the ACGIH TLV for RD = 3 mg/m^3)

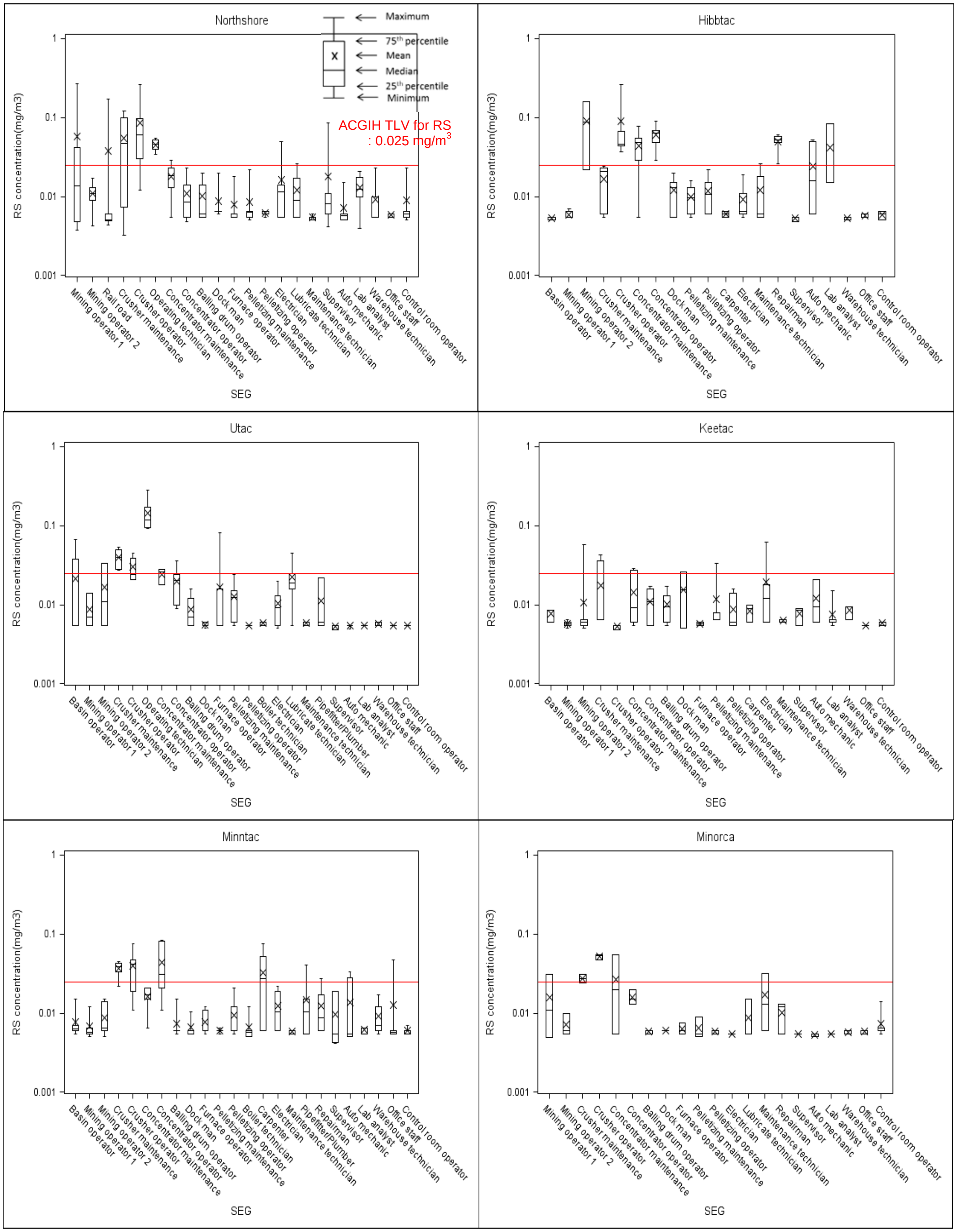

Figure 5-2 Box plots for respirable silica for each SEG in all mines (the horizontal line indicates the ACGIH TLV for RS = 0.025 mg/m^3)

