Summary Report:

Environmental Particulate Matter Characterization

by

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ABSTRACT

The NRRI characterization studies provide physical (size and shape), mineralogical, chemical, geological, geographical, and historical context to the findings of the University of Minnesota's School of Public Health (SPH) and the University of Minnesota Medical School (UMMS). The SPH and UMMS findings (Finnegan and Mandel, 2014) showed that mesothelioma is associated with working longer in the taconite industry. However, the SPH and UMMS investigators "…were not able to state with certainty that the association with EMPs and mesothelioma was related to the ore dust or to the use of commercial asbestos or both."

The NRRI findings indicate the following:

1) Low concentrations of PM_{10}, $PM_{2.5}$, and EMPs in Mesabi Iron Range community air.
2) Elemental iron concentrations in MIR communities were similar to elemental iron concentrations in background sampling locations when taconite mines/plants were inactive. When taconite mines/plants were active, the elemental iron concentrations within communities were found to be statistically higher.
3) Mineralogically and morphologically, the EMPs identified in MIR communities and taconite processing plants were dominated by particles that did not fit the "countable"/"covered" classification criteria. Of the 145 "covered" EMPs identified within the six MIR taconite processing plants, a total of 8 were "countable" (NIOSH, 2011), representing 1.1% of the total number of EMPs, out of 691 total. These EMPs were detected in two taconite plants (seven in one plant and one in another); no other "countable"/"covered" EMPs were detected in the other four plants.
4) The lake sediment study returned similar results, in which 4 of the study's 790 identified EMPs found in the lake sediment samples met the "countable"/"covered" classification.
5) In comparison to the NIOSH standard, for countable particles, the results from this study show that the community air has significantly lower amounts than the standard.
6) Only one plant and two areas in this plant had countable EMPs above the NIOSH benchmark.
7) The highest particulate matter found was for the Minneapolis reference site in comparison for the Range communities and the other two reference sites.
8) The use of MOUDI sampling techniques is a good method for better understanding not only what is in the air, but also the size of the particles that are in the air.
9) Study of lake sediment can be used to interpret some of the impacts of past industrial activities and to gain a better understanding of the impact of local geology.

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ACKNOWLEDGEMENTS

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The NRRI's Particle Study project team extends additional thanks to several individuals for their contributions and input, starting with Anda Bellamy, editor and publications specialist. Anda's editing, organizing, and assembly of the team's final reports was a huge task. Next is Tamara Diedrich, who initiated and oversaw many key components of the NRRI's early project work. We also wish to thank the members of the NRRI Particle Study Science Advisory Board (SAB), composed of D. Wayne Berman (AEOLUS Inc.), Gregory Meeker (United States Geological Survey – USGS), Paul Middendorf (National Institute of Occupational Safety and Health – NIOSH), and Daniel Vallero (U.S. Environmental Protection Agency – USEPA), who provided guidance, recommendations, and constructive criticism during the course of this work; their collective inputs were of significant assistance to the NRRI team, and their advisory role was much appreciated. A deep expression of gratitude is given to Professor Emeriti Dale Lundgren (University of Florida) and the late Virgil Marple (University of Minnesota), as well as Bernard Olson (University of Minnesota), Bryan Bandli (University of Minnesota Duluth), and Francisco Romay (MSP Corporation – MOUDI) for the key roles they played advising the technical aspects of our aerosol science-based experimental designs, sampling, and particulate matter (PM) analysis and characterization. Special thanks are also extended to former NRRI team members Devon Brecke and Megan Schreiber, who played important sample collection and laboratory support roles, and to the NRRI's Meijun Cai for her statistical review and analysis of the project's community, background, and taconite facility datasets. We would also like to thank the dedication and hard work of all the student workers who contributed to this effort over the years, including: Alanna Schwanke, Chris Bicknese, April Severson, Allison Severson, Stuart Kramer, and Matt Chaffee. Minnesota agencies are also thanked for providing feedback to the NRRI's work. They include the MPCA, Minnesota Department of Health (MDH), Minnesota Department of Natural Resources (MDNR), and the Department of Iron Range Resources & Rehabilitation (DIRRR).

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UNIVERSITY OF MINNESOTA, DULUTH – NATURAL RESOURCES RESEARCH INSTITUTE (NRRI) ENVIRONMENTAL PARTICULATE MATTER CHARACTERIZATION

Taconite workers on the Mesabi Iron Range (MIR) in northeastern Minnesota have displayed a higher than normal incidence of the rare disease Mesothelioma (MDH, 2003; 2007). To better-understand this higher level of occurrence, the Minnesota Legislature funded the $4.9-million-dollar Minnesota Taconite Workers Health Study (MTWHS). Initiated in 2008, this five-component study was a collaborative effort between the University of Minnesota's School of Public Health (SPH), the University of Minnesota Medical School (UMMS), and the University of Minnesota Duluth's Natural Resources Research Institute (NRRI). The University's School of Public Health and Medical School performed the study's four health-related components (Finnegan and Mandel, 2014). The study's fifth component, "Environmental Study of Airborne Particulate Matter," was conducted by the NRRI and is summarized in this Executive Summary Report. The study comprised physical, mineralogical, and chemical characterization of airborne particulate matter (PM) in MIR taconite processing facilities and selected MIR communities. Respirable mineral PM of a specific size, shape, and mineralogy – referred to as elongate mineral particles (EMPs) – were of special interest. Ambient aerosol PM samples were collected from five MIR communities, three Minnesota background locations, and the six operating taconite processing plants located on the MIR. Components of this PM characterization included: gravimetric analysis, mineralogical identification, mineralogical concentration evaluations, PM morphological characterizations, and PM chemical characterizations.

The fundamental question addressed by this study is, "What is in the air?" Additionally, a characterization of EMPs from age-dated lake sediments in two MIR lakes was performed in an effort to obtain historical data regarding the characteristics of past PM conditions; in a general sense, to investigate what *had been* in the air. Consequently, the research effort focused on providing answers to the following underlying questions:

- How much PM is in the air?
- What is the size distribution of the PM in the air?
- What are the physical characteristics of the PM in the air?
- What are the mineralogical characteristics of the PM in the air?
- What are the chemical characteristics of the PM in the air? and
- What historical trends in PM mineralogy, physical characteristics, and chemical composition can be identified from studies of dated sedimentary deposits in lakes located in close proximity to taconite operations on the MIR?

Therefore, the NRRI's environmental study of airborne PM was necessarily a characterization study, quantifying the presence of EMPs as well as the mass concentration, mineralogy, and chemistry of the PM. Six separate reports resulted, including:

1. **Standard Operating Procedures (SOP) Report** (Monson Geerts et al., 2019a), which describes methodologies developed through a Science Advisory Board (SAB) and other experts in the field of aerosol science, utilized for all sampling and analytical procedures during the course of the study, as well as the results of experiments that led to the implementation of these procedures. In addition, the SOP contains a "Glossary" that describes the terminology used throughout this series of reports based on the correct usage in relevance to geologic, mineralogic, and morphologic context;
2. **MIR Taconite Plant Gravimetric Report** (Monson Geerts et al., 2019b), which characterizes mass PM concentrations collected at various process locations within the six MIR taconite plants (from west to east: Keetac, Hibtac, Minntac, Utac, Minorca, and Northshore) sampled during the course of the study;

3. **MIR Community Gravimetric Report** (Monson Geerts et al., 2019c), which characterizes mass PM concentrations collected within five MIR communities (from west to east: Keewatin, Hibbing, Virginia, Babbitt, and Silver Bay) as well as three non-MIR background sites (Ely Fernberg site, Duluth NRRI rooftop, and University of Minnesota Mechanical Engineering Building rooftop) sampled during the course of the study;
4. **Mineralogy – Characterization of Elongate Mineral Particles (EMPs) Report** (Monson Geerts et al., 2019d), which identifies the mineralogy of all EMPs detected from PM collected at community sites, background sites, and the taconite plants, and characterizes their physical properties;
5. **Elemental Chemistry of Particulate Matter (PM) Report** (Monson Geerts et al., 2019e), which characterizes the chemistry of the PM collected at community sites, background sites, and taconite plants in terms of elemental mass; and
6. **Lake Sediment Historical Report** (Zanko et al., 2019), which describes the mineralogical, physical, and chemical characteristics of EMP identified in dated lake sediments from two lakes sampled during the study ("North-of-Snort" Lake near Babbitt and Silver Lake in Virginia).

A brief description of the study is presented below, followed by the findings (executive summaries) from the six individual reports.

Disclosure and Disclaimer Statement

The NRRI's aerosol PM sampling protocols — including the equipment utilized and the data collected in this study — are not meant to represent, or to be considered a substitute for, ambient air monitoring as conducted and reported by regulatory agencies such as the Minnesota Pollution Control Agency (MPCA), Minnesota Department of Health (MDH), and the United States Environmental Protection Agency (USEPA). Any reference to State, USEPA, National Ambient Air Quality Standards (NAAQS), Occupational Health and Safety Administration (OSHA), and/or Mine Safety Health Administration (MSHA) standards is for illustrative and comparative purposes and is meant only to provide context to the findings in this NRRI study.

