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Analysis of the Silica Percent in Airborne Respirable Mine Dust Samples From U.S. Operations

ABSTRACT: Exposure to crystalline silica in mining can lead to silicosis, a potentially fatal lung disease, and it may be contributing to the increase of coal workers' pneumoconiosis (CWP) seen in Appalachian miners. Exposure to silica in mines is controlled indirectly by reducing the respirable dust exposure limit through a formula that employs the % of silica in the dust. To reduce this exposure, control technologies and specific monitoring techniques need to be developed and implemented and the knowledge of the % of silica in mine dusts can help this process. This manuscript analyzes the % of silica in dust samples for the U.S. mining industry collected from 1997 to 2011. In the metal/nonmetal (M/NM) industry, metal and sand and gravel mines showed the highest silica % (8.2 %, 9.8 %) along with the highest variability. The silica % was found to be lower for samples collected in underground by comparison to surface and mill. In the coal industry, the samples collected in surface locations showed high silica % in the dust. For both the coal and M/NM industries, the % of silica and the respirable dust concentration were inversely related—i.e., the lower the dust concentration, the higher and more variable silica percentages were observed. The respirable dust limit formula suggests the first explanation: a mine with a high silica % in the dust is required to keep the dust concentration low under the reduced standard. Additional explanations

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are also proposed: the variability of the % of silica in the dust, the selective efficiency of control technologies, and different transport properties for dust with variable silica content. The findings improve the understanding of exposure to silica in mining environments and the data presented will be helpful in developing monitoring strategies for the measurement of silica and for the design of control technologies.

Introduction

Crystalline silica (hereafter referred to as silica) has long been recognized as an occupational hazard. The Occupational Safety and Health Administration (OSHA) in the U.S. estimated in 2003 that nearly 2×10^6 workers were potentially exposed to silica dust in general industry and the mining, construction, and maritime industries [1]. Occupational exposures to silica are associated with the development of silicosis [2], lung cancer [3,4], pulmonary tuberculosis, and airway diseases [5]. Mining is one of the sectors more impacted by the exposure to silica. Recent studies suggest that high silica exposure may explain, in part, the increase of coal workers' pneumoconiosis (CWP) and advanced CWP seen in Appalachian miners [6].

The mining industry in the U.S. is generally categorized by commodity: coal, metal, nonmetal, stone, and sand and gravel (S&G) mines. The Mine Safety and Health Administration (MSHA) divides the mining industry into coal mines and metal/nonmetal mines (M/NM) that include all the non-coal commodities. This division is mainly due to differences in history, mining operators, mining techniques, and geology associated with these different types of mines. In 2008, a total of 14 907 mining operations reported employment data to MSHA. Almost half (47.8 %) were sand and gravel mines, followed by stone mines (31.1 %), coal mines (14.3 %), nonmetal mines (4.8 %), and metal mines (2.0 %). There were 273 496 mine operator employees in 2008, with 85 693 and 187 803 employees reported by coal and M/NM mine operators, respectively. For mine operators, 20.6 % of the employee hours were for underground work locations, while 79.4 % were for surface work locations [7].

Coal and M/NM mines have different monitoring, measurement, and enforcement approaches relative to worker exposure to silica. For both industries, only respirable samples are subjected to analysis for silica, and the non-explicitly stated exposure limit for silica is an 8-h time-weighted average (TWA) concentration of 100 μg/m^3. In coal mines, the respirable dust standard is an 8-h TWA concentration of 2.0 mg/m^3. Silica exposure is controlled by reducing the 2.0 mg/m^3 standard when the content of silica in airborne dust exceeds 5 % by weight. The reduction is made by the following formula [8,9]:

$$\text{Reduced standard } \left(\text{mg/m}^3\right) = 10/[\%] \text{ silica}$$

When applicable, the silica content is determined by using a Fourier-Transform Infra-Red (FTIR) method [10]. In M/NM mines, the exposure limit to respirable silica-bearing dust is also dependent upon the % of silica if this content is greater than 1 %. The exposure limit considers three forms of crystalline silica (quartz, crystobalite, and tridimite) even though the first is the most common. The exposure limit is dependent upon the amount (%) of quartz(Q), cristobalite (C), and/or tridymite (T) present in the dust [11].

