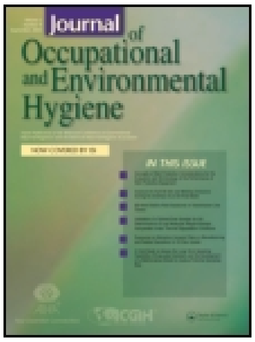

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Variability in Quartz Exposure in the Construction Industry: Implications for Assessing Exposure-Response Relations

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The aims of this study were to determine implications of inter- and intraindividual variation in exposure to respirable (quartz) dust and of heterogeneity in dust characteristics for epidemiologic research in construction workers. Full-shift personal measurements ($n = 67$) from 34 construction workers were collected. The between-worker and day-to-day variances of quartz and respirable dust exposure were estimated using mixed models. Heterogeneity in dust characteristics was evaluated by electron microscopic analysis and electron spin resonance. A grouping strategy based on job title resulted in a 2- and 3.5-fold reduction in expected attenuation of a hypothetical exposure-response relation for respirable dust and quartz exposure, respectively, compared to an individual based approach. Material worked on explained most of the between-worker variance in respirable dust and quartz exposure. However, for risk assessment in epidemiology, grouping workers based on the materials they work on is not practical. Microscopic characterization of dust samples showed large quantities of aluminum silicates and large quantities of smaller particles, resulting in a D_{50} between 1 and 2 μm. For risk analysis, job title can be used to create exposure groups, although error is introduced by the heterogeneity of dust produced by different construction workers activities and by the nonuniformity of exposure groups. A grouping scheme based on materials worked on would be superior, for both exposure and risk assessment, but is not practical when assessing past exposure. In dust from construction sites, factors are present that are capable of influencing the toxicological potency.

Keywords construction workers, dust characteristics, pneumoconiosis, respirable dust, quartz

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Although quartz is an abundant mineral in Earth's crust, human airborne exposure to it is mainly confined to mining operations, the production and use of silica flour, the ceramics industry, foundries, and the construction industry. In the construction industry, quartz exposure can occur during processing of materials containing sand and stone (e.g., brick, concrete, granite, mortar, tile and concrete blocks).

Quartz is a known cause of both obstructive chronic bronchitis(1) (e.g., chronic obstructive pulmonary disease [COPD],) and restrictive respiratory diseases (e.g., pneumoconiosis or, more specifically, silicosis)(2) Lung function loss is described in relation with quartz dust exposure(3) and silicosis.(3–5) Moreover, α-quartz, a crystalline form of quartz, has been classified as carcinogenic to humans (IARC group 1).(6) Although the construction trade is often mentioned on death certificates of individuals with silicosis,(7,8) apart from a recent study among Dutch construction workers,(9) quartz-related respiratory health effects have not been studied in this trade. Silicosis is associated with cumulative exposure to α-quartz, but variability(10,11) and heterogeneity(12,13) in exposure are known to complicate the design of epidemiologic studies. Interindividual variability within groups, considered uniformly exposed, may result in attenuation of exposure-response relationships.(14) Heterogeneity in toxicological properties of the dust(13) may also influence the applicability of exposure proxies. It is also known that surface characteristics and small contaminations of standard quartz particles modify its toxic potency.(15,16)

In construction work, quartz-containing materials do not have a homogeneous composition and a variety of tools generate dust with different characteristics, resulting in differences in biological activity.(12)

Exposure measurements were performed for the assessment of exposure-response relations in an epidemiologic study on respiratory health effects among quartz-exposed construction workers.(9) The aim of this study was to evaluate variability and heterogeneity in exposure to respirable dust and respirable quartz, which potentially affect exposure-response relations.

MATERIAL AND METHODS

Exposure measurements were collected from construction workers subgroups that were included in an epidemiologic study on respiratory health effects.(9) Workers performed the following tasks: concrete drilling, cutting recesses, cleaning of construction sites (sweeping), pointing, grinding mortar between bricks, inner wall construction, and demolition. Three workers (two carpenters, and one worker gluing concrete blocks), who experienced exposure generated by work of colleagues, were included as well. Concrete drilling workers were involved in drilling concrete with jackhammers and hammer drill; they also performed recess milling and sawing in either concrete or lime sandstone. Pointers were involved in filling joints with mortar or grinding mortar with hand-held grinders or jackhammers. Cleaners cleared or swept the work sites. The inner wall bricklayers used concrete blocks (a highly cellular material composed of quartzite, lime, and water). Demolition workers used jackhammers, drills, and excavators equipped with breakers for demolishing. Other tasks included welding, sawing, and clearing of rubble.