BACKGROUND

Higher-than-expected rates of the rare cancer mesothelioma have been identified in a cohort comprising Minnesota taconite workers. Mesothelioma develops in cells of the mesothelium, the protective lining around many of the internal organs of the body, and is most commonly caused by exposure to asbestos (Case et al., 2011). An initial finding by the Minnesota Department of Health (MDH) identified 58 individuals with mesothelioma from an original cohort of 72,000 mine workers (MDH, 2007). This finding prompted the Minnesota Legislature to approve $4.9 million for Minnesota Taconite Worker's Health Study (MTWHS), which was conducted by a collaborative effort between the University of Minnesota's School of Public Health (SPH), the University of Minnesota Medical School (UMMS), and the University of Minnesota Duluth's Natural Resources Research Institute (NRRI). This five-year study comprised five studies as identified below:

- Occupational Exposure Study (SPH)
- Mortality (cause of death) Study (SPH)
- Incidence Study (SPH)
- Respiratory Health Survey of Taconite Workers and Spouses (UMMS)
- Environmental Study of Airborne Particulate Matter (NRRI)

The NRRI's role in this study was to conduct a baseline environmental study of airborne particulate matter within five MIR communities, three background sites, and six taconite processing plants. The environmental study comprised gravimetric determinations as well as chemical, mineralogical, and morphological characterization. Identifying and characterizing ***"What is in the air?"*** in both the communities and taconite plants in the study area is paramount to understanding the nature of mineral dust(s) and their source(s) that may be either directly or indirectly associated with the increase in this disease. The NRRI's characterization study was conducted with an emphasis on Elongate Mineral Particles (EMPs) present in the airborne particulate matter (PM). As defined by the National Institute of Occupational Safety and Health (NIOSH) in 2011, EMPs may comprise particles of any mineral species that have length-to-width ratios greater than 3:1 and that are of inhalable, thoracic, or respirable size. In this Executive Summary and six detailed reports, EMPs were identified utilizing a variety of analytical methods, including Minnesota Department of Health (MDH) Method 851 (TEM (transmission electron microscopy) Analysis for Mineral Fibers in Water), MDH Method 852 (TEM Analysis for Mineral Fibers in Air), International Organization for Standardization (ISO) Method 13794 (Ambient air – determination of asbestos fibres – indirect-transfer electron microscopy method), ISO Method 10312 (Ambient air – determination of asbestos fibres – direct-transfer transmission electron microscopy method), U.S. Environmental Protection Agency (USEPA) Method 540-R-97-028 (Superfund method for the determination of releasable asbestos in soils and bulk materials), as well as Proton Induced X-ray Emission (PIXE) analysis. Exposure to asbestiform (hair-like, flexible fibers) amphibole (and possibly chrysotile) EMPs has been attributed to the development of mesothelioma (Gunter et al., 2007).

Taconite mining and processing activities in northeastern Minnesota can generate mineral dust that may have the potential to produce adverse human health effects. This mineral dust – or PM – is made up of small particles (generally micron- and submicron-sized). Particle mineralogy, morphology, and concentration dictate whether there is potential for adverse health effects (Gunter et al., 2007). Some of the rocks in northeast Minnesota can contain amphibole mineral species. The Biwabik Iron Formation (BIF) comprises rocks that make up the ores mined for iron on the Mesabi Iron Range (MIR). The mineralogy of the BIF dictates the composition of the dust produced when this rock is crushed and ground at MIR taconite operations. The mineralogy of the BIF changes from west to east across the MIR due to contact metamorphism (a type of thermal metamorphism that can be considered as "baking" of the original rocks in the area), which occurred approximately 1.1 billion years ago during the emplacement of a series of mafic (iron- and magnesium-rich) igneous intrusions collectively known as the Duluth Complex. Heat associated with this event raised the temperature of the BIF, with the eastern part of the rock unit reaching the highest temperatures immediately adjacent to the Duluth Complex. The heat associated with this contact or thermal metamorphism caused primary iron-formation minerals to chemically react to locally generate amphiboles and other iron-silicate minerals (Fig. 1 – Red dashed line between Zones 2 and 3). The amphiboles are principally of the cummingtonite-grunerite series and can include actinolite and hornblende (French, 1968; Ross et al., 1993). These amphiboles have the potential to have crystal morphologies that are prismatic, acicular (needle-like), or fibrous (asbestiform).

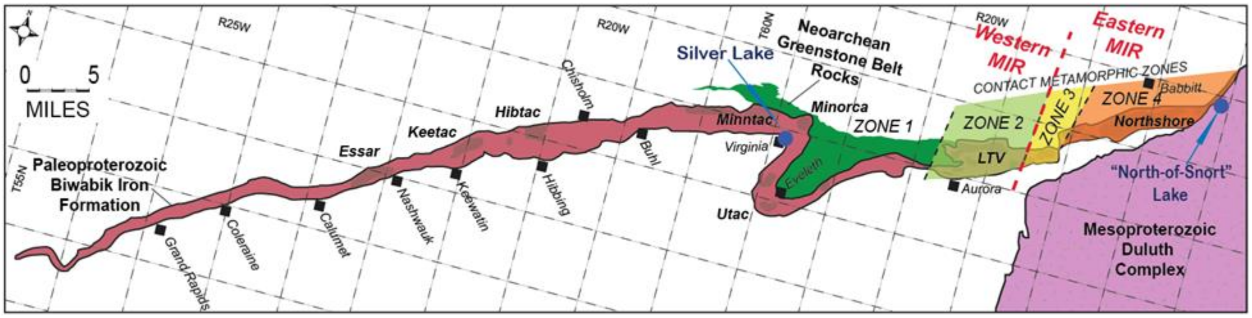

Figure 1. Simplified geologic map of the BIF and contact metamorphic zones adjacent to the Duluth Complex (after French, 1968; Jirsa et al., 2005; McSwiggen and Morey, 2008).

Particulate matter sampling locations for the NRRI study included five population centers on the MIR communities including Keewatin (elementary school), Hibbing (high school), Virginia (city hall), Babbitt (municipal building), and Silver Bay (high school), as well as two separate locations north and south of the MIR for background comparison (Ely (Fernberg site) and Duluth (NRRI rooftop)) (Fig. 2). In addition, a third background site, Minneapolis (Mechanical Engineering Building rooftop – UMN Campus), was added later in the study.

Sampling was opportunistic with respect to location and focused on populated areas rather than a grid-based sampling distribution. Further details regarding MIR community and Minnesota background site sampling protocols can be found in the section titled "Standard Operating Procedure Report" later in this report.

Each of the six currently operating taconite processing plants was also sampled during the NRRI study. From west to east (Fig. 3), these included: Keetac, Hibtac, Minntac, Utac, Minorca, and Northshore. Each taconite plant was sampled a minimum of once during active operation and once during inactivity (no mining or processing activities were taking place), if the occasion arose; those that were sampled during total inactive operation include: Keetac, Hibtac, and Northshore. Two of the plants (Minorca, located in Zone 1, and Northshore, located in Zone 4) were sampled a minimum of three times during active operation to evaluate consistency (statistical analysis), as well as to evaluate potential differences between dust associated with ore mined and processed from the eastern and western parts of the MIR. Further details regarding taconite plant sampling protocols can be found in the section titled "Standard Operating Procedure Report."

Figure 2. MIR community locations.

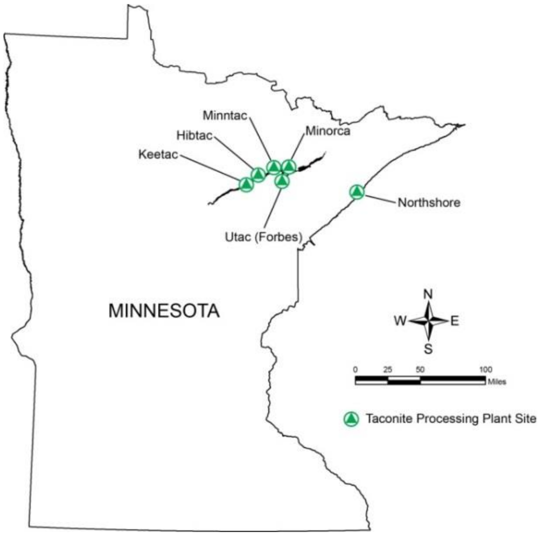

Figure 3. MIR plant locations.

<u>Reporting Structure</u>

Findings from the NRRI's environmental characterization of PM in the MIR communities and taconite processing plants are included in six separate detailed reports:

1. **Standard Operating Procedure Report and Glossary** (Monson Geerts et al., 2019a)
2. **Mesabi Iron Range Taconite Processing Plant Gravimetric Report** (Monson Geerts et al., 2019b)
3. **Mesabi Iron Range Community Gravimetric Report** (Monson Geerts et al., 2019c)
4. **Mineralogy Report – Characterization of Elongate Mineral Particles** (Monson Geerts et al., 2019d)
5. **Particulate Matter Chemistry Report** (Monson Geerts et al., 2019e)
6. **Lake Sediment Historical Report** (Zanko et al., 2019)

A summary of findings for each of the separate reports is included below.