$$\text{Reduced standard mg/m}^3 = \frac{10\ \text{mg/m}^3}{[\%]\text{Q} + 2}$$

$$\text{Reduced standard mg/m}^3 = \frac{5\ \text{mg/m}^3}{[\%]\text{C} + 2}$$

$$\text{Reduced standard mg/m}^3 = \frac{5\ \text{mg/m}^3}{[\%]\text{T} + 2}$$

Quartz composes at least of 99 % of the silica in the MSHA samples and tridimite is rarely present. The mass of silica in M/NM mine samples is measured via an X-ray diffraction (XRD) method [12]. The XRD technique was selected for M/NM samples because of the lower impact of interferences than FTIR. Only respirable (dust) samples greater than 100 μg are subject to analysis for silica. Significant differences do exist between coal and M/NM compliance sampling for silica and respirable dust: in M/NM mines, the compliance samples are collected exclusively by MSHA inspectors; in coal mines, most of the samples are collected by MSHA inspectors, but operators are allowed by law to submit additional samples when a reduced dust standard has been proposed or applied.

In a recent study, Joy showed that the current MSHA approach for regulating coal miner exposure to respirable quartz does not protect miners from excessive exposure in all cases [13]. Specific situations where this problem arises include when the quartz content of the airborne dust increases due to changes in geologic conditions—i.e., more rock, or rock with higher quartz content, is extracted. The overexposure also occurs under reduced standard conditions when the presence of high silica has already been assessed. Adding to the prob lem, if the mine operator submits optional samples for quartz analysis, the pro cess may be extended by several weeks. Overexposure can cause adverse health effects and, potentially, dust concentrations below compliance standards may not be sufficient to protect the workers' health, based on the National Institute for Occupational Safety and Health (NIOSH) hazard review report [5].

The Office of Mine Safety and Health Research (OMSHR) NIOSH, recently initiated an effort for the development of end-of-shift techniques, specifically a laboratory-successful FTIR technique and an innovative elemental analysis technique [14,15]. An end-of-shift monitoring approach would allow operators to estimate the average concentration of silica in the area where they just sampled. Taking this one step further, OMSHR is also conducting research on methods for determining silica exposure during the shift. Due to limited sensitivity of most silica measurement methods, collecting an adequate sample mass in a short time can be problematic. A possible solution is the use of a high-volume sampler, and promising results have been published [16]. Another approach is the determination of silica % in an area of the mine or for a specific job by long-term static sampling and the use of this information in conjunction with real-time dust monitors. This approach requires constant or at least predictable silica content in the mine dust.

From a general perspective, the control and assessment of the exposure to silica in mining is dependent on the knowledge of two quantities: the respirable dust concentration and the % of silica in the respirable sample. While long-term trends of dust concentration and silica concentration in mining have been documented and analyzed [17–19], few studies analyzed in detail the % of silica in mine dust [19]. In a report from 1992, the National Occupational Health Survey of Mining examined the quartz content in bulk dust samples collected over six years in coal and M/NM mines [20]. The goal of the survey was to characterize health-related agents to which U.S. miners are exposed. The survey considered exclusively bulk and not respirable dust; therefore, it could be misleading to apply the findings of that study to respirable samples.

This manuscript investigates the % of silica in respirable dust samples collected in different mining industries and available in the MSHA database. Data gathered from 1997 to 2011 for both the coal and M/NM industries were used for this study. The findings provide valuable information for the development of specific sampling and analytical techniques for the monitoring of crystalline silica in the mining industry. In addition, the results can be useful for the design and evaluation of control technologies implemented for the reduction of crystalline silica exposure in mining.

Methodology

Information from MSHA archived respirable dust samples from 1997 through 2011 was retrieved from the MSHA Standardized Information System (MSIS) Samples database. Different information can be retrieved from the database regarding each sample. From the coal database it was possible to retrieve directly the % of silica in the dust collected and the respirable concentration relative to the sample. From the M/NM database it was possible to obtain the respirable dust concentration for each sample, while data on the % of silica in the dust was derived from the exposure limit associated with each sample. In addition, information related to the location where the sample was collected was retrieved. There are several known limitations of using a similar database: first and foremost the samples are taken for enforcement and not scientific reasons. This approach implies that: (1) MSHA inspectors sample workers suspected to be at the greatest risk of overexposure and not randomly [18], (2) samples collected in M/NM mines and containing less than 1 % of silica are coded differently and they are not used to calculate a reduced exposure limit. The use of these censored samples for the analysis of the % of silica in the mine dust would require several assumptions and potential introduction of errors. The authors decided to not consider these samples. In general, the MSHA database is a partially biased view of the true respirable mine dust and most likely is shifted towards the upper ends of the overall exposure distributions. Because the exposure is measured as silica concentration and it is function of the % silica and the concentration of respirable dust also the data of the % of silica in the dust can be partially biased. In spite of these shortcomings, the MSHA database is uniquely valuable in that it contains information on thousands of respirable dust samples collected in the US mining industry over a relatively long time period.

