Original Article

Uncertainty Estimation and its Effect to Respirable Crystalline Silica Exposure Between Direct and Indirect Method

Suhaily Amran*[1, 2], Mohd Talib Latif[2,3], Shoffian Amin Jaafar[1], Firoz Khan[3], A.M Leman[d]

[1]Consultation, Research and Development Department, National Institute of Occupational Safety and Health (NIOSH), Lot 1, Jln 15/1, Section 15, 43650 Bandar Baru Bangi, Selangor, Malaysia
[2]School of Environmental and Natural Resources Science, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
[3]Centre for Tropical Climate Change System (IKLIM), Institute of Climate Change, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia.
[a] Faculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia (UTHM), Pagoh Higher Education Hub, Johor, Malaysia

* Corresponding author: suhailyamran@gmail.com

Article history

Received 29/1/2023
Accepted (Panel 1) 16/6/2023
Accepted (Panel 2) 17/7/2023

ABSTRACT : *Due to its carcinogenic nature, many countries establish occupational exposure limits for respirable crystalline silica (RCS) at very low levels, typically ranging between 0.025 and 0.1 mg m^{-3}. Consequently, industrial hygienists encounter substantial challenges in selecting suitable sampling methods to adhere to these stringent exposure limits. Factors to consider include selecting between direct or indirect methods and the uncertainties associated with each method. This study aimed to determine the correlation between RCS exposure using direct and indirect analysis methods, validate the application of both methods to underpin compliance status, and evaluate the effects of sampling and analytical uncertainty on exposure levels. Sampling was performed among 31 crusher operators at six quarries, with each worker equipped with a pair of integrated sampling devices to facilitate parallel comparisons between direct and indirect methods. Exposure data from direct and indirect methods showed substantial correlation ($p<0.05$, $r^2 = 0.82$) with no significant differences ($p > 0.05$). For the direct method, 35.9% of crusher operators exceeded the RCS exposure limit compared to 30.9% for the indirect method. The total coefficient variance (CV_T) was 0.10 and 0.09 for the direct and indirect methods, respectively. For both methods, CV_T was influenced more by the coefficient of variance for analytical procedures (CV_A) than by the coefficient of variance for sampling procedures (CV_P). Integration of CV_T values into the upper confidence limit (UCL) calculations revealed an increased number of non-compliance exposures for both methods. The indirect method demonstrated lower uncertainty and better quality assurance compared to the direct method. However, no significant differences ($p>0.05$) were found among the field samples. Industrial hygienists may choose either method that meet their criteria concerning quality, timing, or cost.*

Keywords : *Direct Method, Indirect Method, MDHS 101, NMAM 7500, Respirable Crystalline Silica*

1.0 INTRODUCTION

Exposure to silica has been linked to various respiratory conditions, including silicosis and lung cancer (ALS, 2016; Dong et al., 1995; Kane, 1997; WHSQ, 2011). Recent research suggests that silica exposure may also lead to autoimmune diseases (Rocha-Parise et al., 2014). The International Agency for Research on Cancer (IARC) classified crystalline silica in the form of quartz or cristobalite as a Group 1 carcinogen in 1997 (IARC, 1997; Lin et al., 2012). The carcinogenic properties of silica and the need to limit its exposure to workers have led to the establishment of low occupational limits ranging from 0.025 mg m^{-3} to 0.1 mg m^{-3}. For instance, The American Conference of Governmental Industrial Hygiene (ACGIH) established a threshold limit value (TLV) of 0.025 mg m^{-3} for respirable crystalline silica (RCS) (ACGIH, 2015). Many countries, such as the United Kingdom (HSE 2011), Australia, Belgium, Canada, France, Ireland, Denmark, Singapore, Malaysia (DOSH 2000), Spain, and Sweden, maintain an occupational limit for respirable quartz at 0.1 mg m^{-3}. The limit value for Austria, Hungary, and Switzerland is 0.15 mg m^{-3}, while Finland, South Korea, and Argentina adhere to a limit of 0.05 mg m^{-3} (IFA, 2015; Maciejewska, 2008). Monitoring substances with low occupational thresholds presents challenges for field and laboratory personnel. Industry stakeholders have expressed concerns about the efficacy of current methods in ensuring compliance monitoring and detecting violations at very low levels (Cox Jr et al., 2015).

Selecting the most appropriate sampling and analytical techniques is crucial for achieving minimal exposure limits. The primary concerns are the sampling and analytical errors inherent to each method. Errors at such low occupational limits may introduce uncertainty into the compliance status of exposure results. Uncertainty, in this context, is a parameter associated with the result of a measurement that characterises the dispersion of a value that can reasonably be attributed to the measurement. Uncertainty relates to the concept of doubt (Ellison et al., 2000). There is a lack of information regarding the errors occurring in measurements performed under real conditions (Garcia et al., 2013). Furthermore, the selection of an RSC sampling method necessitates experienced and critical judgement. Consequently, there is ongoing debate between selecting direct (HSE, 2005) or indirect methods (NIOSH, 2003a, b) and choosing between X-ray diffraction (XRD)(NIOSH, 2003b) or infrared (NIOSH, 2003a) methods.

XRD and infrared are commonly utilised analytical techniques for quantifying RCS (Key-Schwartz et al., 2003; Madsen et al., 1995; Miller, 2014; Page, 2006; Verpaele and Jouret, 2013). However, XRD is a more accurate and precise technique compared to the infrared technique (Kuo et al., 2010). Analysis of crystalline silica is considered a selective method as it relies on the individual crystalline properties of the substance (HSE, 2005; Markku et al., 2012). Frequently employed methods include the National Institute of Occupational Safety and Health (NIOSH) Manual Analytical Method No.7500 (NMAM 7500) for silica, crystalline, XRD via filter deposition (NIOSH, 2003b), and Methods for the Determination of Hazardous Substance Guidance No.101 (MDHS 101) for RCS in Airborne Dust (HSE, 2005). NMAM 7500 is fully validated and the only method with any legal significance (Smith, 1992).

In the direct method, field samples are analysed directly without any pretreatment, while the indirect method involves a series of pretreatments such as digestion, filtration, and/or ashing of field samples. Although the sampling techniques of the direct and indirect methods are similar, the analytical procedures differ (Amran et al., 2016). Direct analytical methods minimise sample handling; however, standards must be prepared under conditions similar to those of the samples (Kaufer et al., 2005). An example of a direct method is MDHS 101, where standard filters can be produced in an exposure chamber to simulate conditions similar to those of sampling (HSE, 2005). Indirect methods involve pretreatment of filter membranes, with NMAM 7500 being an example that employs XRD. Another indirect method, NMAM 7602, utilizes Fourier-transform infrared (FTIR) spectroscopy for analysis (NIOSH, 2003a). Kaufer et al. (2005) conducted a study evaluating direct and indirect methods using infrared technology, and a direct method using X-rays. In that study, results obtained through the indirect method using infrared techniques were approximately 13% lower on average compared to the mean results of two direct methods using infrared and XRD.