ENVIRONMENTAL PARTICULATE MATTER CHARACTERIZATION FINDINGS

1. Standard Operating Procedure (SOP) Report (Monson Geerts et al., 2019a)

The standard operating procedure (SOP) for PM sampling associated with this study was developed with the assistance of a Science Advisory Board (SAB) and collaboration with aerosol scientists at the University of Minnesota Department of Mechanical Engineering and University of Florida-Gainesville. This document is a comprehensive narrative that outlines the development of the sampling methodology that was used consistently throughout the study, including a history of in-house experiments that were conducted at the request of the SAB and collaborators to address questions and/or possible concerns that ultimately resulted in the strengthening of the overall sampling design.

Equipment utilized in PM sample collection included the Micro-Orifice Uniform Deposition Impactor (MOUDI) (Marple et al., 1991; 2014), an instrument that collects size-fractionated samples of PM with aerodynamic diameters ranging from 30.0 microns to 0.056 microns (one million microns make up a meter), and a final filter that collects particles with aerodynamic diameters less than 0.056 microns. Results from the MOUDI sampling allow PM to be classified into specific size classifications, including: PM_1 (all particles with aerodynamic diameters less than or equal to 1 micron), $PM_{2.5}$ (all particles with aerodynamic diameters less than or equal to 2.5 microns), and PM_{10} (all particles with aerodynamic diameters less than or equal to 10 microns), that are important for assessing air quality parameters. As well, a total filter sampler (TFS) that collects all size fractions of PM on a single filter was utilized so that particle mineralogy, morphology, and chemistry could be identified. Highlights of the findings from the sampling methodology phase of the study are:

- The sampling equipment used in this study, including the Model 120 MOUDI™ II Cascade Impactor and Savillex TFS, has been proven to be precise and efficient at reproducing PM data in all climate conditions (Marple, 2004; Marple et. al., 1991; 2014). Furthermore, a high degree of precision and consistency was tested experimentally and found to exist between the three individual MOUDI samplers utilized in this study (Appendix D).
- Both test results and references concluded that the most optimum substrates for MOUDI impaction surfaces are polycarbonate (PC) filters, Teflon® substrates for the after filter, and Mixed Cellulose Ester (MCE) filters for TFS collection. These media were chosen also to comply with MDH Method 852 (MDH 852, 1976) and International Organization for Standardization (ISO) methodologies (ISO 13794, 1999).
- Initial concerns over particle bounce in the impactors warranted use of greased substrates. However, due to numerous problems with its use, grease was limited to the top (0) stage for community sampling and the top two stages (0, 1) for plant sampling. Subsequent sampling has shown that a single greased substrate in the top stage of the MOUDI column (top two stages for plant sampling increased PM amounts) is sufficient in controlling particle bounce.
- Experiments where humidity was artificially added in a laboratory setting, in an attempt to minimize particle bounce, were considered difficult to replicate in the field and statistically shown to have little or no effect.
- Regression analysis indicates no statistically significant difference in gravimetric results between earlier samples collected using grease on the upper six stages versus samples collected with grease on only the upper one to two stages.
- The use of field and laboratory blanks for quality assurance (QA) inspired the development of a blank protocol that proved both accurate and dependable. The protocol had minor amounts of variance and great consistency throughout the duration of the study.
- As part of the QA and testing of protocol development, experiments were conducted placing field blanks and their petri slide holders both inside and outside of the MOUDI samplers. No statistically significant differences were found between the two locations.
- Separate experiments focused on the distribution of polystyrene latex (PSL) particle spheres on MCE substrates collected using the TFS and showed that, although the flow into the TFS is laminar, the PM was evenly distributed at a scale that allows for quarter substrate replication results. The tests, in conjunction with quality assurance/quality control (QA/QC) duplicated samples, showed that the distribution in quartered MCE substrates, for the purpose of analysis, was reproducible. Analytical method MDH 852 (Transmission Electron Microscope (TEM) Analysis for Mineral Fibers in Air) involves an indirect preparation dissolving the original MCE filter substrate quarter and then re-depositing the PM on a grid for subsequent TEM analysis.
- Minor modifications, including an insulated (extruded polystyrene) protective aluminum housing and the use of a standard light bulb inside the housing during cold weather sampling, were effectively shown to keep the sampling equipment from freezing in cold climate “winter” conditions and temperatures down to -35˚C.
- Experiments involving potential disturbances in the vicinity of sampling equipment in high-density PM areas (inside taconite plants) had little to no overall effect on the gravimetric results of these particle samples. Only coarse particles from the top stage of the MOUDI with a cut-size of 30.0 µm and higher showed any influence.

2. Mesabi Iron Range Taconite Processing Plant Gravimetric Report (Monson Geerts et al., 2019b)

This report discusses the gravimetric characterization and results from aerosol PM sampling within the MIR taconite processing plants. A total of 14 separate aerosol PM sampling events were conducted at the six taconite processing facilities currently operating on the MIR in northeastern Minnesota. All six facilities (designated as Plants A to F, using the same nomenclature as Finnegan and Mandel (2014)) were sampled at least once during active operation (11 total). Three facilities were sampled during total plant shutdown (inactive) in 2009. Two of the plants were sampled three times each during active operation. These two plant locations were chosen for the spatial relationship of their respective ore sources (mines) to the Duluth Complex. This ~1.1-billion-year-old intrusion provided heat that resulted in thermal metamorphism of the BIF that changed the mineral, chemical, and physical characteristics of the iron formation at its eastern end (French, 1968; McSwiggen and Morey, 2008; Severson et al., 2009).

Sampling of the taconite facilities occurred randomly during both summer season (defined for this study as May through October) and winter season (defined for this study as November through April). Each of the sampling events comprised two-hour-long sample collections at approximately 30L/minute at four specific locations in each facility, including: 1) fine crusher; 2) concentrator – magnetic separator; 3) agglomerator – balling drums/discs; and 4) kiln – pellet discharge. These locations were chosen within the plants because they represent process areas where important physical and/or chemical changes to the raw material, as related to particle size and/or a change in mineralogical characteristics, take place. Equipment utilized in sample collection included a Micro-Orifice Uniform Deposit Impactor (MOUDI: Marple et al., 1991; 2014), as described in the previous section, "Standard Operating Procedure (SOP) Report," and a Total Filter Sampler (TFS) that collects all PM on a single substrate.

This report specifically discusses the results of PM gravimetric analysis within the taconite processing facilities. The findings from this report are:

- The equipment and collection protocol used by the NRRI to sample the six plants was consistent throughout the sample collection period.
- Based on the relative consistency of the geology of the BIF and the similarities of mining and processing the ore, statistical power of analysis was achieved through the 10 cumulative sample series collected in the five most western plants. Four additional sample series were collected from the sixth plant on the east end of the BIF, where the geology/mineralogy of the iron formation has been changed due to thermal metamorphism.
- Substantial and statistically significant differences in the amount of PM existed between the inactive and active plant processing operations. Although PM in the areas of active crusher and concentrator were found to be up to three times greater than in inactive plants, these amounts were not statistically significant. The process areas of active agglomerator and kiln were the dustiest areas of the plants, and statistically significant results indicate these process areas contain up to 30–40 times the amount of PM compared with inactive plants.
- Although some variations were observed from plant to plant (due to varying ventilation systems, housekeeping, etc.), our findings indicate that the four locations in the plants contained PM. Ranked from 'most' (1) to 'least' (4), average PM levels (measured in (μg/m^3), including total and PM$_{2.5}$ fraction (respirable fraction), were: 1) the kiln areas; 2) the agglomerator areas; 3) fine crusher areas; and 4) the concentrator/magnetic separator areas (Fig. 4). Note that Total PM and PM$_{2.5}$ concentrations in communities sampled are generally orders of magnitude lower than concentrations in the various plant process areas.