Personal respirable dust samples were collected from 34 construction workers on 1 to 3 different days in November and December 1999. Personal air sampling for respirable dust was conducted during full workdays (average duration of 6½ hours), using Dewell-Higgins cyclones obtained from the Casella Group Ltd. (Bedford, UK), connected to Gilian Gilair5 portable pumps (Sensidyne Inc., Clearwater, Fla.) at a flow rate of 1.9 L/min. Of a total of 88 samples, 21 were not valid, due to irregularities or pump failure during measurements, leaving 67 samples for analysis. Five field blanks and 12 duplicate samples were included. After gravimetric determination of dust on the PVC filters (diameter 25 mm; pore size 0.2 μm), quartz was determined by infrared absorption spectrophotometry (IR) according to National Institute for Occupational Safety and Health (NIOSH) method 7602.(17)

The limit of detection (LOD) was assessed as the average weight of the blank filters plus three times the standard deviation, and was 0.15 mg for respirable dust on the filters. Dust samples with dust levels below 0.15 mg ($n = 5$), were assigned a value of two-thirds of this limit divided by the average sampling volume (0.72 m^3), which resulted in a limit of detection for dust measurements of 0.14 mg/m^3. The analytical limit of detection for α-quartz on the filters was 1.7 μg.

Based on four parallel stationary samples, the coefficient of variation (CV) was estimated to be 13% for respirable dust and 7% for respirable quartz. The percentage of quartz was calculated from the mass of quartz and the total amount of gravimetrically determined respirable dust on the filter. Samples with quartz levels below 1.7 μg ($n = 4$), were set at two-thirds of this value divided by the average sampling volume, which resulted in a limit of detection for quartz measurements of 1.6 $\mu g/m^3$.

For a more detailed characterization of dust, six respirable dust samples, acquired during demolition, cutting recesses, grinding in both lime and cement based mortar, clearing rubble and pile top crushing, were studied with Scanning Electron Microscopy (SEM, Philips 515; Philips, Eindhoven, The Netherlands) and transmission electron microscopy (TEM, Philips CM12). Stationary sampling was performed on Nucleopore filters (polycarbonate) with a diameter of 25 mm and pore size of 0.2 μm, coated with carbon before sampling, in Dewell-Higgins cyclones, coupled with a vacuum pump (Becker Equipment Inc., Wuppertal, Germany) connected to a gas meter at a flow of 2 L/min. Before analysis, samples were coated with a thin layer of gold. Particle count, particle diameters and composition of the samples were estimated by TEM. The scanning electron microscope was used for analysis of morphology and was equipped with an elemental dispersive X-ray analyzer (EDAX; Philips) for chemical analysis. Hydroxyl radical generation in samples from drilling in concrete and pile top crushing was measured by electron spin resonance (ESR) spectroscopy.(18) Measurement of soluble transition metals was done in aqueous suspension with 0.08N HNO_3 using inductively coupled plasma mass spectroscopy (IOP-MS)(19) after filtration through 0.2 μm filters.

Statistical Analysis

Distributions of dust and quartz exposure were examined to ascertain logarithmic distribution. Exposure levels were described for different jobs in terms of arithmetic and geometric means as well as the corresponding geometric standard deviations and ranges. Variance components were estimated using multiple linear mixed models.(20) Job title was introduced as a fixed effect, while the worker identity was introduced as a random effect. The models have the following general form:

$$\mathrm{Y_{ijk}} = \mu + \beta_k + \chi_{i(k)} + \varepsilon_{j(ik)} \qquad \textbf{(1)}$$

In this model, $\mathrm{Y_{ijk}}$ represents the natural logarithm of the exposure concentration measured on the $\mathrm{j^{th}}$ day of the $\mathrm{i^{th}}$ worker in a group k; μ is the true underlying mean of log-transformed exposure averaged over all groups; β_k is the fixed effect of group k; $\chi_{i(k)}$ is the random effect of the $\mathrm{i^{th}}$ worker in group k; and $\varepsilon_{j(ik)}$ is the random within-worker variation on day j for worker i in group k.