Results and Discussion

An analysis of the % of silica in the respirable dust samples collected in the mining industry was initiated by dividing the industry by commodities and locations. Figure 1 presents the % of silica in the M/NM respirable dust for the period 1997–2011 and Table 1 summarizes the statistical data of all the charts. Because the data of silica % in the dust is distributed in a lognormal fashion—Rankit test passed—the boxplots are reported in lognormal scale. The bottom and top of the box are the 25th and 75th percentiles (the lower and upper quartiles, respectively), and the band near the middle of the box is the 50th percentile (the median). The ends of the whiskers represent one standard deviation above and below the mean of the data and the additional dots represent 95 % and 5 %. In addition, the geometric mean of each set is included in Table 1 for each set. The silica % is substantially lower in the samples collected underground, and also shows a low variability. For every M/NM commodity, a single factor analysis of variance (ANOVA) ($\alpha = 0.05$) was conducted on the log-transformed data for the samples collected in the three locations. A significant difference was found among locations and a post hoc Tukey–Kramer analysis identified underground as different from both mill and surface locations. The reason for this is not clear, but it is likely a function of mine geology, as well as mining practices (i.e. methods of excavation and ore handling). It is also possible that the crushing and refining processes and subsequent handling of the ores in mill and surface locations can generate a respirable dust that is richer in silica.

The Tukey–Kramer analysis underscored also that the % of silica in samples collected in mill and surface locations are different for both stone and

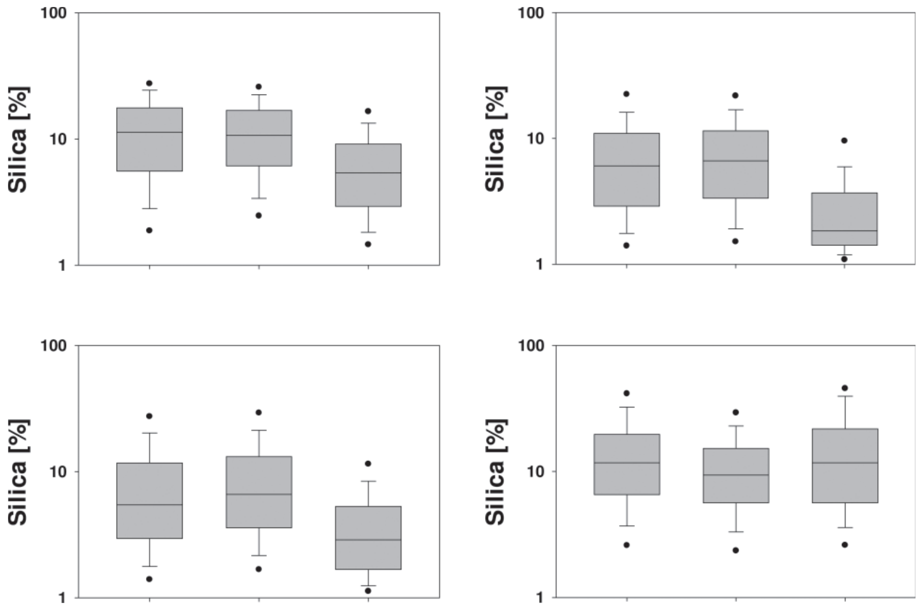

FIG. 1—*Percent of silica in respirable samples collected in M/NM mines from 1997 to 2011. For each chart the data are (left to right): mill, surface, and underground. Metal mines (top left), nonmetal mines (top right), stone mines (bottom left), S&G mines (bottom right).*

S&G industry. These findings can have implications in the development of monitoring strategies: in specific underground locations, the estimation of an area's silica % by using long-term stationary sampling might produce relatively good accuracy. On the other hand, simply employing dust monitors in mill and surface locations and assuming constant and reliable information on silica % might induce poor estimation in the exposure to silica. In this case, a timely measurement of the silica % in the dust by the end-of-shift approach could be beneficial.