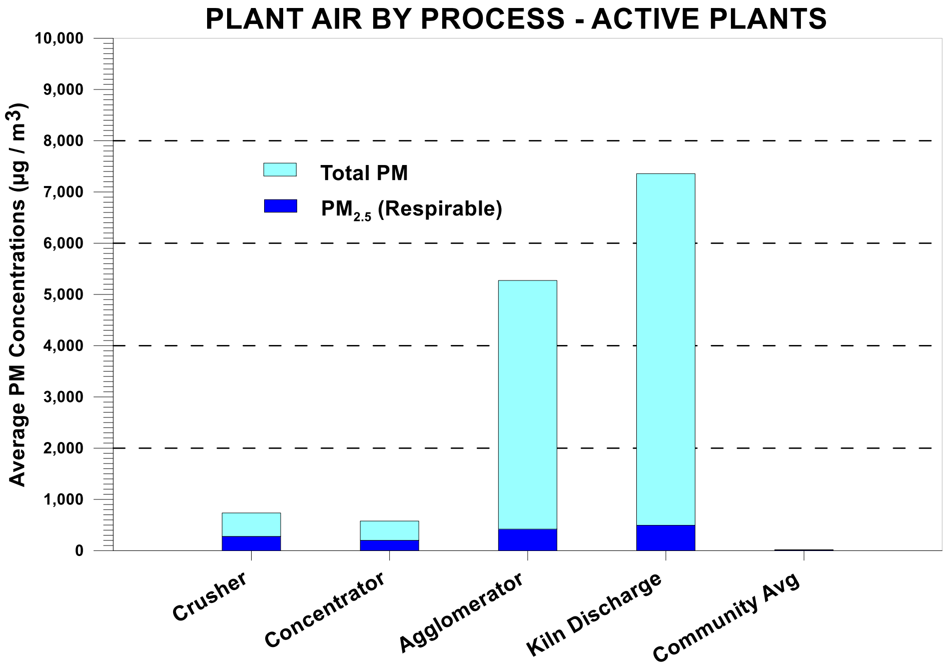

Figure 4. Relationship between average Total PM and $PM_{2.5}$ concentrations in plant process areas and communities sampled during this study.

- Total PM and $PM_{2.5}$ concentrations were higher in active plants versus inactive plants. However, the finer $PM_{2.5}$ fraction comprised a higher percentage of the total percent PM in inactive versus active plants, most probably due to the fact that most PM in active plants is coarse-fraction and settles out more rapidly.
- ▪ Comparisons of the PM (active) results from Plant A and Plant F (plants receiving three sampling events each) were statistically insignificant and were found to be similar to the average levels found in all active plants across the MIR.
- ▪ Average, high, and low total PM concentrations (measured in $\mu g/m^3$) from samples collected using the MOUDI in the four areas of the active plants were as follows (Table 1):

Table 1. Average, high, and low Total PM concentrations (in $\mu g/m^3$) for the four taconite plant process areas sampled during this study.

Active Plant MOUDI Total PM Concentrations ($\mu g/m^3$)

	Average ($\mu g/m^3$)	**High** ($\mu g/m^3$)	**Low** ($\mu g/m^3$)
Crusher	735.88 (488.23)*	1,570.79	278.39
Concentrator	578.28 (208.94)	885.82	141.01
Agglomerator	5,272.63 (4,831.32)	16,684.25	1,287.18
Kiln	7,357.82 (6,640.14)	20,980.17	445.53

*(standard deviation)

- Average PM concentrations in the respirable $PM_{2.5}$ fraction (measured in $\mu g/m^3$), including their respective percentage of the MOUDI total, from samples collected using the MOUDI in the four areas of the active plants are as follows (Table 2):

Table 2. Average, high, and low $PM_{2.5}$ concentrations (in $\mu g/m^3$) for the four taconite plant process areas sampled during this study.

Active Plant MOUDI $PM_{2.5}$ Concentrations ($\mu g/m^3$)

	Average ($\mu g/m^3$)	High ($\mu g/m^3$)	Low ($\mu g/m^3$)
Crusher	277.80 (154.04)*	577.31 – 41.6% $PM_{2.5}$	118.09 – 34.0% $PM_{2.5}$
Concentrator	201.49 (72.26)	311.41 – 44.9% $PM_{2.5}$	72.09 – 51.1% $PM_{2.5}$
Agglomerator	420.89 (213.07)	944.29 – 5.7% $PM_{2.5}$	239.28 – 5.2% $PM_{2.5}$
Kiln	495.66 (250.75)	948.76 – 7.7% $PM_{2.5}$	188.76 – 40.4% $PM_{2.5}$

*(standard deviation)

3. Mesabi Iron Range Community Gravimetric Report (Monson Geerts et al., 2019c)

As part of the NRRI's "Environmental Study of Airborne PM on the Mesabi Range of northeastern Minnesota," a total of 54 separate aerosol PM sampling events were conducted at five MIR communities in northeastern Minnesota. From west to east, these included the communities of Keewatin, Hibbing, Virginia, Babbitt, and Silver Bay. The communities were chosen for their spatial distribution across the 110-mile-long iron-formation, their relevance as population centers, and their proximity to the six taconite processing plants. In conjunction with the sampling at MIR communities, 24 aerosol PM samples were also collected from three background sites, including Ely (rural and north of the MIR); Duluth (mid-sized city and south of the MIR); and Minneapolis (large metropolitan/industrial area not in northeastern Minnesota). All five MIR communities and background sites were sampled at least three times during both summer season (defined as May through October) and winter season (defined as November through April). Each of the sampling events consisted of 120- to 168-hour sample collections at a rate of approximately 30L/minute at a central location within each community. Equipment utilized in sample collection included a Micro-Orifice Uniform Deposit Impactor (MOUDI: Marple et al., 1991; 2014), and a Total Filter Sampler (TFS), which collects all PM without size discrimination on a single substrate. The findings from this report, which focuses on gravimetric analysis of aerosol PM collected from the MIR communities, are as follows:

- The NRRI's aerosol PM sampling and characterization is not meant to represent, or to be considered a substitute for, ambient air sampling as conducted and reported by regulatory agencies in the state of Minnesota; neither is it intended to be used for purposes of calculating exposure or risk assessment. References in this report to Minnesota ambient air quality standards (MAAQS), USEPA national ambient air quality standards (NAAQS), OSHA, and MSHA standards are for illustrative and comparative purposes and are only to provide context to the findings in this study. The NAAQS PM_{10} and $PM_{2.5}$ standards are established and accepted air quality benchmarks and provide a logical frame of reference. **Study conclusions find that all MIR community samples have concentration less than the NAAQS limits for PM_{10} and $PM_{2.5}$. (*Please keep in mind that the NRRI's community air sampling methodology and reported $PM_{2.5}$ (and PM_{10}) data are not meant to be considered a substitute for ambient air sampling as conducted and reported by regulatory agencies***

***such as MPCA and USEPA. NRRI has referenced the NAAQS PM$_{2.5}$ standard for illustrative and comparative purposes and to provide context to its community (and background location) air sampling findings, because the NAAQS PM$_{2.5}$ standard is an established and accepted air quality benchmark.*)**

- The equipment and methods of sample collection used by the NRRI to characterize PM in the five MIR communities, including the three Model 120 MOUDI II cascade impactors, were in good agreement and shown to provide consistent measurements throughout the sample collection period (Marple et al., 2014). Statistical analysis indicates that NRRI PM$_{2.5}$ and PM$_{10}$ from the Virginia site (2009) showed no significant difference with PM$_{2.5}$ and PM$_{10}$ concentrations utilizing high-volume samplers associated with state regulatory sampling (MPCA, 2010) that was performed using USEPA-approved protocols. A detailed explanation of all statistical analyses can be found in the Sample Methodology – Preliminary Statistical Analysis section of this study report.
- Compared with PM concentration variances from 10+ years of regulatory sampling by the MPCA, coupled with the increased sample collection times used by the NRRI (120 to 168 hours), the statistical power of community sampling completed by the NRRI for PM characterization in this study was conducted at confidence rates ranging from 85% to 95%.
- An index, called the “Taconite Particle Source Index” (TPSI), was assigned to each sample with the intention of correlating PM concentrations with meteorological data collected at the time of sampling. The index largely represents the percent time that winds greater than 1.8 meters/second (~4 mph) were blowing from a potential source of mining-related PM (including: mine pits, haul roads, tailings basins, and active waste rock piles) and local taconite processing facilities/plants. Statistically significant correlation between PM and TPSI was found in the communities of Virginia (negative) and Babbitt (positive). There was no correlation between the TPSI and PM in the other three MIR community locations.
- Statistical analysis concluded that there was no significant difference in mean PM concentration levels at the five MIR community locations when local mine/plants were active compared to when they were inactive. Similarly, there was no significant difference identified for PM concentrations between MIR communities and the Ely background location.
- Average winter season PM concentrations were statistically significantly higher than summer season.

- Specific **total PM** average concentrations (measured in µg/m^3) from samples collected (n=number of samples) using the MOUDI in the five MIR community locations are as follows (Table 3):

Table 3. Average Total PM concentrations (in µg/m^3) for all sampling events, sampling events during plant inactivity, and sampling events while plants were active for Mesabi Range communities and background sampling sites away from the Mesabi Range.

MOUDI Total PM

	Average (µg/m^3)	**Inactive Plant (µg/m^3)**	**Active Plant (µg/m^3)**
Keewatin Elementary School **n=7**	9.07 (2.91)*	9.49 (n=6) (2.95)	6.55 (n=1) (NA)
Hibbing High School **n=9**	10.61 (3.05)	10.76 (n=3) (4.24)	10.54 (n=6) (2.77)
Virginia City Hall **n=10**	12.70 (3.75)	13.11 (n=4) (3.46)	12.43 (n=6) (4.24)
Babbitt Municipal Building **n=16**	10.41 (3.38)	9.59 (n=7) (2.29)	11.04 (n=9) (4.06)
Silver Bay High School **n=12**	11.42 (4.82)	9.94 (n=7) (3.70)	13.49 (n=5) (5.84)
BACKGROUND			
Ely Fernberg Site **n=6**	8.15 (3.63)	Not Applicable	Not Applicable
Duluth NRRI Building **n=12**	8.43 (4.35)	Not Applicable	Not Applicable
Minneapolis UMN Mech. Eng. **n=6**	15.25 (3.08)	Not Applicable	Not Applicable

*(standard deviation based on concentrations)

Since there are no ambient air standards for “total PM,” PM_{10} is used for comparative purposes: NAAQS PM_{10} limit value is 35 µg/m^3 for a three-year average.