This study aimed to assess the correlation between direct and indirect methods for measuring RCS exposure, validate the application of both methods for compliance assessment, and examine the impact of sampling and analytical uncertainty on the resulting exposure data.

2.0 METHODOLOGY

The study protocol was approved by the NIOSH Malaysia Ethics Committee. Sampling was conducted at six quarries in Kuala Lumpur and Selangor, located in the central region of Peninsular Malaysia. All quarries used jaw crushers at primary crusher plants and Hydrocone® crushers at secondary and tertiary crusher plants. Each crusher was equipped with water sprinklers as a dust suppression system.

Sampling involved 31 crusher workers. The minimum number of samples in a similar exposure group (SEG) was determined based on the NIOSH USA Occupational Exposure Sampling Strategy Manual, 1977 (NIOSH 1977). For a sample size representing the top 10% with a confidence level of 0.95 ($\alpha = 0.05$) and an estimated population of over 200 crusher operators, the minimum sample size of 29 workers was required. This sample size exceeded the Department of Occupational Safety and Health (DOSH) guidelines for Monitoring of Hazardous Substances, which recommends 18 samples representing the top 10% with a confidence level of 0.9 (DOSH 2002). Crusher operators were selected owing to their excessive exposure to quarry dust. This group was classified as a similar exposure group (SEG) considering the exposure pattern and duration. SEG is defined as a group of workers with the same general exposure profile to an agent because of the similarity in the frequency of tasks performed, materials used, and processes (Bullock & Ignacio 2006). Sampling was conducted throughout the 8-hour work shift. Crusher plant activities were monitored and controlled from an enclosed air-conditioned room where operators spent most of their time. Occasionally, operators performed troubleshooting or maintenance outside the control room, exposing them to dust and crystalline silica produced during crushing, grinding, screening, loading, and unloading processes.

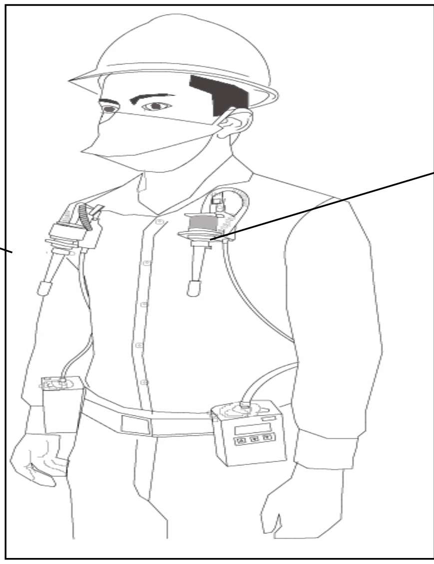
The sampling and analysis for the direct method strictly followed the Methods for the Determination of Hazardous Substance Guidance No.101 (MDHS 101) (HSE, 2005), while the indirect method followed the NIOSH Manual Analytical Method No.7500 (NMAM 7500) which uses crystalline silica by X-ray diffraction via filter deposition (NIOSH, 2003b). Both methods utilised XRD for analysis. Table 1 presents the comparison criteria between the direct and indirect methods.

Table 1 Comparison Between Direct and Indirect Methods

	Direct Method	Indirect Method
Reference method	MDHS 101	NMAM 7500
Method Published by	HSL, HSE, UK	NIOSH USA
Sampling pump	Standard flow	Standard Flow
Filter type	25 mm polyvinyl chloride (PVC) filter	37 mm polyvinyl chloride (PVC) filter
Cyclone type	SKC GS3 cyclone	SKC GS3 cyclone
Standard preparation by	By exposure chamber	By funnel filtration system
Sample preparation by	Direct filter method	Digestion with acid and ashing
Analytical instrument	XRD	XRD

Each worker was provided with two integrated sampling sets. All samples were collected in pairs to create parallel sets for comparison between the direct and indirect methods (Fig. 1). Each set consisted of a standard flow SKC sampler (AirChek XR5000, 2013, Dorset, USA), attached to an SKC GS3 cyclone and a 3-piece cassette loaded with a 5.0 µm polyvinyl chloride (PVC) filter. We used an SKC GS3 cyclone flow rate of 2.75 L min^{-1} as the separating device for the respirable fraction. The specification patterns matched the definitions of the respirable conventions (SKC, 2014). Volume calculations were based on the average pre- and post-calibration flow rates (SKC, 2014) and sampling duration. Air velocity and humidity were measured to determine environmental factors on the monitoring day, using an electronic integrated hot-wired anemometer and hygrometer (TSI, 8386-M-GB, 2014, Shoreview, USA). All equipment was in good condition, calibrated, and traceable to international standards. The final results were corrected for normal temperature (25 ˚C or 298 K) and pressure (760 mmHg).

Assembly for indirect method. A 37 mm PVC filter in 3-piece cassette equipped with a GS3 cyclone



Assembly for direct method. A 25 mm PVC filter in a 3-piece cassette equipped with a GS3 cyclone

Figure 1: Assembly of Sampling Pump on Workers

Quartz measurements were performed using an XRD (Rigaku, Multiflek, Tokyo, Japan) equipped with six holder autosamplers and a sample spinner. An X-ray generator was used to obtain a maximum output of 2000 watts (tube voltage 40 kV; tube current 50 mA). The scanning range of the primary quartz peak was 26.0°–27.14°. Results were obtained from the integral signal intensities.

In total, 31 pairs of personal samples were collected, with each pair representing both the direct and indirect methods. The main difference between the direct and indirect methods related to sample preparation. In the direct method, the filters were analysed using XRD without any pretreatment procedures. Analyses using the indirect method involved acid treatment, ashing, and filtration processes to transfer dust from the 37 mm sampling filter onto the 25 mm XRD filter holder. The recovery rates for the direct and indirect methods were 95.6% and 90.7%, respectively. The total combined standard uncertainty was 19.54% for the direct method and 17.14% for the indirect method. Replication for airborne samples were not performed as it is nearly impossible to duplicate the same conditions owing to the variability of worker movement, machine operation, and environmental factors such as temperature, humidity, and wind direction. However, laboratory investigations, such as recovery and reproducibility, were based on the replication of seven to ten samples. Table 2 lists the performance characteristics of the direct and indirect methods.