For all the M/NM industries, the samples collected in mill and surface locations showed similar geometric mean % of silica (Table I). The geometric mean is close to 10 % for both metal and S&G mines (11.2 %, 9.0 %, 9.6 %, and 9.6 %, respectively for mill and surface locations) and it is substantially lower for nonmetal and stone mines in both locations. The upper quartile is highest for samples collected in S&G mill locations where in general high values for all the parameters are found. The difference between the upper and lower quartile is a good indication of the variability in the silica % in non-underground locations. This value is in general around 10 % and, on average, slightly higher for metals (11.4 %) and S&G (11.4 %) dust samples than for nonmetal (8.1 %) and stone (9.1 %). This implies that the industries with higher

TABLE 1—*Statistical summary data on the percent of silica in respirable dust samples collected in M/NM mines from 1997 to 2011.*

			Mill	Surface	Underground	Total
Metal		*Sample #*	*1328*	*1016*	*866*	
	% silica	Geometric Mean	9.6	9.6	5.3	8.2
		Median	11.3	10.7	5.4	9.2
		1 quartile	5.6	6.1	2.9	4.6
		3 quartile	17.6	16.9	9.1	15.2
		5 %	1.9	2.5	1.5	1.8
		95 %	27.4	25.8	16.5	25.8
Nonmetal		*Sample #*	*2431*	*1101*	*156*	*3688*
	% silica	Geometric Mean	5.8	6.2	2.3	5.7
		Median	6.1	6.6	1.8	6.0
		1 quartile	2.9	3.4	1.4	2.8
		3 quartile	11.0	11.5	3.7	11.0
		5 %	1.4	1.5	1.1	1.4
		95 %	22.4	21.8	9.4	21.8
Stone		*Sample #*	*8188*	*16 131*	*1084*	*25 403*
	% silica	Geometric Mean	5.9	6.8	3.1	5.3
		Median	5.5	6.6	2.9	6.0
		1 quartile	3.0	3.6	1.7	3.2
		3 quartile	11.7	13.2	5.3	12.3
		5 %	1.4	1.7	1.1	1.5
		95 %	27.4	29.3	11.5	27.4
S & G		*Sample #*	*6807*	*11 048*	*134*	*17 989*
	% silica	Geometric Mean	11.2	9.0	11.2	9.8
		Median	11.7	9.4	11.7	10.2
		1 quartile	6.6	5.6	5.6	5.9
		3 quartile	19.7	15.2	21.7	16.9
		5 %	2.6	2.4	2.8	2.4
		95 %	41.5	29.3	45.6	35.0

silica % (median) in the dust also have a higher likelihood of variability in silica %. In order to compare the % of silica in the dust samples collected in different M/NM commodities, a single factor ANOVA ($\alpha = 0.05$) was conducted by considering a single combined set of samples from each commodity: the post hoc Tukey–Kramer analysis showed that the means for each industry are significantly different. This finding is probably affected by the very large number of samples for each industry.

The MSHA database also reports the year when each sample was collected, which allowed for an investigation of how the silica % in the dust evolved through the years in the respirable dust samples collected in the M/NM