- Specific PM levels in the respirable **$PM_{2.5}$ fraction** (measured in µg/m^3 – and by representative percent of total sample) from MOUDI totals in the five MIR community locations are as follows (Table 4):

Table 4. $PM_{2.5}$ concentrations (in µg/m^3)for all sampling events, sampling events during plant inactivity, and sampling events while plants were active for Mesabi Range communities and background sampling sites away from the Mesabi Range.

$PM_{2.5}$ Fraction of MOUDI Total

	Average (µg/m^3)	**Inactive Plant (µg/m^3)**	**Active Plant (µg/m^3)**
Keewatin **n=7**	6.54 (72.5%) (2.08)*	6.92 (73.7%) (2.00)	4.27 (65.1%) (NA)
Hibbing **n=9**	6.45 (61.2%) (1.92)	6.75 (63.6%) (2.38)	6.31 (60.0%) (1.88)
Virginia **n=10**	7.04 (55.7%) (2.23)	7.69 (57.8%) (2.62)	6.61 (54.4%) (2.08)
Babbitt **n=16**	6.64 (62.8%) (2.64)	5.63 (57.5%) (2.08)	7.42 (67.0%) (2.86)
Silver Bay **n=12**	6.37 (56.5%) (2.75)	5.78 (58.5%) (2.39)	7.18 (53.6%) (3.28)
BACKGROUND			
Ely Fernberg Site **n=6**	5.83 (71.5%) (2.73)	Not Applicable	Not Applicable
Duluth NRRI Building **n=12**	5.21 (66.9%) (1.94)	Not Applicable	Not Applicable
Minneapolis UMN Mech. Eng. **n=6**	9.73 (63.5%) (2.24)	Not Applicable	Not Applicable

*(standard deviation based on concentrations)

For comparative purposes: NAAQS $PM_{2.5}$ limit value is 12 µg/m^3 for a three-year average.

- The Minneapolis background site had the highest average total PM (15.25 µg/m^3) and PM$_{2.5}$ (9.73 µg/m^3) concentrations of the study, while Virginia had the highest average total PM (12.70 µg/m^3) and PM$_{2.5}$ (7.04 µg/m^3) concentrations of MIR communities. Ely had the lowest average total PM (8.15 µg/m^3) and Duluth the lowest PM$_{2.5}$ (5.21 µg/m^3) concentrations of the study, while Keewatin had the lowest average total PM (9.07 µg/m^3) and Silver Bay the lowest PM$_{2.5}$ (6.37 µg/m^3) concentrations of MIR communities. Those relationships are shown graphically as follows (Fig. 5):

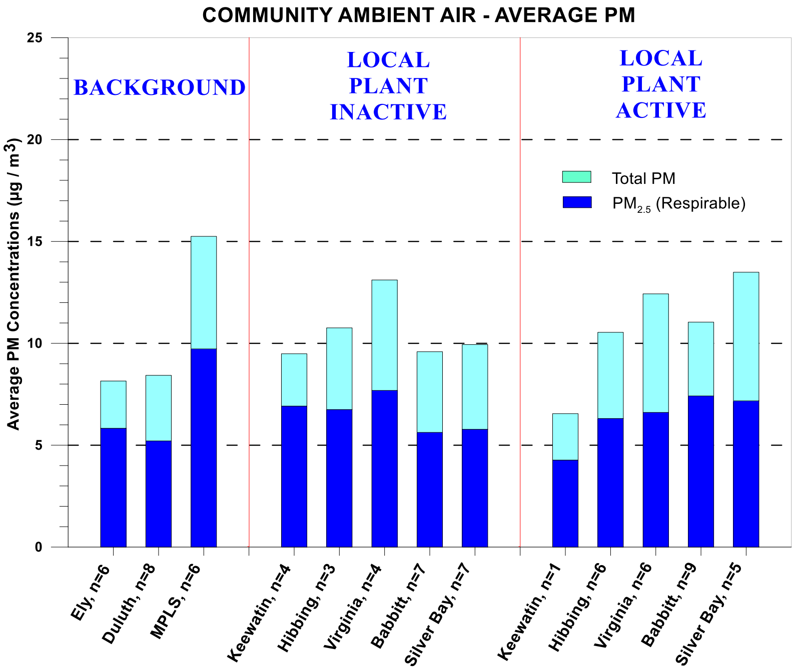

Figure 5. Summary of average Total PM and PM$_{2.5}$ concentrations (in µg/m^3) for community sampling sites and background sites. Community sampling site data is shown during plant activity and while plants were inactive.

4. Mineralogy – Characterization of Elongate Mineral Particles (EMPs) (Monson Geerts et al., 2019d)

"NIOSH considers asbestos to be a "Potential Occupational Carcinogen"[*] *and recommends that exposure be reduced to the lowest feasible concentration. The NIOSH recommended exposure limit (REL) for airborne asbestos fibers and regulated EMPs is 0.1 countable EMP from one or more covered minerals per cubic centimeter, averaged over 100 minutes, where:*

- *a* ***"Countable" elongate mineral particle (EMP)*** *is any fiber or fragment of a mineral longer than 5 µm with a minimum aspect ratio of 3:1 when viewed microscopically with use of NIOSH Analytical Method 7400 ("A" rules) or its equivalent; and*
- *a* ***"Covered" mineral*** *is any mineral having the crystal structure and elemental composition of one of the asbestos varieties (chrysotile, riebeckite asbestos [crocidolite], cummingtonite-grunerite asbestos [amosite], anthophyllite asbestos, tremolite asbestos, and actinolite asbestos) or one their nonasbestiform analogues (the serpentine minerals antigorite and lizardite, and the amphibole minerals contained in the cummingtonite-grunerite mineral series, the tremolite-ferroactinolite mineral series, and the glaucophane-riebeckite mineral series).*

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[*] NIOSH's use of the term "Potential Occupational Carcinogen" dates to the OSHA classification outlined in 29 CFR 1990.103 (OSHA, 1990), and, unlike other agencies, is the only classification for carcinogens that NIOSH uses.

This clarification of the NIOSH REL for airborne asbestos fibers and related EMPs results in no change in counts made, as defined by NIOSH Method 7400 ('A' rules). However, it clarifies definitionally that EMPs included in the count are not necessarily asbestos fibers" (NIOSH, 2011).

Mineral particles detected in this study that correspond to the constraints of these NIOSH parameters will be reported as "countable" structures and/or "covered" minerals, by definition (NIOSH, 2011). Particles that fit both of these definitions will be identified as "countable"/"covered" EMPs. Analytical techniques for identifying EMPs in this report utilized two methods by two separate certified commercial laboratories, including: 1) Transmission Electron Microscopy (TEM) at Braun Intertec Corporation (Minneapolis, MN), using the Minnesota Department of Health (MDH) 852 Method (1976), which is an indirect preparation for determining asbestos fibers from the total filter samples (TFS); and 2) TEM at EMSL Laboratories (Minneapolis, MN) using the International Standardization Organization (ISO) Method 13794 (1999), also an indirect preparation method for determination of asbestos fibers from PM_1 substrates. Both of these TEM analytical methods are consistent with NIOSH Method 7402 (1994). In addition to PM substrate analyses, crushed/milled core samples of the ore horizon were also analyzed using a Modified Elutriator Method (EPA 540-R-97-028; USEPA, 1997) coupled with the ISO Method 10312 (1995), a direct preparation for determining the concentration of asbestos structures in the respirable portion of PM. The Natural Resources Research Institute (NRRI) also conducted a scanning electron microscope (SEM) studies to detect, identify, measure, and photograph EMPs from the copper grids generated by Braun Intertec for purposes of additional characterization and confirmation.

This report discusses the results of mineral characterization of the PM within the MIR communities, taconite processing facilities, and background locations. Highlights of the mineral characterization results include:

- The sampling methodology remained constant throughout the study, with the exception of increasing the sampling duration in MIR communities and background locations from five to seven days (120 to 168 hours), in an effort to increase analytical sensitivities for asbestos analysis.
- With the exception of the industrial asbestos mineral chrysotile, detected EMP mineralogy for both community/background and taconite plant locations was consistent with mineralogy identified in the local bedrock and glacial sedimentary deposits in northeastern Minnesota.