Table 2 Performance Characteristics Between Direct and Indirect Methods

Performance Characteristic	Unit	Direct method	Indirect method
Calibration range	µg	17.33-570	10-500
Calibration slope (r^2)	-	0.8	0.7
Calibration regression	-	0.996	0.999
Limit of detection (LOD)	µg	13.04	4.11
Limit of Quantification (LOQ)	µg	43.46	13.70
Recovery	%	90.4	88.6
Precision	%	5.48	3.64
Trueness	%	5.97	6.77
Accuracy	%	±15	±13
Combined uncertainty component, U	%	8.10	7.69
Expanded Uncertainty, *U (k* value =2*)*	%	16.20	15.37

Uncertainty from recovery was obtained using the following equation:

$$u(\bar{R}_m) = \bar{R}_m \times \sqrt{\left(\frac{s_{obs}{}^2}{n \times \bar{C}_{obs}{}^2}\right) + \left(\frac{u\left(C_{spike}\right)}{C_{spike}}\right)^2}$$

where:

$C_{obs}{}^2$ = mean of the replicate analyses of the spiked sample
C_{spike} = concentration of the spiked sample
S_{obs} = standard deviation of the results from replicate analyses of the spiked sample
$u(C_{spike})$ = standard uncertainty in the concentration of the spiked sample.
n = number of replicates

Combined uncertainty, $u(y)'$ was obtained using the following equation:

$$u(y)' = \sqrt{u\,(p)^2 + u\,(q)^2 + u\,(r)^2 + \ldots}$$

Where $u(p)$, $u(q)$, $u(r)$ represents the uncertainty for each factor

3.0 RESULTS AND DISCUSSION

3.1 Comparison of Exposure Pattern Between Direct and Indirect Methods

Fig. 2 shows the exposure patterns of personal samples using direct and indirect methods. The highest personal respirable dust exposure for the direct method was 0.41 mg m^{-3} while for the indirect method, it was 0.26 mg m^{-3}. The lowest exposure was 0.02 mg m^{-3} for the direct method and 0.17 mg m^{-3} for the indirect method. Overall, the exposure patterns fluctuated without any distinct trends. Although exposure among similar groups is expected to be similar, slight changes in environmental conditions and worker movements may cause irregularities. This variability was supported by Garcia et al. (2013), who attributed the findings to in situ errors, such as worker movement, machine influences, technology used, and irregular environmental factors, such as wind velocity, temperature, and humidity (Garcıa et al., 2013).

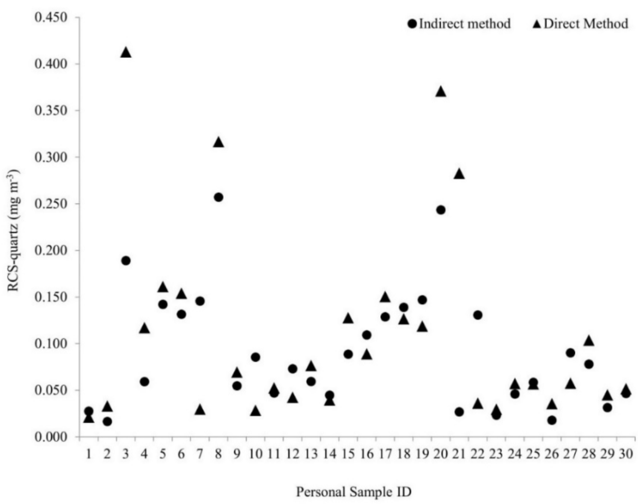


Figure 2: A Plot of Personal RCS-Quartz Exposure Based on Direct and Indirect Methods

3.2 Correlation and Regression Between Direct and Indirect Methods

The Wilcoxon signed-rank test revealed no significant differences ($p>0.05$) between the exposure data from both direct and indirect methods. Furthermore, the Spearman rank-order showed a positive correlation ($p<0.05$) between the two datasets. Regression analysis of personal exposure to RCS-quartz based on the two methods (Fig. 3) indicated a high correlation between the datasets ($r^2 = 0.8172$).

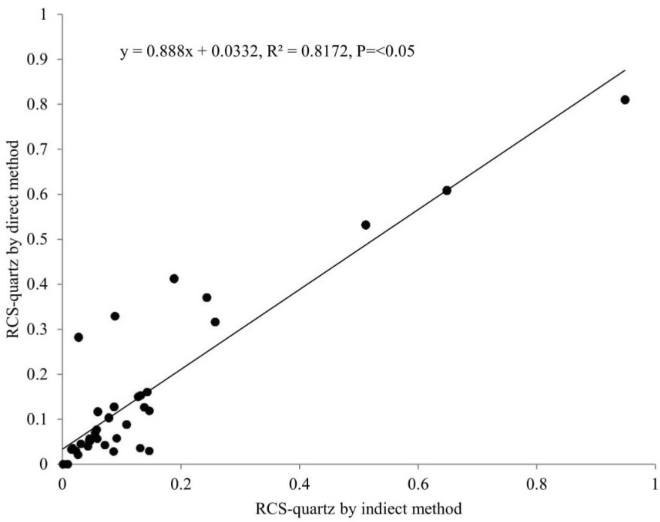


Figure 3: A Regression Plot of Personal RCS-Quartz Exposure Based on Two Methods

3.3 Compliance Status of RCS-Quartz Between Direct and Indirect Methods

Table 3 provides descriptive and inferential statistics for the time-weighted average (TWA) personal exposure to personal respirable dust based on the direct and indirect methods. The datasets displayed different arithmetic means (AM) and geometric means (GM). The AM for RCS-quartz exposure using the direct method was 0.106 mg m^{-3}, exceeding Malaysia's PEL. However, the AM for RCS-quartz exposure using the indirect method was 0.092 mg m^{-3}, which is lower than the PEL of 0.1 mg m^{-3} according to Malaysian requirements under the Occupational Safety and Health (Use and Standard of Exposure of Chemical Hazardous to Health) Regulations 2000 (USECHH Regulations 2000) (DOSH, 2000). The overall exposure range was between 0.010 and 0.413 mg m^{-3}. The highest AM for personal RCS-quartz exposure using the direct method was 0.189 mg m^{-3} in Quarry A, followed by 0.170 mg m^{-3} in Quarry E. The lowest AM was 0.058 mg m^{-3} recorded at Quarry F, followed by 0.071 mg m^{-3} in Quarry C. For the indirect method, the highest AM for RCS-quartz exposure was 0.163 mg m^{-3} in Quarry D, followed by 0.125 mg m^{-3} in Quarry B. The lowest AM was 0.055 mg m^{-3} in Quarry F, followed by 0.058 mg m^{-3} in Quarry E.