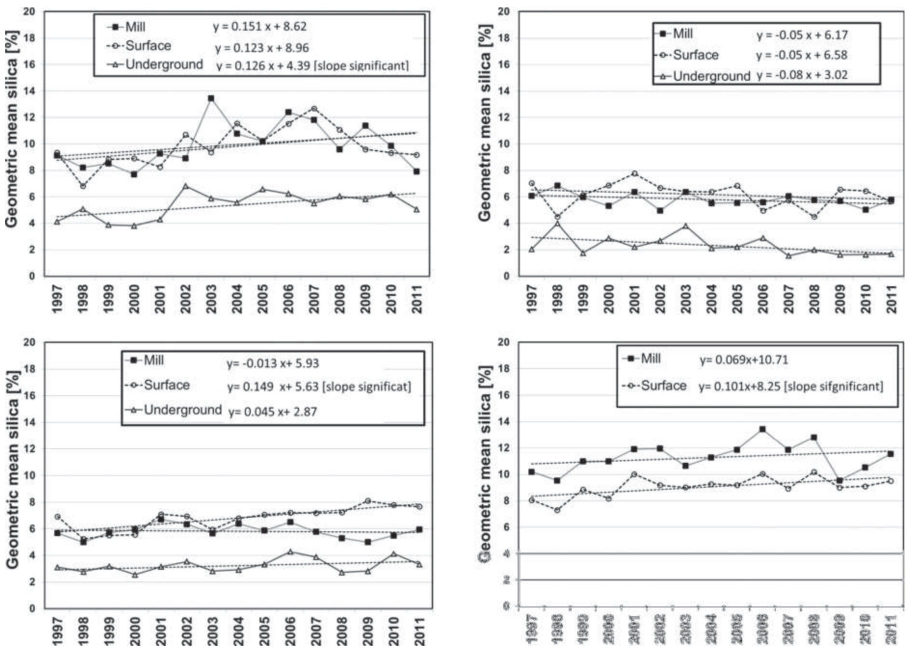

FIG. 2—*Year by year geometric mean % of silica in the M/NM dust from 1997 to 2011. Metal mines (top left), nonmetal mines (top right), stone mines (bottom left), S&G mines (bottom right).*

industry (Fig. 2). For the metal and S&G industries and for the samples collected in surface stone mines, the regression for each line underscores a pronounced positive trend—an increase in the geometric mean % of silica—with time for the years 1997–2011. An analysis of regression ($\alpha = 0.05$) showed that the slope are significantly different than zero only for the samples collected in underground metal mines, in surface stone mines, and in surface S&G locations.

The % of silica in the respirable dust samples collected between 1997 and 2011 in coal mines is substantially different for underground and surface coal locations (Fig. 3). The variability is significantly higher for samples collected in surface locations. The data summarized in Table 2 shows that the geometric mean for samples collected in surface locations is slightly higher than for underground (5.6 versus 4.7)—and that the difference is significant (single factor ANOVA $\alpha = 0.05$) In underground locations, 95 % of the samples did not show a silica % higher than 20 %, while this value reached almost 40 % for surface locations. The % of silica in the respirable dust is also much more variable if the sample was collected on the surface. In addition, 50 % of the samples around the median showed a silica % between 17 % and 1.7 %. For underground samples, these values were narrower: 8.4 % and 2.8 %. An

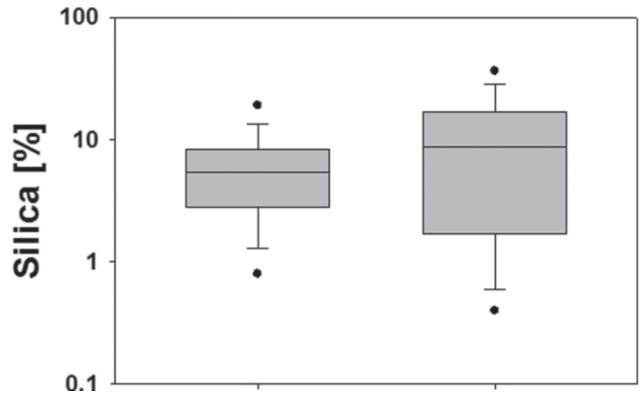

FIG. 3—*Percent of silica in respirable samples collected in coal mines from 1997 to 2011*.

explanation for this finding could be found by considering the different mining process—i.e., the presence of non-coal silica-rich dust is much more prevalent in surface coal mines or surface locations of underground coal mines. Surface mine operators might need to mine through a substantial amount of rock material in order to retrieve sufficient coal and this process can generate dust with variable silica %.

Figure 4 provides a means for visualizing how the median silica % in the respirable dust in coal mines varied yearly from 1997 to 2011. For both samples collected in surface and underground locations, the geometric mean of the silica % decreases during the years with a slope that is significantly different than zero. The decrease is more pronounced in the first years for both surface and underground samples. For this reason, a second analysis was conducted only on samples collected between 2003 and 2011; in this case, the positive relationship is still significant, but substantially reduced for underground and reversed for surface location.

TABLE 2—*Statistical summary data on the percent of silica in respirable dust samples collected in coal mines from 1997 to 2011*.