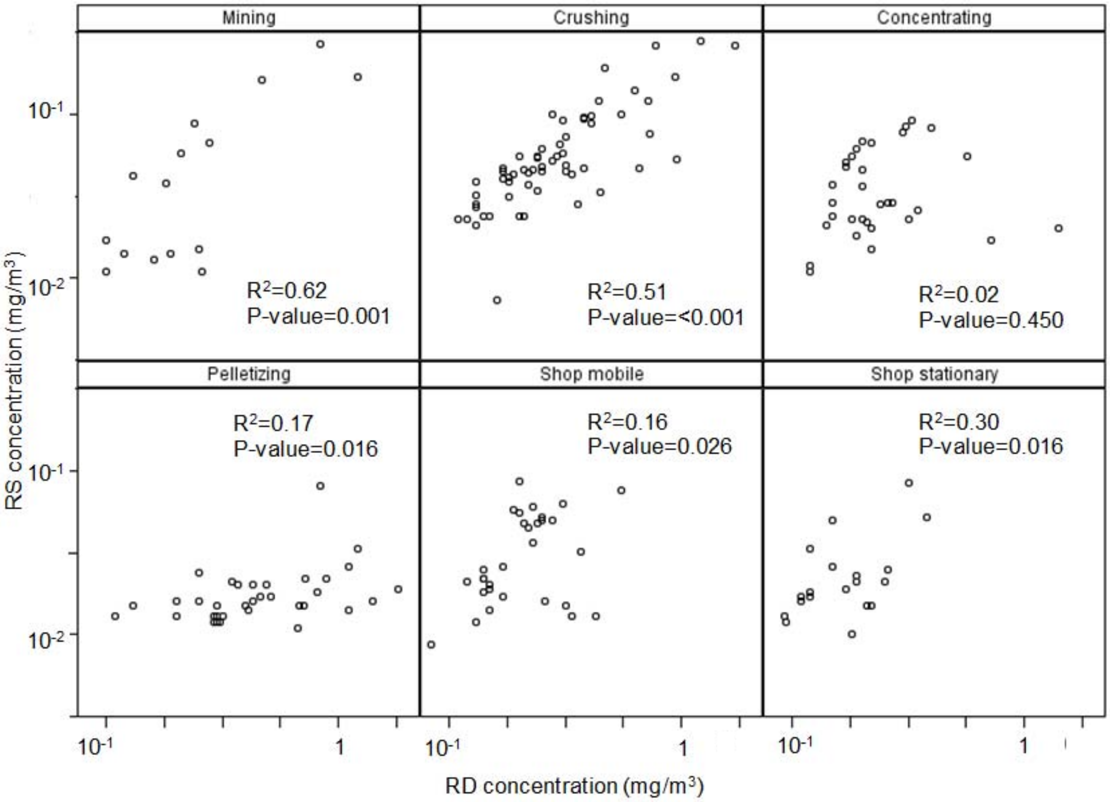

Figure 5-3 Coefficient of determination (R^2) between respirable dust and respirable silica