Table 3 Summary of Personal Exposure to RCS-Quartz Using Direct and Indirect Methods

Quarry	Method	N	AM (mg m^{-3})	GM (mg m^{-3})	GSD	Range (mg m^{-3})		% ≥ PEL
						Min	Max	
A	Direct	3	0.136	0.066	4.98	0.021	0.413	39.8
	Indirect	3	0.071	0.043	3.69	0.016	0.188	25.9
B	Direct	7	0.138	0.092	2.46	0.028	0.316	46.2
	Indirect	7	0.125	0.110	1.73	0.055	0.257	57.1
C	Direct	6	0.071	0.065	1.58	0.040	0.128	17.6
	Indirect	6	0.069	0.065	1.44	0.043	0.108	11.8
D	Direct	4	0.189	0.170	1.70	0.119	0.371	84.3
	Indirect	4	0.163	0.158	1.34	0.128	0.243	94.3
E	Direct	3	0.170	0.067	3.48	0.030	0.283	37.5
	Indirect	3	0.058	0.044	2.59	0.024	0.131	19.3
F	Direct	6	0.058	0.055	1.43	0.036	0.104	4.8
	Indirect	6	0.055	0.047	1.82	0.018	0.091	10.7
Overall	Direct	31	0.106	0.073	2.42	0.010	0.413	35.9
	Indirect	31	0.092	0.067	2.26	0.010	0.258	30.9

RCS-quartz, Respirable Crystalline Silica–quartz; AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; PEL, permissible exposure limit [a]; 8-hour TWA based PEL by Malaysian USECHH 2000 at 0.1 mg m^{-3}

Figs. 4-7 compare RCS-quartz exposure between direct and indirect methods using log probability plots, least-squares best-fit lines, and log-normal distributions. The GM for RCS-quartz exposure using the direct method of analysis was 0.073 mg m^{-3} and 0.067 mg m^{-3} for the indirect method. The geometric standard deviation (GSD) for RCS-quartz for the direct method and indirect method was 2.42 mg m^{-3} and 2.26 mg m^{-3}, respectively. The GSDs showed that the data represent similar exposure groups; however, the direct method had greater variance compared to the indirect method. These findings are reliable and may be used for inferential purposes. The variability of results may be due to environmental fluctuations and errors during sampling and analysis. Using the direct method, 35.9% of crusher operators were exposed to RCS-quartz levels exceeding the occupational limit (Figs. 4 and 6). However, when using the indirect method, 30.9% of crusher operators were exposed to RCS-quartz levels above the PEL (Figs. 5 and 7), based on Malaysian USECHH 2000 standards.

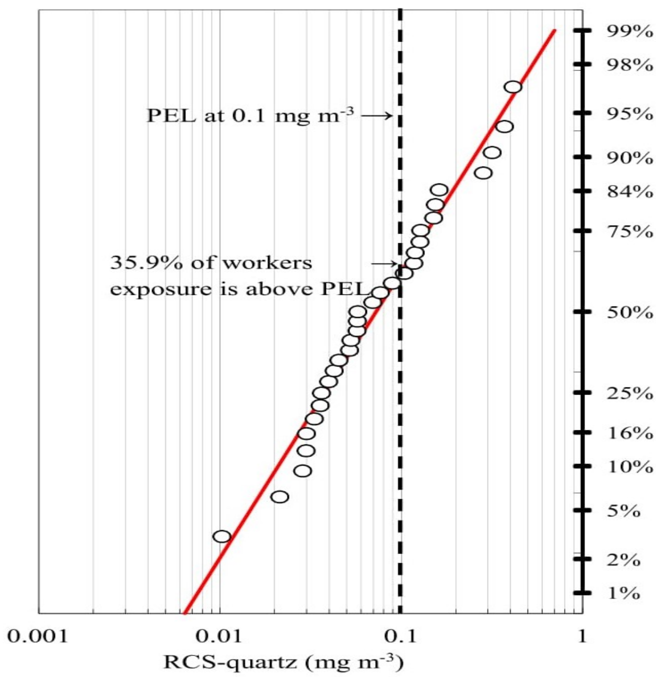


Figure 4: Log Probability Plot and Least-Squares Best-Fit Line for RCS-Quartz Exposure Using the Direct Method. PEL by Malaysian USECHH Regulations 2000 is at 0.1 mg m^{-3}

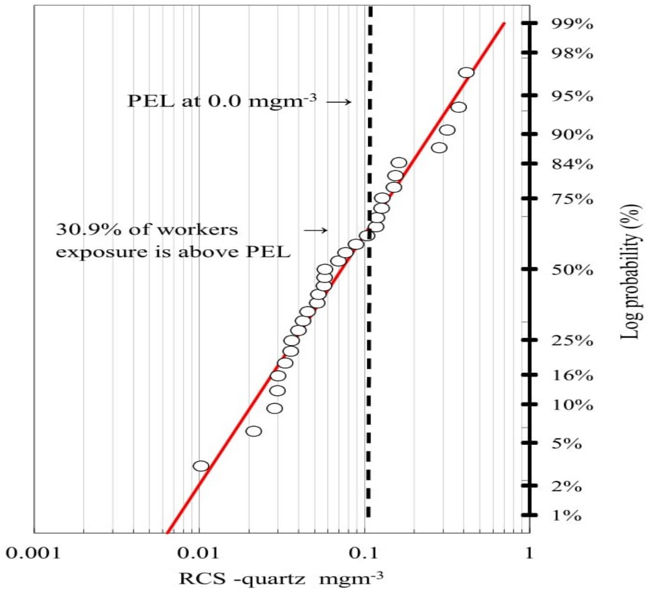


Figure 5: Log Probability Plot and Least-Squares Best-Fit Line for RCS-Quartz Exposure Using the Indirect Method. PEL by Malaysian USECHH Regulations 2000 is at 0.1 mg m^{-3}