Separate models were constructed for three measures of exposure: respirable dust, quartz and percentage of quartz. It is assumed that $\chi_{i(k)}$ and $\varepsilon_{j(ik)}$, which are mutually independent, are normally distributed with zero means and variances ${}_{bw}\sigma^2_{yk}$ and ${}_{ww}\sigma^2_{yik}$, respectively. Measurements on the same worker were assumed to be correlated (compound symmetry covariance structure). Variances are estimated as between-worker (${}_{bw}\hat{\sigma}^2_{yk}$) and within-worker (${}_{ww}\hat{\sigma}^2_{yik}$) variance components. To assess whether a more restrictive model could also describe the data when job was used as grouping scheme, variance components were assumed common for all jobs.(20,21) The effect of pooling the variance components when job-title was used to group exposure data, was examined using the likelihood ratio test.(22) The p-values for these tests were approximated by comparing –2 times the difference in log likelihoods between the models to a χ^2-distribution with 2*(number of jobs-1) degrees of freedom. A reduced model should be considered when the difference in –2 log likelihood between models is smaller than χ^2 statistic for given p-value and degrees of freedom.(22) For other exposure models the effect of pooling was not evaluated, because of the potential of a number of combinations: exposure samples comprised dust generated by more than one material, tool or worksite.

The hypothetical performance of a reduced exposure model was evaluated by percentage of explained between-worker variance. In addition, the attenuation ratio of the exposure-response relations (the hypothetical bias in the exposure-response relation towards zero due to non-differential misclassification) was calculated considering either a group-based strategy(14) or an individual based strategy (model with worker only).(10,11) For calculation of the attenuation ratio, the number of workers in each job-title category (n) and the number of repeated measurements (m) were averaged ($n = 4.9$, $m = 1.97$). Statistical analyses were performed with SAS statistical software (version 6.12, SAS Institute, Inc., Cary, N.C.). Statistical significance was reached at $p < 0.05$.

RESULTS

The mean respirable dust and mean respirable quartz concentrations were 2.4 mg/m^3 and 0.40 mg/m^3, respectively. Hypothesis of log normal distribution of exposure data could not be rejected (Shapiro-Wilk statistic: 0.97; p = 0.1 and 0.98; p = 0.3 respectively). The ranges of exposure are large, in particular for quartz (GSD = 7.0). For respirable dust and percentage of quartz the geometric standard deviations were smaller (3.5 and 3.3, respectively) (Table I). The full-shift average exposure levels for respirable nuisance dust exceeded the Dutch maximum acceptable concentration (MAC) of 5 mg/m^3 in 15% (n = 10) of the measurements. Respirable quartz dust concentrations exceeded the Dutch MAC for respirable quartz (0.075 mg/m^3) in 58% (n = 39) of the measurements.

Comparison of the models with distinct variance components of each job title with the models with pooled between- and within-worker variance components for job titles, showed that these were not statistically different for both dust, quartz and percentage of quartz (−2 log likelihood ratio test, χ^2 statistic = 4.11, df = 12, p = 0.9; χ^2 statistic = 4.03, df = 12, p = 0.9, and χ^2 statistic = 11.3, df = 12, p = 0.5, respectively).

TABLE I. Exposures to Respirable Dust (mg/m^3), Respirable Quartz (mg/m^3), and Percentage of Quartz by Job Title