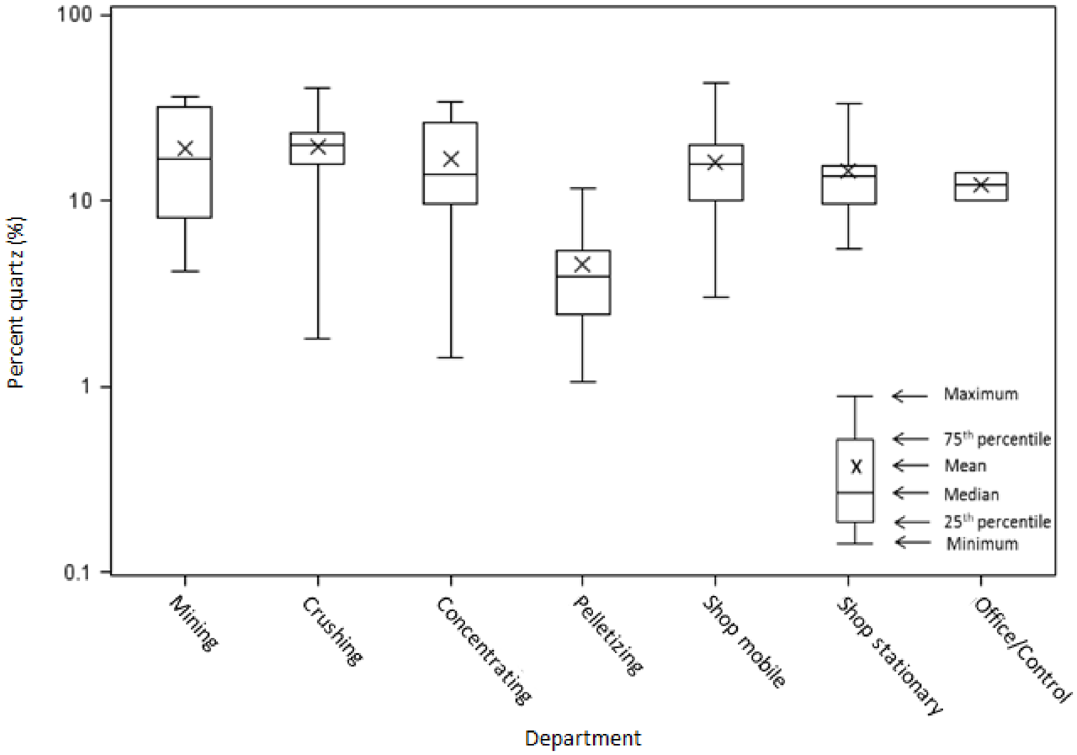

Figure 5-4 Percent of quartz in respirable dust samples by department

Chapter 6 Conclusions And Future Directions

Overall Conclusions

The purpose of this research was to describe a multi-faceted approach to assessing present-day and historical exposure levels to EMP (elongate mineral particles), respirable dust, and respirable silica in the taconite mining industry in northeastern Minnesota. This effort was part of a larger epidemiological study assessing the respiratory health effects of exposure to components of taconite dust. The main goals of the occupational exposure assessment were to accurately group workers into SEGs (similar exposure groups) and to assess the present-day and historical exposures to the taconite dust components that are health-relevant. There are four key areas of research within this dissertation, each of which is the subject of a paper that has either been submitted or will be soon. The primary conclusions of these four papers are:

1. Insufficient quantitative exposure data have hampered investigations of the relationship between cumulative exposures to EMP in taconite dust and adverse health effects. Specifically, no research on exposure to taconite dust, which includes EMP, had been conducted since 1990. The first paper describes a comprehensive assessment of present-day exposures to total and amphibole EMP in the taconite mining industry. SEGs were established to assess present-day exposure levels and buttress the sparse historical data. Personal samples were collected to assess the present-day levels of worker exposures to EMP at six mines in the Mesabi Iron Range. The samples were analyzed using NIOSH

methods 7400 and 7402. For many SEGs in several mines, the exposure levels of total EMP were higher than the permissible exposure limit (PEL) for EMP. 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. The concentrations of amphibole EMP were well controlled across all mines and were much lower than the concentrations of total EMP, indicating that amphibole EMP are not major components of taconite EMP. Two different approaches were used to evaluate the variability of exposure between-SEGs, between-workers, and within-workers. The related constructs of contrast and homogeneity were calculated to characterize the SEGs. Contrast, which is a ratio of between-SEG variability to the sum of between-SEG and between-worker variability, provides an overall measure of whether there are distinctions between the SEGs. Homogeneity, which is the ratio of the within-worker variance component to the sum of the between-worker and within-worker variance components, provides an overall measure of how similar exposures are for workers within an SEG. Using these constructs, it was determined that the SEGs are formed well enough when grouped by mine for both total and amphibole EMP to be used for epidemiological analysis.