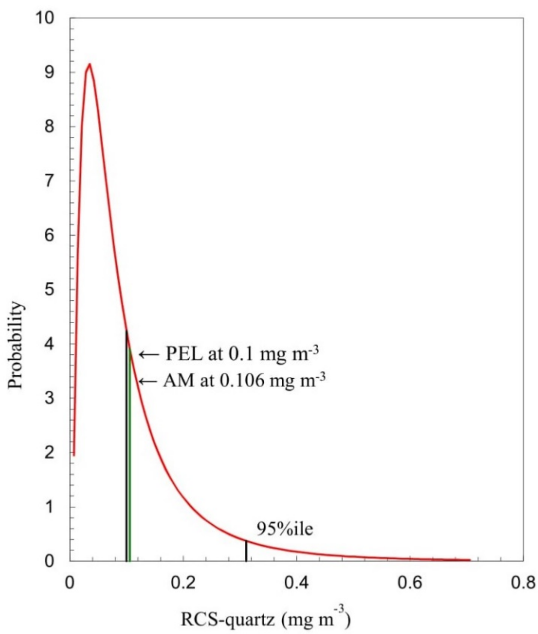


Figure 6: Log-Normal Distribution for RCS-Quartz Exposure using the Direct Method. AM, Arithmetic Mean

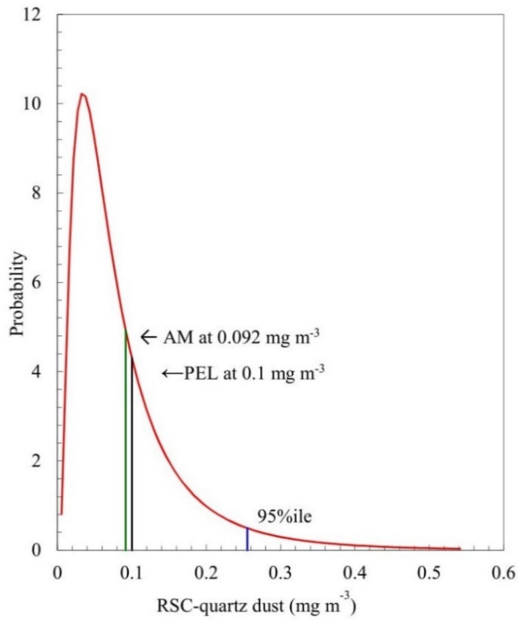


Figure 7: Log-Normal Distribution for RCS-Quartz Exposure using the Indirect Method. AM, Arithmetic Mean

3.4 Uncertainty Associated with Sampling and Analysis for Direct and Indirect Methods

Exposure-based 8-hour TWA was used to determine compliance with regulated or referred occupational limits. Statistical decision-making involves the concept of confidence intervals. When the average concentration is calculated, the exposure is unlikely to be the same as the true average concentration. The discrepancy between the calculated and true concentrations was due to sampling and analysis errors, thereby contributing to uncertainty in the final result. Based on the NIOSH sampling strategy, the accuracy of airborne concentration measurements considers four main causes of airborne concentration measurement errors: 1) random variations in the sampling device, 2) random variations in the analytical procedure, 3) systematic errors in the sampling method, and 4) systematic errors in the analytical procedure (NIOSH, 1977).

Based on this error, we calculated the uncertainty factor in the form of the coefficient of variance (CV) of the final exposure result. CV can be divided into the coefficient of variance for sampling (CV_P) and the coefficient of variance for analysis (CV_A). The CV_P arises from random variations in sampling devices and systematic errors in the sampling method (NIOSH 1977), whileCV_A originates from random and systematic analytical procedures errors (NIOSH, 1977). The total coefficient of variance (CV_T) is derived from the values of CV_A and CV_P using the following formula:

$CV_T= [(CV_P)^2+(CV_A{}^2)]^{½}$

Where:
CV_P: coefficient of variance for sampling
CV_A: coefficient of variance for analytical
CV_T: total coefficient of variance

Table 4 lists the values of each uncertainty factor used in this study. The uncertainty calculations were based on the standard deviation (SD) of each factor. To standardise the unit of calculation, the SDs of each factor were converted to relative standard deviation (RSD) as percentage values. A lower RSD indicates less uncertainty or variability in the data.

Table 4 Uncertainty Values for CV_P, CV_A, and CV_T of the Final Results

	Uncertainty Factor	Unit	RSD for Direct Method (%)	RSD for Indirect Method (%)
Sampling Variation	Flow rate	>C	2.11	0.95
	CV for Sampling (CV_P)		0.02	0.01
Analytical Variation	Reproducibility	%	5.44	3.92
	Recovery	%	8.16	7.62
	Combined uncertainty	%	9.81	8.57
	CV for analytical (CV_A)		0.10	0.09
	Total CV(CV_T)		0.10	0.09

CV_P, coefficient of variance for sampling; CV_A, coefficient of variance for analysis; CV_T, total coefficient of variance

The CV_P for the direct method is 0.02 and 0.01 for the indirect method. The RSD of the sampling flow rate was slightly lower for the indirect method than that for the direct method because of the application of a different set of pumps. The sampling pumps used for both methods were of the same specifications and were interchangeable, but the data showed that the set of pumps used for the indirect method had fewer fluctuations and better performance than those used for the direct method. However, this factor was directly incorporated into the final uncertainty estimation.

The CV_A is 0.10 for the direct method and 0.09 for the indirect method, showing less uncertainty for the direct method compared to the direct method. The RSD for reproducibility was 5.44% for the direct method and 3.92% for the indirect method. The RSD of recovery was 8.16% for the direct method and 7.69% for the indirect method.

Overall, the total coefficient of variance (CV_T) was 0.10 for the direct method and 0.09 for the indirect method. In both methods, the CV_T was more influenced by CV_A than CV_P. The CV_T values indicated that the indirect method had less variance and better quality assurance than the direct method. This final CV_T can be incorporated into each exposure value to assess compliance status.

3.5 Effect of Uncertainty Estimation on Final Compliance Determination.

Using the CV_T values, statistical methods were used for the calculation of interval limits for estimated TWA concentrations at a 95% confidence level. The range of the average concentrations was determined using this procedure. A numerically larger limit is defined as the upper confidence limit (UCL), and a numerically smaller limit is defined as the lower confidence limit (LCL). The LCL and UCL were calculated as follows:

$$LCL\ (95\%) = Y - 1.645\ (CV_T)$$
$$UCL\ (95\%) = Y + 1.645\ (CV_T)$$

where Y is the exposure or TWA concentration divided by the occupational limit (0.1 mg m^{-3}). This formula is based on a 95% confidence interval and full-period single-sample sampling technique.