		Underground	Surface	Total
	Sample #	*66 721*	*11 104*	*77 825*
% silica	Geometric Mean	4.7	5.6	4.8
	Median	5.4	8.7	5.6
	1 quartile	2.8	1.7	2.7
	3 quartile	8.4	17.0	9.1
	5 %	0.8	0.4	0.7
	95 %	19.2	36.6	22.6

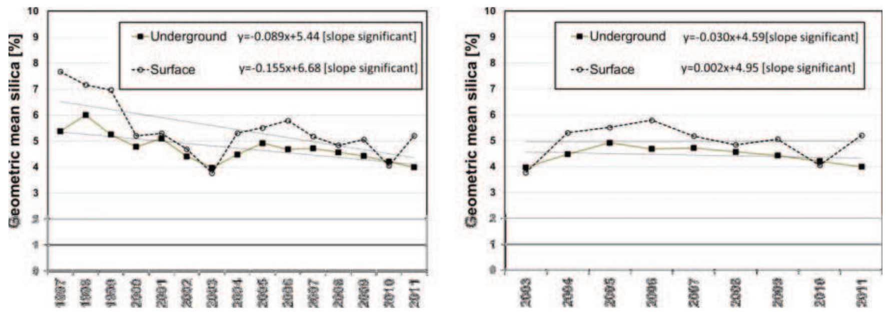

FIG. 4—*Year by year geometric mean % of silica in the coal mine dust from 1997 to 2011 (left) and from 2003 to 2011 (right).*

The relationship between the % of silica in the respirable dust samples collected and the number of samples collected was also explored. This analysis investigates how the sampling strategy by MSHA inspectors is affected by the industry and if the % of silica in the dust is a factor in this selection. The geometric mean for the % of silica in dust samples collected every year from 1997 to 2011 in each M/NM industry and location were plotted against the relative number of samples (Fig. 5 left panel). Preliminary analysis of the plot showed how a positive correlation was visually detected only if the number of samples were log-normally transformed; in this case, a regression slope significantly different from zero was assessed. In a similar fashion, the geometric mean of the % of silica in dust samples collected every year from 1997 to 2011 in coal mines were compared with the number of samples collected. In this case, the results were divided between samples collected in underground and surface locations (Fig. 5 central and right panel); a positive significant relationship was found even without the log-transformation of the number of samples.

As described in the Introduction, silica % is the metric used to calculate the reduced dust exposure limit in both the coal and M/NM mine industries,

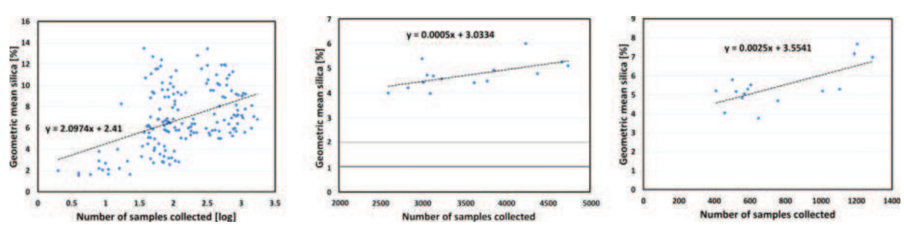

FIG. 5—*Relationship between silica % in the dust samples and number of samples collected in the mining industry: (left) samples collected in M/NM industry between 1997 and 2011, (central) samples collected in underground coal mines between 1997 and 2011, (right) samples collected in surface coal mines between 1997 and 2011.*

and the actual respirable dust concentration is used in conjunction with the reduced limit to verify the compliance status. For this reason, it is important to investigate the relationship between the respirable dust concentration and the silica % in the dust. The data of the two values were plotted for each sample collected from 1997 to 2011 in metal, nonmetal, stone, and S&G mines (Fig. 6). The black line in each chart represents the dust standard. Intuitively, the samples located on the right of the line were characterized by a silica content higher than 100 μg. In each chart, the % of silica is inversely correlated with the dust concentration—i.e., the lower the dust concentration, the higher the variability of the silica %. This data trend is similar for every location in the four industries. The similarity of the boundaries of the area populated by data and the reduced standard lines is evident.