Community and Background Sampling

- Statistical analyses between separate EMP analytical results from Babbitt, MN were conducted using the Wilcoxon test (a ranking-based test for non-parametric data) (Wilcoxon, 1945). Comparison of two datasets: 1) MPCA (2008; 2009) – 20 samples, December 2007 through June 2008; and 2) NRRI – 16 samples, November 2008 through September 2009, indicated no significant differences in the results between the two data sets.
- Community sampling resulted in 73 individual samples (53 MIR community and 20 background) totaling 9,874 hours of ambient air sampling with 17,513 m^3 of air sampled by the MOUDI and 5,291 m^3 by the TFS.
- From this, a total of 669 EMPs were detected including: 1) 117 (17.5%) "covered" mineral EMPs (chrysotile and listed amphiboles cummingtonite/grunerite, actinolite, and tremolite with aspect ratios ≥ 3:1; NIOSH, 2011); 2) 507 (75.8%) "Non-Amphibole/Non-Chrysotile" EMPs; and 3) 45 (6.7%) "Ambiguous" EMPs. One tremolite EMP cleavage fragment was detected.
- Approximately 50% of the community/background samples contained one or more "covered" mineral EMPs (by NIOSH definition, 2011) with calculated concentrations ranging 0.003 to 0.0147 structures per cubic centimeter of air (s/cm^3). The NIOSH recommended

exposure limit (REL) for airborne asbestos fibers and regulated EMPs is 0.1 countable EMP structures from one or more covered minerals per cubic centimeter, averaged over 100 minutes.

- Of the 117 identified “covered” EMPs, a total of ten were “countable” (“covered” ≥ 5 µm length; NIOSH, 2011) EMPs, representing 1.5% of the total number of EMPs. These 10 EMPs were detected from the MIR communities of Virginia (2), Babbitt (1), and Silver Bay (7), in concentrations ranging from 0.003 to 0.0018 s/cm^3. No “countable”/”covered” structures were detected in the communities of Keewatin or Hibbing or at any of the background sites in Minneapolis (MN), Duluth (MN), or northeast of Ely (MN).
- Average concentrations of “countable”/“covered” EMPs for background sites, community sites sampled during plant operations, and community sites sampled during plant inactivity consistently are below 0.0005 EMPs/cm^3 (Fig. 6).

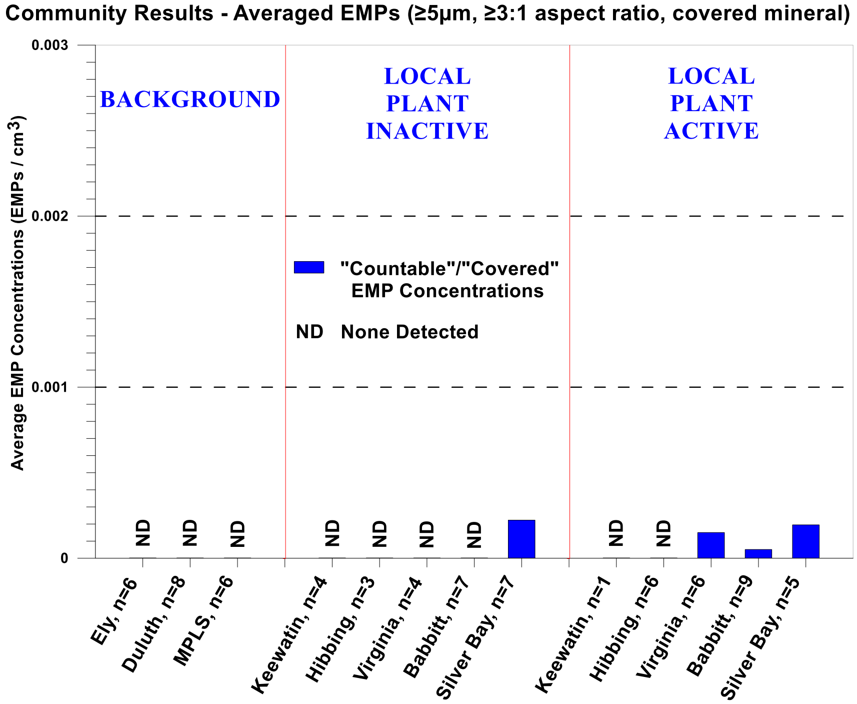


Figure 6. Summary of EMP concentrations (in EMPs/m^3) identified for community sampling sites and background sites. Community sampling site data is shown during plant activity and while plants were inactive.

- With the exception of the industrial asbestos mineral chrysotile, 97% of the measured morphological dimensions of EMPs detected in the communities and background locations do not meet the NIOSH (2011) definition of “countable”/”covered.”
- Of the 1,559 EMPs that were detected during community and background sampling using both analytical methods, one (0.06%) “countable”/”covered” amphibole mineral EMP was identified from the community location of Silver Bay that had an aspect ratio ≥ 15.
- Thirty-four EMPs (2.8%), categorized as “Non-Amphibole/Non-Chrysotile” and “Ambiguous,” were detected in the TFS samples from MIR communities that had aspect ratios ≥ 15.

Taconite Plant Sampling

- Taconite plant sampling resulted in 55 individual samples from four process locations in the plants: 1) crusher; 2) concentrator; 3) agglomerator; and 4) kiln (Table 5). A total of 138 hours of plant air sampling took place, with 247 m^3 of air sampled by the MOUDI and 51 m^3 by the TFS.

Table 5. Summary of "countable"/"covered" EMPs detected per process area in the six taconite plants evaluated during this study.

PROCESS AREA	"COUNTABLE"/"COVERED" EMP NOT DETECTED	"COUNTABLE"/"COVERED" EMP DETECTED (Average, EMP/cm^3)
Secondary Crusher	**5 Plants**	**1 Plant (0.2 EMP/cm^3)**
Concentrator	**4 Plants**	**2 Plants (0.03 and 0.1 EMP/cm^3)**
Agglomerator	**6 Plants**	**No Plants**
Kiln Discharge	**6 Plants**	**No Plants**

***Point source samples not to be confused with exposure measurements**
Countable/Covered (EMP = ≥ 5 µm, ≥ 3:1 aspect ratio, covered minerals)

- From the taconite plant TFS sampling, a total of 691 EMPs were detected: 1) 145 (21%) "covered" (NIOSH, 2011) mineral EMP; 2) 510 (73.8%) "Non-Amphibole/Non-Chrysotile"; and 3) 36 (5.2%) "Ambiguous" EMPs.
- Chrysotile (concentration of 0.1099 s/cm^3) was detected in one plant sample series during the study. The samples came from the Plant D kiln during active operation.
- Of the 145 "covered" EMPs identified, a total of 8 were "countable" (NIOSH, 2011), representing 1.1% of the total number of EMPs. These EMPs were detected in taconite plants A (seven) and E (one). The mineralogy of these EMPs included seven EMPs (six cummingtonite/grunerite and one actinolite) from four separate samples from the crusher/concentrator at Plant A, and one cummingtonite/grunerite EMP in a concentrator sample at Plant E.
- Plant air concentrations of "countable"/"covered" structures ranged from 0.0346 s/cm^3 (one EMP at Plant E) to 0.3219 s/cm^3 (single sample maximum three EMPs in the concentrator at Plant A). The NIOSH recommended exposure limit (REL) for airborne asbestos fibers and regulated EMPs is 0.1 countable EMP structures from one or more covered minerals per cubic centimeter, averaged over 100 minutes.
- Four of the 55 taconite plant samples had concentrations of "countable"/"covered" structures that exceeded 0.1 s/cm^3; all were from one plant.
- Twenty-eight EMPs that had aspect ratios ≥ 15 (categorized as "Non-Amphibole/Non-Chrysotile" and "Ambiguous") were detected in five of the six MIR taconite plants.

5. Particulate Matter Chemistry Report (Monson Geerts et al., 2019e)

Chemical characterization of the aerosol PM was completed through elemental analysis on 270 sample substrates (260 aerosol samples plus 10 blanks and duplicates) using Proton-Induced X-Ray Emission (PIXE) spectrographic technique. Seventy-two inorganic elements were analyzed using the PIXE technique. For the purposes of this study, 11 of the most abundant elements were compared, including those that are common constituents in the geological materials (rocks, glacial deposits, soils) of northeastern Minnesota. In this report, we have organized the order in which the different elements are reported to represent specific groups of minerals in these geological materials:

- Aluminum (Al), sodium (Na), potassium (K), calcium (Ca), silicon (Si), magnesium (Mg), and iron (Fe): Inosilicates (i.e., amphiboles), Phyllosilicates (i.e., stilpnomelane), and Quartz: minerals common to rocks and sediments in northeastern Minnesota;
- Fe, manganese (Mn), and titanium (Ti): Oxides – e.g., magnetite, hematite, etc. minerals common to iron formation; and
- Sulfur (S) and chlorine (Cl): volatiles, common to processes of combustion.

Their concentrations were found to be lognormally distributed and underwent $\log_{10}$ transformation.