Table 5 compares RCS-quartz personal exposure using direct and indirect methods before and after considering the variances. The LCL is only calculated if results are slightly higher and UCL if results are slightly lower than the PEL value. The CV_T values are generally small and not significant enough to be incorporated into the final result if the exposure is relatively low or high compared to the occupational limit. However, in this study, UCL was calculated for each personal exposure datum. However, LCL was not reported because in the prevention approach, prediction must be based on the worst-case scenario. Table 5 shows a comparison of RCS-quartz personal exposure using direct and indirect methods after considering uncertainty factors. Adding CV_T values to the UCL calculation increased the AM, GM, and percentages above the PEL. However, the GSD for each method was reduced, indicating that each dataset had less variance and was more reliable for inferential discussions.

Table 5 Summary of RCS-Quartz Personal Exposure Using the Direct and Indirect Methods Before and After Consideration of Total Coefficient of Variance (CV_T)

	N	Direct Method				Indirect Method			
		AM	GM	GSD	% above PEL	AM	GM	GSD	% above PEL
Exposure results based on 8h TWA (before consideration of CV_{T})	31	0.106	0.073	2.42	35.9	0.092	0.0667	2.26	30.9
Exposure results based on UCL (after consideration of CV_{T})	31	0.121	0.093	2.1	46.1	0.103	0.085	1.90	51.8

RCS-quartz, Respirable Crystalline Silica–quartz; AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; PEL, permissible exposure limit [a]; 8-hour TWA based PEL by Malaysian USECHH 2000 at 0.1 mg m^{-3}

When interpretating each personal exposure result independently, both methods showed an increase in non-compliance cases. The direct method exposure results, based on the UCL, showed 13 non-compliance results compared to only 12 previously. Meanwhile, the indirect method showed 14 non-compliance results based on the UCL, compared to only 11 previously. These changes from compliance to noncompliance indicate possible overexposure due to uncertainty or errors during sampling and analysis. In such cases, management may struggle to make final decisions, as UCL results are not conclusive, while the results based on 8-hour TWA are questionable due to uncertainty. Therefore, it is advisable for management to consider resampling to verify actual conditions.

3.6 Variability Due to Environmental Factors

Table 6 shows the RSD values for environmental factors during sampling. Wind velocity, with an RSD reaching up to 83.3%, was the main factor affecting the uncertainty value. In this study, sampling was performed around the crusher plants, and all crusher plants were erected in open areas where the wind direction and speed are unpredictable and easily influenced by weather conditions. However, if sampling is performed under indoor conditions, these factors are minimised. Because the quarries are located in an equatorial region, humidity also plays a role in uncertainty, with an RSD of 13.9%. Temperature had the least contribution (RSD = 0.93%). The temperature differences between sampling and calibration were corrected for each sample volume calculation.

Table 6 Relative Standard Deviation for Environmental Factors

Uncertainty Factor	Unit	RSD for Direct Method (%)	RSD for Indirect Method (%)
Temperature	°C	0.93	0.93
Humidity	L min^{-1}	13.9	13.9
Wind Velocity	M s^{-1}	83.3	83.3

RSD, relative standard deviation,

In this study, variability in environmental factors was not included in the uncertainty calculations. These factors are considered random errors and should not be included in the CV_P calculation. These environmental factors may be minimised through the use of good sampling strategies.

4.0 CONCLUSION

Despite differences in the field sampling results, both the direct and indirect methods showed substantial correlation ($p<0.05$, $r^2 = 0.82$) and no significant differences ($p>0.5$) between the two sets of field data. Arithmetic mean (AM) exposure levels for RCS-quartz were reported to be 0.106 m gm^{-3} and 0.092 m gm^{-3} for the direct and indirect methods, respectively. Log probability plots indicated that 35.9% of crusher operators were exposed to RCS-quartz levels exceeding the exposure limit using the direct method, compared to 30.9% of crusher operators using the indirect method.

The total coefficient of variance (CV_T) was 0.10 for the direct method and 0.09 for the indirect method, with CV_T being more influenced by CV_A than by CV_P in both methods. Overall, the CV_T values indicated that the indirect method had less uncertainty and better quality assurance compared to the direct method. This final CV_T can be incorporated into each exposure value to determine compliance status. Herein, integrating the CV_T value into the UCL calculation showed an increasing number of non-compliance results. The direct method exposure results based on the UCL showed 13 non-compliance results compared to only 12 previously. Meanwhile, the indirect method showed 14 non-compliance results based on the UCL compared to only 11 previously.

In addition to instrument accuracy during sampling and analysis, the variability may also be affected by environmental factors. Wind velocity was the main factor contributing to environmental uncertainty, with an RSD of up to 83.3%. Humidity was also a determinant of environmental variation, with an RSD of 13.9%, while temperature contributed the least, with an RSD of only 0.93%. This study showed that there were no significant differences between the direct and indirect methods, providing industrial hygienists and researchers with flexibility to utilize the direct method based on cost-effectiveness, efficiency, and ease of implementation.

ACKNOWLEDGEMENTS

The authors would like to thank the National Institute of Occupational Safety and Health (NIOSH) of Malaysia for providing a research grant (06/NIOSH/03–06/NG 0018), laboratory facilities, and manpower for this study. Special thanks go to Dr. Rose Norman for assistance in proofreading this manuscript.

REFERENCES

ACGIH, 2015. TLVs and BEI. American Conference of Governmental Industrial Hygiene (ACGIH), Cincinnati.

ALS, 2016. American Lung Association. Learn about silicosis. http://www.lung.org/lung-health-and-diseases/lung-disease-lookup/silicosis

Amran, S., Latif , M.T., Leman, A.M.L., Goh , E., Jaafar, S.A., Khan, M.F., Zainal Abidin, A.S., 2016. Evaluation of performance characteristics between direct and indirect sampling method for respirable crystalline silica (RCS) exposure in granite quarry ARPN J. of Eng. and Appl. Sci. 11.

Bullock, W.H. & Ignacio, J.S. 2006. Appendix IX: Glossary. In. A strategy for assessing and managing occupational exposure, Bullock, W. H. & Ignacio, J. S. (ed.). Third Edition. Fairfax: American Industrial Hygiene Association pp. 423-434.

Cox Jr, L.A., Van Orden , D.R., Lee, R.J., Arlauckas, S.M., Kautz, R.A., Warzel, A.L., Bailey , K.F., Ranpuria , A.K., 2015. How reliable are crystalline silica dust concentration measurements? Regul. Toxicol. Pharmacol. 73, 126-136.

Dong, D., Xu, G., Sun, Y., Hu, P., 1995. Lung cancer among workers exposed to silica dust in Chinese refractory plants Scand. J. Work. Environ. Health, 21, 69-72

DOSH, 2000. Occupational Safety and Health (Use and Standard of Exposure Chemical Hazardous to Health) Regulations 2000. Department of Occupational Safety and Health: Kuala Lumpur, Malaysia.