			Respirable Dust (mg/m^3)		Respirable Quartz (mg/m^3)		Percentage of Quartz in Respirable Dust (%)	
Group	n^A	n^B	AM^C min-max	GM^D (GSD^E)	AM min-max	GM (GSD)	AM min-max	GM (GSD)
Total	34	67	2.4	1.2	0.40	0.091	13	7.6
			0.14–14	(3.5)	0.0016–4.7	(7.0)	0.41–53	(3.3)
Recess milling and concrete drilling workers	8	14	3.7	1.9	1.09	0.42	25	22
			0.3–14	(3.3)	0.036–4.7	(5.0)	5.8–53	(1.8)
Pointers (grinding mortar)	4	10	3.5	2.4	0.56	0.35	16	15
			0.5–8.0	(2.7)	0.09–1.6	(2.8)	7.9–24	(1.4)
Pointers	1	3	0.30	0.27	0.003	0.0023	0.94	0.86
			0.14–0.41	(1.8)	0.0016–0.005	(1.9)	0.45–1.2	(1.8)
Demolition workers	10	21	2.4	1.4	0.25	0.14	14	10
			0.2–9.4	(3.0)	0.038–1.3	(2.7)	2.6–38	(2.5)
Inner wall constructors	2	4	2.0	1.5	0.043	0.04	2.6	2.3
			0.5–4.0	(2.5)	0.016–0.084	(2.0)	1.1–4.2	(1.8)
Construction site cleaners	6	12	1.0	0.58	0.032	0.017	3.8	2.9
			0.14–2.5	(3.2)	0.0016–0.1	(3.6)	0.41–8.4	(2.4)
Background exposed group	3	3	0.20	0.19	0.006	0.004	2.4	1.9
			0.14–0.3	(1.6)	0.0016–0.02	(3.5)	1.1–4.8	(2.2)

[A] Number of measured workers.
[B] Number of measurements.
[C] Arithmetic mean.
[D] Geometric mean.
[E] Geometric standard deviation.

TABLE II. Assessment of Variances and Percentage of Explained Between-Worker Variance of Several Exposure Models

Exposure Models	G[A]	$_{ww}\hat{\sigma}_k^{2B}$	$_{bw}\hat{\sigma}_k^2$	% Explained Between-Worker Variance[C]	Predicted Attenuation (%)
Respirable dust					
Worker only	—	0.86	0.77	—	36
Job title	7	0.81	0.45	41	17
Work site	11	0.84	0.26	66	21
Material	7	0.95	0.087	89	14
Tools	2	0.82	0.50	36	8.7
Material and tools	8	0.84	0.091	88	14
Respirable quartz					
Worker only	—	1.0	3.2	—	14
Job title	7	1.0	0.51	84	3.8
Work site	11	1.0	0.65	80	5.7
Material	7	1.5	0.12	96	5.0
Tools	2	1.0	1.6	51	1.8
Material and tools	8	1.2	0.22	93	4.6

Note: Between- and within-variance components were considered common for each exposure model.

[A] G = number of levels of fixed effects (determinants of exposure) in the model.

[B] $_{ww}\hat{\sigma}_k^2$ and $_{bw}\hat{\sigma}_k^2$: restricted maximum likelihood estimates of the within- and between-worker variance components, respectively.

[C] Relative to model with worker only.

Introducing job title with pooled variance components, compared to the models with worker only, resulted in statistically significant different models (χ^2 statistic = 15.8, df = 8, p = 0.046, and χ^2 statistic = 29.4, df = 8, p = 0.0003 for dust and quartz, respectively).

When comparing respirable dust and quartz exposure models, which considered job title as grouping variable, the model for quartz explained more of the between-worker variance (84%) than the model for respirable dust (41%). For quartz, grouping by job title also resulted in less predicted attenuation of a hypothetical dose-response relation (from 14% to 4%). For respirable dust, the predicted attenuation dropped from 36% for the individual approach to 17% for the grouping approach (Table II).

Different exposure models for respirable dust did not alter the within-worker variability (Table II). The model with tools and materials explained 88% of the between-worker variance, opposed to 41% for the model with job-title only. For respirable quartz, an exposure model based on material or material and tools, resulted in a higher within-worker variance ($_{ww}\hat{\sigma}_k^2 = 1.5$ and 1.2, opposed to 1.0 for the model with worker only). In these two models 96 and 93% of between-worker variance was explained compared to the model with worker only, resulting in low between-worker variances ($_{bw}\hat{\sigma}_k^2 = 0.12$ and 0.22, respectively).