2. Different dimensions of EMP have been proposed as being relevant to respiratory health end-points such as mesothelioma and lung cancer. In the second paper, a methodology for converting personal EMP exposures measured using the NIOSH

7400/7402 methods to exposures based on different size-based definitions has been proposed and illustrated. Air monitoring of 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 according to the different definitions were estimated by (1) a linear regression model based on data across all locations and (2) a location-specific ratio that converted NIOSH 7400/7402 personal measurements into exposures based on the different definitions. The highest fractions of EMP concentrations were found for the size defined as 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. However, similar exposure patterns were observed based on the different EMP size definitions, consistent with the high degree of correlation between the exposure levels. Therefore, 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.

3. The goal of the third paper was to develop a matrix of exposure concentrations as a function of SEG, mine, and year for a mesothelioma case-control study and to estimate cumulative exposures to total and amphibole EMP as defined by NIOSH in the Mesabi Iron Range. Historical exposure data for mining jobs in the Iron Range are sparse; however, by combining comprehensive present-day exposure levels with the minimal historical data, we generated exposure matrices that provide estimates of exposure levels. Historical EMP measurements (n=682) were extracted from databases maintained by two mining companies and the Mine Safety and Health Administration. Present-day EMP concentrations (n=1280) were monitored and analyzed using NIOSH 7400 for total EMP and NIOSH 7402 for amphibole EMP. Using the measured data and regression model estimates, we reconstructed the exposure for each SEG by mine between 1955-2010. The resulting exposure matrix was combined with employment history to estimate a cumulative exposure for each worker in our cohort. Our exposure matrix contains estimates of total EMP exposure for 28 SEGs and 7 mines over 56 years. Based on the cumulative distribution, ~60% of workers in the mesothelioma case-control study had lifetime average total EMP exposure levels greater than the occupational exposure limit (NIOSH REL: 0.1 EMP/cm^3), calculated using a time-varying exposure model, and ~10% of workers had EMP levels exceeding the REL, calculated using a constant exposure model. The average total EMP exposure concentration for the entire cohort population was calculated using both models. The cumulative EMP exposure concentration for the control group and all

(both control and case) workers were not markedly different, as the number of workers in the control group was much larger than the case group. Similarly, the cumulative distributions were not markedly different for the case and control groups.

4. The potential relationship between silica-containing respirable dust and health risks in northeastern Minnesota's Mesabi Iron Range has raised concerns in the taconite mining industry and the surrounding communities. Therefore, our study assessed the present-day levels of exposure to respirable dust (RD) and respirable silica (RS) in the taconite mines and evaluated how the mining process influences the exposure levels. Personal samples (n=679) were collected to assess the present-day exposure levels of workers to RD and RS at six mines in the Mesabi Iron Range. The RD and RS concentrations were calculated using NIOSH 0600 and NIOSH 7500, respectively. Between-mine, between-SEG, and within-SEG components of variability for RD and RS exposures were also estimated using a one- or two-way nested random-effects ANOVA model. The concentrations of RD in all mines fell below the ACGIH TLV for RD. With a few exceptions, the concentrations of RS in the crushing and concentrating processes were higher than those measured in the other mining processes, as well as higher than the ACGIH TLV for RS. The magnetic separation and flotation processes in the concentrating department may have reduced the levels of RS significantly, as well as lowered the percentage of quartz in RD in the pelletizing department. There

was little to no variability between the six mines for either RD or RS exposures. The between-SEG variability for RD did not differ across mines; however, significantly smaller between-SEG variability for RS was observed in the Keetac mine. This finding could be the result of the characteristics of the taconite rock, seasonal effects during sampling, or the tasks assigned for each job.

Limitations of Study and Future Directions

This research study provides a comprehensive set of present-day EMP exposure levels, data that previously was non-existent. Therefore, future research could focus on performing additional analyses or extensions of the current study, including