DOSH. 2002. Guidelines on monitoring of airborne contaminant for chemicals hazardous to health: under the occupational safety and health (use and standard of exposure of chemicals hazardous to health) Regulations, Department of Occupational Safety and Health: Kuala Lumpur. 2000 (P.U. (A) 131).

Ellison, S.L.R., Rosslein, M., Williams, A., 2000. Eurachem/Citac Guide; quantifying uncertainty in analytical measurement., in: Eurachem/Citac (Ed.). Eurachem, Citac, United Kindom.

Garcıa, J.M., Cabo, P.M., Pasarın, J.F., Rodrı´guez, J.F., Vilas, E.F., Mene´ndez, M.C., Martı´nez-Camblor, P., 2013. Measurement precision of respirable dust and silica in workplace conditions. STP 1565, Silica and Associated Respirable Mineral Particles. www.astm.org.

HSE, 2005. Methods for the Determination of Hazardous Substances (MDHS) Guidance No.101: respirable crystalline silica in respirable airborne dust. Health and Safety Executive. 101: 1-16. Link: http://www.hse.gov.uk/pubns/mdhs/pdfs/mdhs101.pdf

HSE, 2011. EH40/2005 Workplace exposure limits, in Executive, H.a.S. (Ed.), second ed.

IARC, 1997. Silica, Some Silicates, Coal Dust and Para-Aramid Fibrils: IARC Monographs on the Evaluation of Carcinogenic Risks to Humans. International Agency for Research Cancer (IARC), World Health Organization(WHO), France.

IFA, 2015. GESTIS International Limit Value. Institute fur Arbeitsschutz der. link: http://limitvalue.ifa.dguv.de/.

Kane, F., 1997. The Campaign to End Silicosis. Job Safety Health. pp. 16-19. Link: http:/www.osha.gov

Kaufer, E., Masson, A., Moulut, J.C., Lecaque, T., Protois, J.C., 2005. Comparison of direct (X-ray diffraction and infrared spectophotometry) and indirect (infrared spectrometry) methods for the analysis of α-quartz in airborne dust Ann. Occup. Hyg. 49, 661-671.

Key-Schwartz, R.J., Baron, P.A., Bartley, D.L., Rice, F.L., 2003. Determination of airborne crystalline silica Hazard Review: Health Effect of Occupational Exposure to Respirable Crystalline Silica. pp. 260-280. Link: https://www.cdc.gov/niosh/docs/2002-129/

Kuo, C.T., Uang, S.N., Huang, H.T., 2010. Application of χ-ray diffractometry on analysis crystalline free silica in foundries, Link: https://laws.ilosh.gov.tw/Book/MP_Publish, in: Department of Public Health, C.M.U. (Ed.).

Lin, C.N., Goh, E., Devi, N., 2012. Quarry Directory Malaysia 2012. IQM, Selangor, Malaysia.

Maciejewska, A., 2008. Occupational exposure assessment for crystalline silica dust: Approach in Poland and worldwide. Int. J. Occup. Med. Environ. Health .

Madsen, F.A., Rose, M.C., Cee, R., 1995. Review of quartz analytical methodologies: present and future needs. Appl. Occup. Environ. Hyg. 10, 991-1002.

Markku, L., Reetta, R., Timo, A., Virpi, V., 2012. Comparison of measuring methods for respirable quartz dust, International Occupational Hygiene Association (IOHA) International Scientific Conference. Poster Tracking 128, Kuala Lumpur.

Miller, A., 2014. In pursuit of a direct-on-filter method for measuring silica in filter samples of airborne dust, Colloqium Mechanical Engineering.

NIOSH, 1977. Occupational exposure sampling strategy manual, Cincinnati, Ohio.

NIOSH, 2003a. Manual Analytical Method No 7602: Silica, Crystalline by Infrared (KBr pellet). . National Institute on Occupational Safety and Health. Link: http://www.cdc.gov/niosh/docs/2003-154/pdfs/7602.pdf.

NIOSH, 2003b. National Institute on Occupational Safety and Health. Manual Analytical Method No 7500: Silica, Crystalline, by X-ray diffractometer (XRD) via filter deposition. National Institute on Occupational Safety and Health. link: http://www.cdc.gov/niosh/docs/2003-154/pdfs/7500.pdf.

Page, S.J., 2006. Crystalline silica analysis: a comparison of calibration material and recent coal mine dust size distribution. Journal of ASTM International, 3.

Rocha-Parise, M., Santos L. M. B., Damoiseaux, J.G.M.C., Bagatin, E., Lido, A.V., Torello, C.O., Tervaert, J.W.C., Queiroz, M.L.S., 2014. Lymphocyte activation in silica-exposed workers. Int. J. Hyg. Environ. Health 217, 586–591.

SKC, 2014. GS-3 respirable dust cyclone, listed in OSHA proposed silica rule. In: SKC Inc. Dorset, U. (Ed.).

Smith, D.K., 1992. Issue and controversy: the measurement of crystalline silica, International Symposium. Department of Geosciences and Material Research University, Pennsylvania State University. pp. 1-70.

Verpaele, S., Jouret, J., 2013. A comparison of the performance of a sampler for respirable dust in workplaces and laboratory analysis for respirable quartz. Ann Occupational Hygiene 57, 54-62.

WHSQ, 2011. Workplace Health and Safety Queensland. Silica-identifying and managing crystalline silica dust exposure. PN 10121 version 2. Department of Justice and Attorney-General. Queensland. Australia.

REFERENCES

ACGIH, 2015. TLVs and BEI. American Conference of Governmental Industrial Hygiene (ACGIH), Cincinnati.

ALS, 2016. American Lung Association. Learn About Silicosis. Link: http://www.lung.org/lung-health-and-diseases/lung-disease-lookup/silicosis

Amran , S., Latif , M.T., Leman, A.M.L., Goh , E., Jaafar , S.A., Khan , M.F., Zainal Abidin , A.S., 2016. Evaluation of performance characteristics between direct and indirect sampling method for respirable crystalline silica (RCS) exposure in granite quarry ARPN J. of Eng. and Appl. Sci. 11.

Bullock, W. H. & Ignacio, J. S. 2006. Appendix IX: Glossary. in. Bullock, W. H. & Ignacio, J. S. (ed.). Third Edition. A Strategy For Assessing and Managing Occupational Exposure, Third Edition. pp. 423-434. Fairfax: American Industrial Hygiene Association

Cox Jr , L.A., Van Orden , D. R., L., R. J., , Arlauckas , S.M., Kautz , R.A., Warzel , A.L., Bailey , K.F., Ranpuria , A.K., 2015. How reliable are crystalline silica dust concentration measurements? Regul. Toxicol. Pharmacol. 73, 126-136.