Possible explanations for this pattern can be made. The first explanation is based on the reduced standard formula: if a mine is regulated under a reduced standard, its dust is more likely to have a high silica % and consequently the mine is required to keep the dust concentration low. In other words, the trend proves that the regulation as it is structured is effective: the higher the % of silica, the lower is the respirable dust concentration. On the other hand, it is more difficult to explain the complete absence of samples with both high respirable mass concentration and high silica %—in other words, to understand why the samples in non-compliance status also present the inverse relationship. The periodic MSHA inspections provide the operators information on the % of silica in the dust. This value is used to generate the reduced dust standard, but it might change before the following MSHA inspection. This change can move the point in the chart from the left side of the reduced standard line to the right side. This transposition from the left side (in compliance) to the right side (not in compliance) of the line can occur even if the same dust concentration is maintained or, in other words, even if the same dust control strategy is implemented. Described from a different perspective, the charts show that the % of silica in M/NM mine dust is extremely variable and it can span from a few % up to 80–90 %. Because of this high variability, it is extremely difficult for operators to predict with accuracy and precision the % of silica in the dust and its evolution over time with the tools currently available. However, the operators might have knowledge of an area with a high silica % dust, and for this reason, the data trend with a pattern similar to the reduced standard line. However, the lack of timely characterization in silica % connotes a *limited* and *non-complete* knowledge by operators of the dust's characteristics present in the mine.

Other possible explanations for the trends are connected with dust control technologies and their performance. If the efficiency of the dust control approaches is more selective to dust with lower silica percentage because of size distribution effects, the results of their application is a lower respirable dust concentration but a higher silica %. Along the same lines, different transport properties for aerosols with low and high % in silica could explain this effect. The transport of an aerosol from the generation of the dust to the sampler is affected by physical (size) properties—i.e., if the aerosol with a higher % in silica is more likely to reach the sampler because of its smaller size, there will be a lower mass concentration but a higher silica %. The authors did not find any reference to support these hypotheses but feel they should be further explored.

A similar analysis of the relationship between the % of silica and the relative respirable dust concentration has been carried out on samples collected in surface and underground coal mine locations (Fig. 7). The black line is the reduced standard for coal dust affected by the % of silica. The samples at the right of the black line have a silica content higher than 100 μg. As noted for the M/NM industry, the trend for the % of silica in coal dust samples also has a distinctive evolution—the lower the dust concentration, the higher the

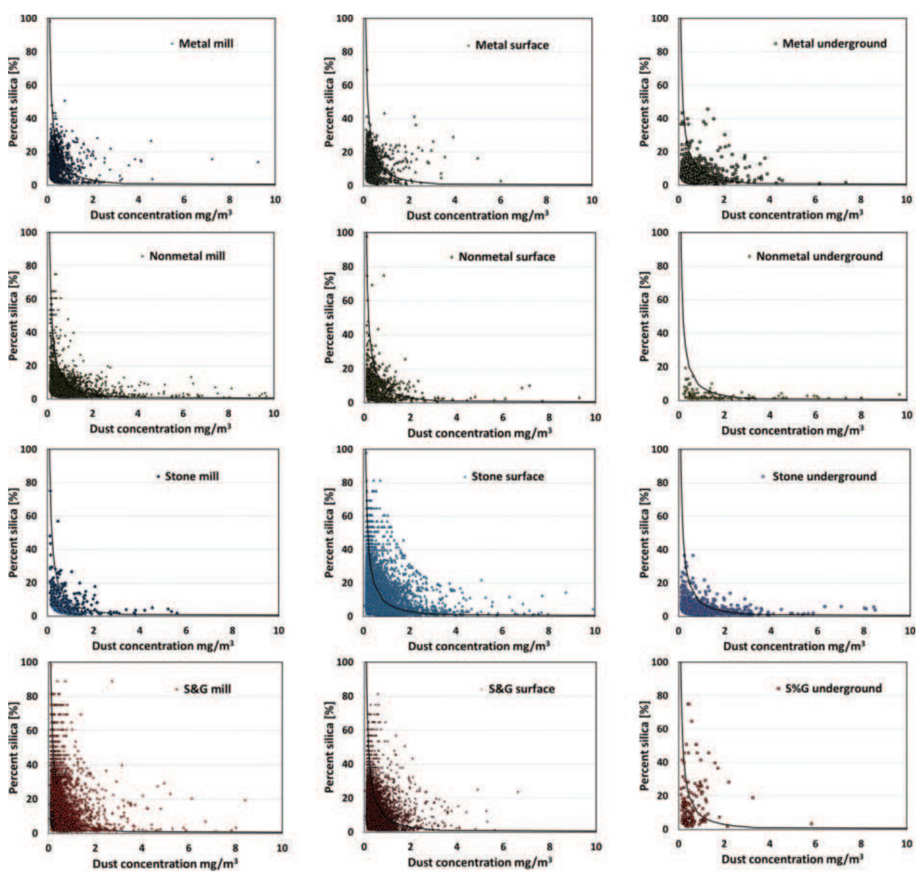

FIG. 6—*Percent of silica versus respirable dust concentration for M/NM dust samples.*