1. An analysis of personal samples using ISO 13794 TEM

   In this work, personal exposure assessment was conducted using standard methods NIOSH 7400 and NIOSH 7402. While NIOSH 7400 using PCM can be used to count the number of EMP, it cannot differentiate between asbestiform and non-asbestiform EMP. NIOSH 7402 using TEM can be used to identify EMP that are amphiboles or chrysotile. However, it does not distinguish between asbestiform and non-asbestiform EMP either. These methods are limited by the absence of detailed EMP dimensions (lengths and widths), which are needed to investigate different size-based definitions. Therefore, a future study could analyze all archived personal samples using ISO 13794 TEM using different size-based EMP definitions as well as analysis of some samples by Scanning Electron Microscopy (SEM) to study morphology. This investigation would have two outcomes: (1) an assessment of present-day and historical exposures by SEG based on personal samples measured using alternative size-based EMP definitions; (2) the identification of methodological differences in each exposure matrix between the indirect (applying conversion factors to personal samples using area measurements) and direct (using personal samples directly) methods; (3) the relationship between ISO 13794 and SEM analysis will yield a greater understanding of whether and how only dimensional information can be used to infer morphological information.

2. The reconstruction of exposures using other mathematical methods

In the current study, since the historical data, especially for EMP, are sparse, the estimates were strongly influenced by the present-day data. Therefore, a second study could follow a different and complementary approach to reconstructing the historical exposure levels: mathematical modeling. This approach would require additional information such as dust generation rates, ventilation rates, and production rates, engineering controls, administrative controls, and existing PPE, and worker activity patterns. Exposure modeling, supplemented by Bayesian methods that can synthesize sparse data and priors based on the exposure models, can be used to reconstruct historical exposures. The new approach may help improve estimates of exposure levels to taconite dust components for each SEG by mine when the exposure matrix is reconstructed.

3. The measurement of area exposure levels by process

For the area measurements, we set up air monitoring devices, including MOUDI (Micro Orfice Uniform Deposit Impactor), adjacent to the places in which employees were working. Therefore, the area measurements are characterizations of EMP based on SEGs (similar exposure groups). The purpose of SEG usage is to understand exposures to taconite dust by workers' tasks, job titles, and locations. However, in some instances the exposure characterization information can be confounded by the process, and the extent of this confounding is unclear. Within an SEG, different processes can cause different levels of exposure to taconite dust. For instance, several major processes take place in the location corresponding to the Pelletizing operator SEG: (1) The disc filter process in the Pelletizing operator location is relatively wet, but generates respirable dust because the powdered materials are screened for moisture content less than 10%. However, this process is less likely to generate respirable silica because the prior process (Concentrating) removes respirable silica during the magnetic separator process. (2) When materials such as bentonite or limestone are added in the mixer in the Pelletizing operator location, the levels of exposure to respirable silcia may be increased. Area measurements can be set up by process within the SEGs for a better understanding of exposure levels. Our current area measurements only covered 81 locations across all mines. This coverage can be increased using a detailed area sampling strategy by process to also cover all locations missing from the current area measurements.

4. The measurement of exposures to other contaminants related to adverse health effects

   EMP and respirable dust containing silica were the taconite dust components of interest in this study, in which we were looking at respiratory disease. EMP are related to mesothelioma and lung cancer and respirable silica is related to silicosis and lung cancer. However, there are other contaminants of concern, such as hexavalent chromium, diesel particulate matter, and heavy metals (e.g., iron and nickel), also relevant to respiratory and cardiovascular health in the taconite mining industry. Future research could measure exposures to these other contaminants for a full investigation of respiratory and cardiovascular health in the epidemiological study.

5. A comparison of respirable dust between personal and area sampling

   The only area monitoring data reported in this dissertation were collected using MOUDI. Besides MOUDI, we measured the mass, number, and surface area of particles in the lung using various other monitoring devices. Among these devices, the DustTrak (Model 8520, TSI Inc., Shoreview, MN, USA) was used to monitor aerosol mass respirable fraction concentration (mg/m3). Since respirable dust was one of the personal sampling agents considered along with EMP, we could obtain more accurate data on the respirable fraction concentration as well as directly compare it with the respirable dust concentration from the real-time area monitoring data.

6. A comparison of differences between analyses

Personal and area samples for EMP and respirable dust containing silica were analyzed by a contract laboratory. Although the laboratory was AIHA (American Industrial Hygiene Association) accredited and well-established, results from its analysis could be compared with analyses of other laboratories. This additional comparison might reveal inter- and intra-laboratory analytical bias along with sampling bias.

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