Dong, D., Xu, G., Sun, Y., Hu, P., 1995. Lung cancer among workers exposed to silica dust in Chinese refractory plants Scand. J. Work. Environ. Health Vol. 21, 69-72

DOSH, 2000. Occupational Safety and Health (Use and Standard of Exposure Chemical Hazardous to Health) Regulations 2000. Department of Occupational Safety and Health, Kuala Lumpur, Malaysia.

DOSH. 2002. Guidelines On Monitoring of Airborne Contaminant For Chemicals Hazardous to Health: Under the Occupational Safety and Health (Use And Standard of Exposure of Chemicals Hazardous to Health) Regulations 2000 (P.U. (A) 131). Kuala Lumpur. Department of Occupational Safety and Health

Ellison, S.L.R., Rosslein, M., Williams, A., 2000. Eurachem/Citac Guide; quantifying uncertainty in analytical measurement., in: Eurachem/Citac (Ed.). Eurachem, Citac, United Kindom.

Garcıa, J.M., Cabo, P.M., Pasarın, J.F., Rodrı´guez, J.F., Vilas, E.F., Mene´ndez, M.C., Martı´nez-Camblor, P., 2013. Measurement precision of respirable dust and silica in workplace conditions. STP 1565, Silica and Associated Respirable Mineral Particles; link: www.astm.org.

Garcia, J.M., Cabo, P.M., Pasarin , J.F., Rodriguez , P.F., Vilas , E.F., Menendez M. C. , Martinez-Camblor, P., 2013. Measurement precision of respirable dust and silica in workplace conditions. Silica and Associated Respirable Mineral Particles; link: www.astm.org.

HSE, 2005. Methods for the Determination of Hazardous Substances (MDHS) Guidance No.101: Respirable crystalline silica in Respirable Airborne Dust, 2005 ed. Health and Safety Executive. Link: http://www.hse.gov.uk/pubns/mdhs/pdfs/mdhs101.pdf, pp. 1-16.

HSE, 2011. EH40/2005 Workplace exposure limits, in Executive, H.a.S. (Ed.), second ed.

IARC, 1997. Silica, Some Silicates, Coal Dust and Para-Aramid Fibrils: IARC Monographs on the Evaluation of Carcinogenic Risks to Humans. International Agency for Research Cancer (IARC), World Health Organization(WHO), France.

IFA, 2015. GESTIS International Limit Value. Institute fur Arbeitsschutz der. link: http://limitvalue.ifa.dguv.de/.

Kane, F., 1997. The Campaign to End Silicosis. Job Safety Health. Link: http:/www.osha.gov, pp. 16-19.

Kaufer, E., Masson, A., Moulut, J.C., Lecaque, T., Protois, J.C., 2005. Comparison of direct (X-ray diffraction and infrared spectophotometry) and indirect (infrared spectrometry) methods for the analysis of α-quartz in airborne dust Ann. Occup. Hyg. 49, 661-671.

Key-Schwartz, R.J., Baron, P.A., Bartley, D.L., Rice, F.L., 2003. Determination of airborne crystalline silica Hazard Review: Health Effect of Occupational Exposure to Respirable Crystalline Silica, Link: https://www.cdc.gov/niosh/docs/2002-129/, pp. 260-280.

Kuo, C.T., Uang, S.N., Huang, H.T., 2010. Application of χ-ray diffractometry on analysis crystalline free silica in foundries, Link: https://laws.ilosh.gov.tw/Book/MP_Publish, in: Department of Public Health, C.M.U. (Ed.).

Lin, C.N., Goh, E., Devi, N., 2012. Quarry Directory Malaysia 2012. IQM, Selangor, Malaysia.

Maciejewska, A., 2008. Occupational exposure assessment for crystalline silica dust: Approach in Poland and worldwide. Int. J. Occup. Med. Environ. Health 21.

Madsen, F.A., Rose, M.C., Cee, R., 1995. Review of quartz analytical methodologies: present and future needs. Appl. Occup. Environ. Hyg. 10, 991-1002.

Markku, L., Reetta, R., Timo, A., Virpi, V., 2012. Comparison of measuring methods for respirable quartz dust, International Occupational Hygiene Association (IOHA) International Scientific Conference. poster tracking #128, Kuala Lumpur.

Miller, A., 2014. In pursuit of a direct-on-filter method for measuring silica in filter samples of airborne dust, Colloqium Mechanical Engineering.

NIOSH, 1977. Occupational exposure sampling strategy manual, Cincinnati, Ohio.

NIOSH, 2003a. Manual Analytical Method No 7602: Silica, Crystalline by Infrared (KBr pellet). . National Institute on Occupational Safety and Health. Link: http://www.cdc.gov/niosh/docs/2003-154/pdfs/7602.pdf.

NIOSH, 2003b. National Institute on Occupational Safety and Health. Manual Analytical Method No 7500: Silica, Crystalline, by X-ray diffractometer (XRD) via filter deposition. National Institute on Occupational Safety and Health. link: http://www.cdc.gov/niosh/docs/2003-154/pdfs/7500.pdf.

Page, S.J., 2006. Crystalline silica analysis: a comparison of calibration material and recent coal mine dust size distribution. Journal of ASTM International 3.

Rocha-Parise, M., Santos L. M. B., Damoiseaux, J.G.M.C., Bagatin, E., Lido, A.V., Torello, C.O., Tervaert, J.W.C., Queiroz, M.L.S., 2014. Lymphocyte activation in silica-exposed workers. Int. J. Hyg. Environ. Health Volume 217, 586–591.

SKC, 2014. GS-3 respirable dust cyclone, listed in OSHA proposed silica rule., in: SKC Inc. Dorset, U. (Ed.).

Smith, D.K., 1992. Issue and controversy: the measurement of crystalline Silica, International Symposium. Department of Geosciences and Material Research University, Pennsylvania State University., pp. 1-70.

Verpaele, S., Jouret, J., 2013. A comparison of the performance of a sampler for respirable dust in workplaces and laboratory analysis for respirable quartz. Ann Occupational Hygiene 57, 54-62.

WHSQ, 2011. Workplace Health and Safety Queensland. Silica-Identifying and managing crystalline silica dust exposure. PN 10121 Version 2. Department of Justice and Attorney-General. Queensland, Australia.