The purpose of chemically analyzing the PM was to evaluate chemical signatures that may reveal the source of PM in the MIR communities background sampling sites located away from the MIR and within taconite processing plants. The findings from this report are as follows:

- There are statistically significant higher concentrations of elemental iron (Fe) in MIR communities when the local mines/plants are active versus inactive.
- Average MIR community iron concentrations (collected during active local mines/plants) were compared with PM concentrations collected during inactive mines/plants and those from the Ely background site. These data indicate that, on average, iron concentrations in air were approximately 3.0 times background levels during sampling periods when local mines/taconite plants were active. In essence, when mines/plants were inactive, the elemental iron concentration in MIR communities was similar to the Ely background site.

Community Sampling Results

- Of the 11 elements analyzed statistically using Tukey's Honest Significant Difference (HSD) method (Tukey, 1949), iron was the only element demonstrating statistically significant difference in each of the individual communities during active local mine/taconite processing plant operations. The highest iron concentration occurred in Virginia, while the lowest iron concentration was in Babbitt. Principal Component Analysis (PCA) plots also confirmed this finding.
- The Tukey's HSD method was also used to compare the elemental concentrations in MIR community air by seasonal sampling at each individual community. Silicon (Si) was found to be statistically significantly higher in Virginia during the winter, and sodium (Na) was found to be significantly lower in Silver Bay during the summer.
- Combining the MIR community data and analyzing it using T-tests with unequal variance indicated that both iron and silicon had statistically significantly higher concentrations when local mine/plants were active. Iron and silicon concentrations were also found to be higher during the winter season versus the summer using this type of analysis.
- T-tests with unequal variance indicated that the MIR communities of Hibbing and Virginia had statistically significantly higher iron and silicon in the air than the Ely background site. It was also found that Silver Bay had significantly higher iron than Ely.

- Pearson correlations were tested between both iron and silicon (the two largest components of iron formation rocks) and between the Taconite Particle Source Index (TPSI) – a measure of the percent time during aerosol sampling that winds were blowing from a local taconite mining/taconite processing plant location. These statistical analyses indicated no significant correlation between the elemental concentrations and the TPSI.

Taconite Plant Sampling Results

- Given the elemental components of locally identified inosilicates, i.e. amphiboles, that occur preferentially in the thermally metamorphosed eastern end of the MIR (French, 1968; MCSwiggen and Morey, 2008), both T-tests and Tukey HSD tests using mean ranks indicated statistically significant differences in elemental concentrations of iron, silicon, calcium, and magnesium (Fe, Si, Ca, and Mg, respectively), but were unable to relate these differences specifically to amphibole mineralogy in the Zone 4 plant compared with the other five Zone 1 plants, which reflects the isochemical nature of thermal metamorphism.
- Tukey's HSD tests were used to compare elemental concentrations between four plant process areas that were sampled, including in order of taconite processing, crusher, separator, agglomerator, and kiln. The process areas of the crusher and concentrator generally had lower elemental iron concentrations than the process areas of the agglomerator and kiln. Comparison among the four processes shows that the concentration of silicon attained maximum values at the crusher process area, and silicon was found to have its lowest elemental concentrations at the kiln; however, statistical significance was not achieved for these results. Iron concentrations are statistically significantly higher in the agglomerator and kiln process areas, and lowest in the crusher and concentrator process areas.

Ternary diagrams were generated for each process location using the elemental concentrations of calcium, magnesium, iron, and silicon (Ca, Mg, Fe, and Si, respectively). Beginning with the ore in the crusher, the diagrams display a transition from the chemical composition of the ore in the crusher process area to the chemical composition of the finished product (taconite pellet) in the kiln process area at each of the plants. The type of pellet produced in these plants (acid or flux) can be distinguished by the elemental concentrations of the PM sampled within the plants.

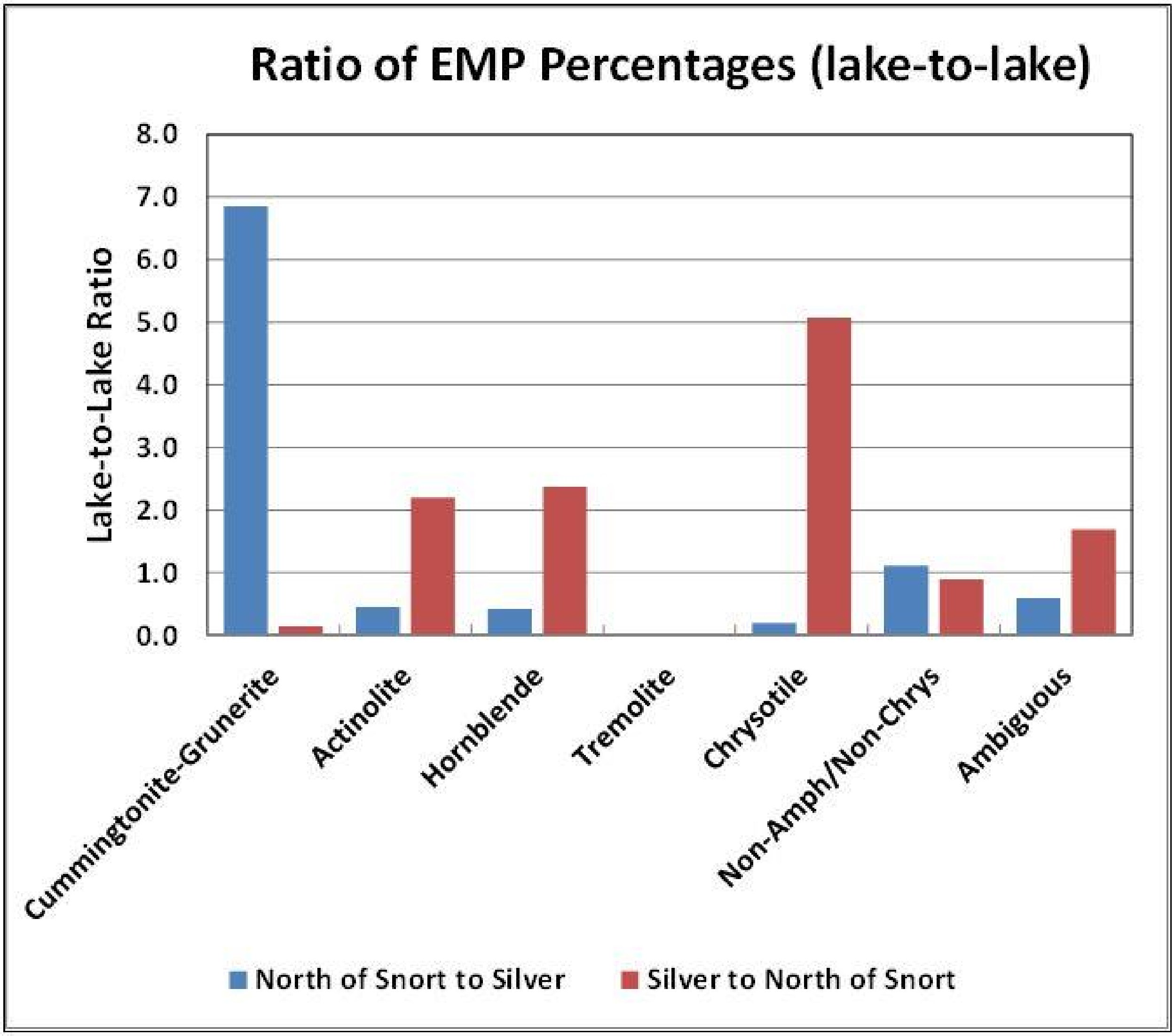
6. Lake Sediment Historical Report (Zanko et al., 2019)

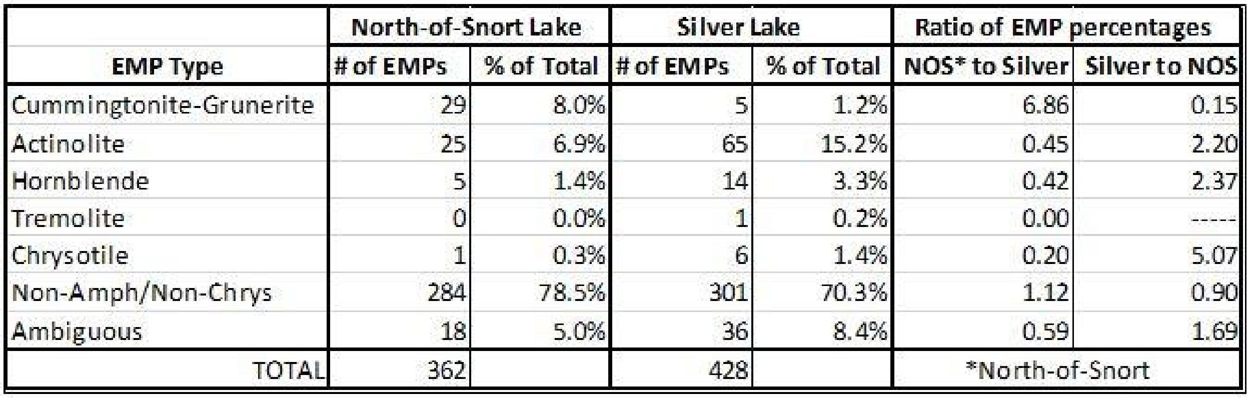
Atmospheric deposition of airborne PM such as fugitive dust contributes to sediment that accumulates at the bottom of a lake. Because of this phenomenon, lake sediment can provide an historic mineralogical and chemical record of what may have been in the air at the time of its atmospheric deposition. This point is important, because the NRRI's role in the MTWHS was to not only help answer the question "What is in the air?" by conducting present-day in-plant and community air sampling, but – and even more challengingly – to potentially answer the question "What *was* in the air?" by collecting and analyzing historic samples. Lake sediment was the only historic sampling medium available that could allow the investigators to make an attempt at assessing what might have been present in the air in the past on Minnesota's Mesabi Iron Range (MIR). The lake sediment study was conceived, in part, to also assess if the mineralogical and physical characteristics (morphology) of insoluble airborne PM components of MIR lake sediment could indicate the presence and/or past use of commercial asbestos.