Six stationary respirable dust samples, taken for characterization of dust particles, ranged from 0.1 mg/m^3 when clearing rubble in the open air to 4 mg/m^3 when demolishing with hand held hammers. The size distribution of the samples was almost the same in all samples (Figure 1), and showed that the number of small particles was much larger than would be expected from the characteristics of the respirable dust convention. The mean diameter size (D_{50}) was between 1 and 2 μm.

Morphological analysis (Table III) showed that most samples contained aggregates, some round particles, some soot particles, and fume and that every sample contained fibrous particles. The fibers either consisted of organic matter or of gypsum. X-ray analysis and X-ray mapping showed an abundance of aluminosilicates in all samples and quite a lot of quartz particles in samples from demolition, cutting recesses and grinding mortar (lime) (Table III). The other samples consisted of aluminosilicates in larger amounts than quartz. SEM and TEM images and X-ray analysis of a sample from demolition work, illustrate the morphology and composition of samples (Figure 2). Especially the amount of gypsum, fibers, calcium-rich (Ca), iron-rich (Fe), aluminum-rich (Al) particles differed among samples. Electron spin resonance analysis showed that hydroxyl radical activity was 13 and 22 times larger in samples from drilling in concrete and pile top crushing, compared to activity on blank filter. Concentrations of soluble aluminum were high in both samples (24 and 959 μg/L, respectively).

DISCUSSION

All groups included in the study except the background-exposed group and the pointers placing fresh mortar between bricks were at risk of exposure to levels exceeding the Dutch MAC value for respirable quartz.

A grouping strategy based on job title would be appropriate for epidemiologic risk assessment because it reduced

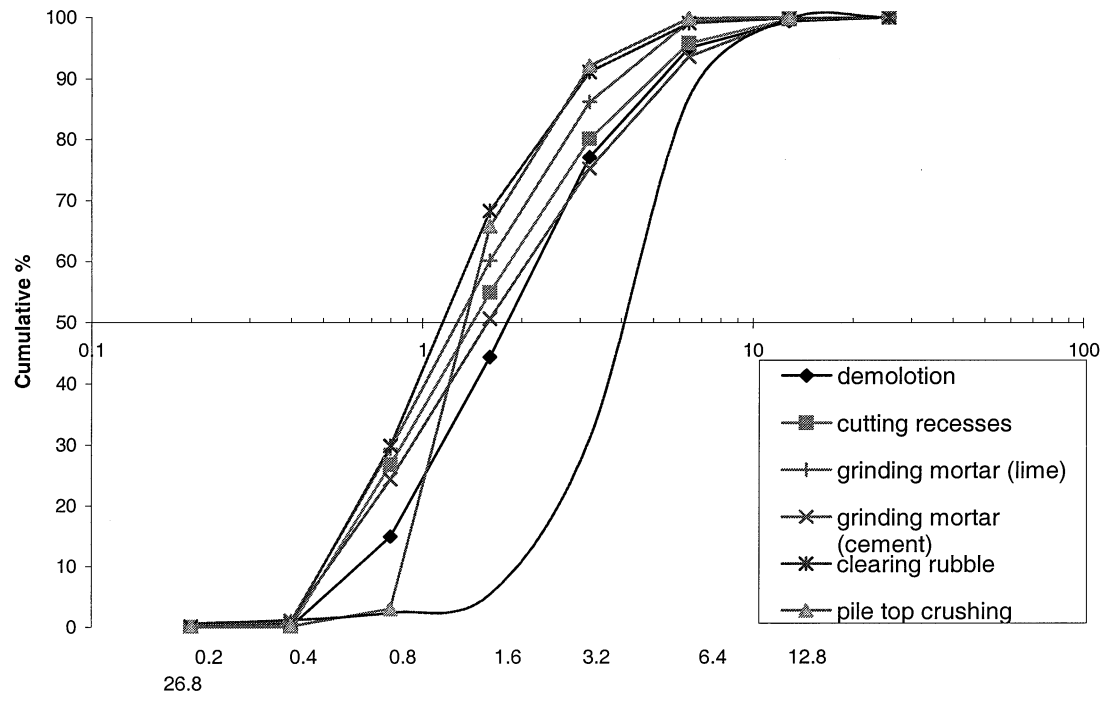

FIGURE 1. Size distribution of respirable dust samples. Data prepared by electron microscopy

expected attenuation relative to the individual based approach. Respirable quartz should be considered a better measure of exposure than respirable dust because of the larger percentage of explained between-worker variance for any of the exposure models presented. The percentage of quartz appeared to be less appropriate as a measure of exposure for risk analysis in epidemiology because there is a lot of unexplained variability and the variability differs between jobs.