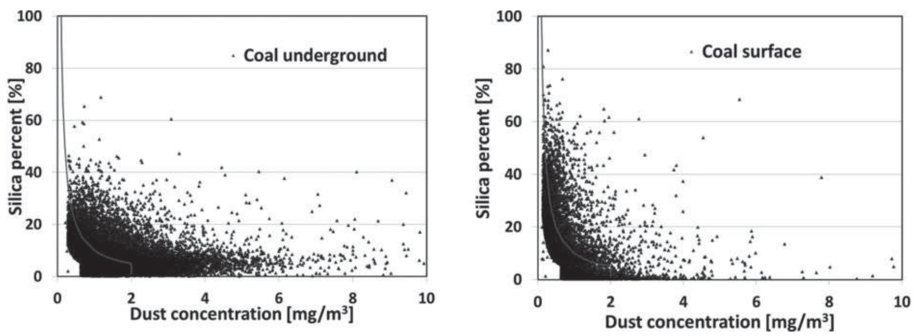

FIG. 7—*Percent of silica versus respirable dust concentration for coal dust samples.*

variability of silica %. This is particularly true for surface coal samples where this inverse trend is more pronounced. This reinforces the idea that the dust samples collected are characterized by a % of silica that is not random.

As proposed above, it is possible that the changing conditions in the % of silica in the dust do not allow the mine operators to predict the dust concentration level below the reduced standard. In other words, the lack of certainty about the silica % in the dust is a possible limiting factor for the operators in maintaining the concentration below the reduced dust standard. Also in this case, the hypothesis of the effect of more selective dust control approaches towards samples less rich in silica dust cannot be excluded. A study conducted in 1987 by Penn State University on the size and elemental composition of airborne coal mine dust showed coal mine dust present underground in bi-modal distributions with a smaller mode around 2 μm [21]. A previous study on silica (more than 90 % silica) particle distribution in respirable coal mine dust samples (surface and underground) showed a median around 1 μm [22]. It should be noted that these studies are more than 20 years old and they might not reflect the current conditions in the coal mine industry. Nevertheless, their findings are an indication of the possible co-presence in the coal mine atmosphere of different separate aerosols with different % in silica, which could explain the trends in Fig. 7. These findings should be considered when designing specific monitoring and control technology strategies with the focus on silica dust in coal mines.

Summary

This study analyzed the % of crystalline silica in respirable dust samples collected by MSHA inspectors from around the U.S. between 1997 and 2011. The results for the M/NM industry showed that the % of silica in the dust was significantly higher and more variable for samples collected in surface and mill locations than for those underground. The % was also found to be higher and more variable in sand and gravel and metal mines, as opposed to other M/NM mines. The % of silica in respirable samples collected in underground metal mines, surface stone mines, and S&G surface mines is slowly increasing over the years of the study. In coal mines, the silica % is significantly higher and more variable for samples collected in surface locations. While the % of silica in samples collected from 1997 to 2011 has been decreasing for both surface and underground locations, the trend stopped or reversed in the last 8 years. The analysis also showed that a positive relationship exists between the geometric mean of the % of silica in respirable dust samples collected in a certain year in both M/NM and coal industry and the number of samples collected. For both coal and M/NM dust samples, the relationship between the silica % and the respirable dust concentration showed a distinctive negative trend: the lower the dust concentration, the higher the variability in the % of silica. A few explanations were proposed to explain these trends, and the possible mathematic relationships need to be further evaluated and verified via specific testing or data analysis by considering different variables.

Acknowledgments

The mention of any company or product does not constitute an endorsement by the National Institute for Occupational Safety and Health. The findings and conclusions in this paper are those of the authors and do not necessarily represent the views of the National Institute for Occupational Safety and Health.

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