The NRRI therefore core-sampled, age-dated (using lead-210 (^{210}Pb) and cesium-137 (^{137}Cs) isotopes), and characterized intervals of sediment from two MIR lakes – Silver Lake in Virginia, on the central MIR, and a lake informally named "North-of-Snort" Lake on the eastern end of the MIR, near Babbitt (see Fig. 7). The sampled sediment spanned pre-mining years through the transition from natural ore to taconite mining, to the present. Core intervals – 19 from Silver Lake and 17 from North-of-Snort Lake – were processed using a water elutriation method developed by Dr. James Webber (Webber, 1999; Webber et al., 2008) to isolate insoluble PM having an aerodynamic diameter of less than 2.5 micrometers (µm) ($PM_{2.5}$); such particles are considered respirable. The objective was to determine if fugitive mineral dust generated by past iron ore/taconite mining activity could be discerned in mineral PM deposited and preserved in the sediment of both lakes. Respirable mineral PM of a specific size, shape, and mineralogy – referred to as elongate mineral particles (EMPs) – were of primary interest. The study's operating hypotheses were: 1) some variable fraction of the EMPs identified during this study was of atmospheric origin; and 2) the source for a portion of these EMPs was fugitive dust generated by iron ore mining activities.

Key findings:

- Both North-of-Snort Lake and Silver Lake received atmospheric inputs from fugitive dust generated by mining and other human-related activities, with North-of-Snort Lake sediment more clearly exhibiting mineralogical evidence of probable taconite mining-related inputs post-1950, reflective of the eastern metamorphosed portion of the BIF.
- Changes occurred in the sediment's mineralogical and chemical composition over time. These findings also indicate that at least a portion of the insoluble PM within each lake's sediment very likely came from atmospheric inputs of fugitive dust coincidental with (and likely generated by) historic iron ore/taconite mining activity.
- It is probable that some of this PM has a non-geological (commercial) origin, as suggested by the presence of the only true asbestos mineral identified in the study – chrysotile – in five sediment intervals from Silver Lake. Most of the potentially respirable PM identified by the study is non-amphibole/non-chrysotile. Based on sediment age-dating, all chrysotile occurrences post-date the commencement of mining and other industrial and urbanization activity on the Mesabi Range. This finding may suggest that the chrysotile EMPs originated from commercial asbestos used in building materials or other industrial products, applications, equipment, and machinery as mining and mining support industries expanded, and as the Mesabi Range's population centers grew.
- In total, 4 of the study's 790 identified EMPs found in the lake sediment samples meet the "countable"/"covered" NIOSH Roadmap EMP criteria. Four (0.51%) were identified as actinolite (three from North-of-Snort Lake and one from Silver Lake), and two (0.25%) were identified as non-amphibole/non-chrysotile (one each from both lakes).
- The majority of the EMPs have a blocky rather than acicular (needlelike) morphology. This particle size distribution and morphology is similar to those of EMPs collected and identified during the community air sampling portion of the MTWHS (Monson Geerts et al., 2019c).
- As a percentage of total amphibole EMPs present in the selected lake sediment intervals, cummingtonite-grunerite (CG) is much more common in North-of-Snort sediment (49.2% vs. 5.9% for Silver Lake), while actinolite (76.5%) and hornblende (16.5%) are more prevalent in Silver Lake sediment, compared to 42.4% and 8.5% in North-of-Snort Lake. The following figure further emphasizes and illustrates these mineralogical differences with a plot of the lake-to-lake ratios of the percentage of each EMP type (Fig. 7). These differences suggest that the bedrock and glacial geology in the vicinity of both lakes – and mining and development activity from two mineralogically distinct portions of the BIF – have impacted the mineralogical composition of both lakes' sediments.





	North-of-Snort Lake		Silver Lake		Ratio of EMP percentages	
EMP Type	# of EMPs	% of Total	# of EMPs	% of Total	NOS* to Silver	Silver to NOS
Cummingtonite-Grunerite	29	8.0%	5	1.2%	6.86	0.15
Actinolite	25	6.9%	65	15.2%	0.45	2.20
Hornblende	5	1.4%	14	3.3%	0.42	2.37
Tremolite	0	0.0%	1	0.2%	0.00	-----
Chrysotile	1	0.3%	6	1.4%	0.20	5.07
Non-Amph/Non-Chrys	284	78.5%	301	70.3%	1.12	0.90
Ambiguous	18	5.0%	36	8.4%	0.59	1.69
TOTAL	362		428		*North-of-Snort	

Figure 7. EMP compositions of North-of-Snort and Silver Lakes.

- The clearest and most significant distinction between the mineralogy of the western and eastern (metamorphosed) portions of the BIF was revealed by the elutriation and analysis of samples obtained from the dust collection systems of two western (BIF metamorphic Zone 1) and one eastern (BIF metamorphic Zone 4) taconite production sources. In effect, the dust collection system samples acted as “control” samples because they unambiguously reflected the geology and mineralogy of the BIF at their respective locations. EMPs from both Zone 1 dust collection system samples were 100% non-amphibole/non-chrysotile (no amphibole EMPs were identified). In comparison, 82.1% of the EMPs identified in the Zone 4 dust collection system sample were identified as either cummingtonite-grunerite (35.7%) or actinolite (46.4%).
- The dust collection system findings lend strong support to the interpretation that amphibole EMPs found in Silver Lake sediment (Zone 1) were derived from older Archean bedrock or younger (Quaternary) glacial materials, and not from activities associated with the mining or processing of iron ore/taconite, while the amphibole EMPs found in North-of-Snort Lake sediment – especially post-1950 – were more reflective of the eastern BIF’s mineralogy (see French, 1968; McSwiggen and Morey, 2008) and mining activity. These findings underscore the importance of geological context and taking into account all potential geological sources of fugitive dust when conducting similar studies.
- Chemical analyses of as-is age-dated sediment intervals show potential mining-related influences, but more so in the Silver Lake sediment.

The study confirmed that lake sediment and the water elutriation technique used to isolate respirable PM from lake sediment can be used for historic evaluation of anthropogenic (human-caused) impacts to air quality as well as for other “mineral forensics” types of applications.

CONCLUSIONS

The six NRRI characterization studies provide physical (size and shape), mineralogical, chemical, geological, geographical, and historical context to the findings of the University of Minnesota’s School of Public Health (SPH) and the University of Minnesota Medical School (UMMS). The SPH and UMMS findings (Finnegan and Mandel, 2014) showed that mesothelioma is associated with working longer in the taconite industry. However, the SPH and UMMS investigators “…were not able to state with certainty that the association with EMPs and mesothelioma was related to the ore dust or to the use of commercial asbestos or both.”

The NRRI findings indicate the following:

1) Low concentrations of PM_{10}, $PM_{2.5}$, and EMPs in Mesabi Iron Range community air
2) Elemental iron concentrations in MIR communities were similar to elemental iron concentrations in background sampling locations when taconite mines/plants were inactive. When taconite mines/plants were active, the elemental iron concentrations within communities were found to be statistically higher.
3) Mineralogically and morphologically, the EMPs identified in MIR communities and taconite processing plants were dominated by particles that did not fit the “countable”/”covered” classification criteria. Of the 145 “covered” EMPs identified within the six MIR taconite processing plants, a total of 8 were “countable” (NIOSH, 2011), representing 1.1% of the total number of EMPs, out of 691 total. These EMPs were detected in two taconite plants (seven in one plant and one in another); no other “countable”/”covered” EMPs were detected in the other four plants.
4) The lake sediment study returned similar results, in which 4 of the study’s 790 identified EMPs found in the lake sediment samples met the “countable”/”covered” classification.

5) In comparison to the NIOSH standard, for countable particles, the results from this study show that the community air has significantly lower amounts than the standard.
6) Only one plant and two areas in this plant had countable EMPs above the NIOSH benchmark.
7) The highest particulate matter found was for the Minneapolis reference site in comparison for the Range communities and the other two reference sites.
8) The use of MOUDI sampling techniques is a good method for better understanding not only what is in the air, but also the size of the particles that are in the air.
9) Study of lake sediment can be used to interpret some of the impacts of past industrial activities and to gain a better understanding of the impact of local geology.

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