An exposure model based on material worked on explained more of the between-worker variance compared to the model with job title, for both respirable dust and quartz exposure. Characteristics of worksites, worker activities, and use of tools have been used successfully to model workers' historic exposure in other settings,(11) but for risk assessment in construction workers it is more efficient and convenient to describe exposures based on job title. It is unlikely that accurate, detailed historic or current information on materials worked on can be obtained from workers, to accurately estimate their exposure. Based on predicted values originating from information on materials used over longer periods of time, assessment of cumulative exposure for construction workers will not be accurate. Construction workers typically work on a variety of materials on different working days, and sometimes even within a working day, as was the case in this study. Worksites differ periodically, especially for specialized workers who work at different locations within one day.

The predicted attenuation of a hypothetical exposure response relation improved satisfactorily for quartz exposure when grouped by job title. The attenuation ratios were calculated using some assumptions, such as an average number of workers per group and an average number of repeated measurements, which are likely to result in some inaccuracy. The coefficients for a number of factors used in the grouping are described elsewhere.(23)

The electron microscopic analysis showed large numbers of particles in the range of the respirable dust convention ($D_{50} = 4.0\ \mu$m). The abundance of small particles implies that a large number of particles can penetrate deep into the lungs potentially causing inflammation. The relatively high concentration of soluble aluminum in the samples might reduce the fibrogenic and acute inflammatory effects of quartz to a certain extent, as has been shown in various animal species.(24,25)

Some samples consisted more of aluminosilicates than quartz. Aluminosilicates are not known to cause silicosis or lung cancer. In some studies of quartz exposure among construction workers, the amount of quartz may have been somewhat overestimated if the interference of aluminosilicates in the X-ray diffraction analysis of quartz has not been accounted for.

The morphology of the dust particles, the presence of impurities, soluble aluminum, and the activity of hydroxyl radicals, show different characteristics in the samples from the different construction workers' activities. Strong indications exist that not only the quartz content, but the extent and properties of the particle surface(15,16,18,26) determine the biological effects. Even among pure workplace quartz samples (quartz content above 90%), enormous differences in biological activity were noted.(18) It is expected that among mixed dust with quartz levels below 20%, effects will even be more influenced by impurities (e.g., metallic iron and aluminum), matrix effects, and mechanical treatment. Several biological assays exist for

TABLE III. Morphology and Qualitative Results of EDX Analysis (SEM) and X-ray Mapping (TEM)

	Demolition	Cutting Recesses	Grinding Mortar (lime)	Grinding Mortar (cement)	Clearing Rubble	Pile Top Crushing
Nonfibrous particles: aggregates	Mostly large; single particles seldom	Lot of aggregates with large sharp edges, combined with fibers	Large and small	Large and few small	Small	More small
Round particles/ fume	−	+/ fume	+	−	+/ fume	+/fume
Soot particles	+++ Combined with other particles	(−) Aggregates and chainlike aggregates	+ Large aggregates and chainlike aggregates	+ Large aggregates and chainlike aggregates	+ Large aggregates and few chainlike aggregates	++ Mostly aggregates and few chainlike aggregates
Fibrous particles	Mostly thick fibers with crystalline structure; some short, thin organic fibers	Fibers with different sizes and crystalline structures	Short, thin fibers (organic)	Short, thin fibers (organic)	Short fibers	Short fibers (organic) and larger fibers
Quartz (>90% Si; 0−2% Al)	++	++	++	+	+	+
Aluminosilicate (>50% Si; >10% Al)	+++	++	+++	++	++	++
Gypsum (>18% S; >47% Ca)	(−)	+++	(−)	(−)	++	(−)
Ca rich (>70%)	+	(−)	++	++	−	−
Fe rich	−	+ (fume and particles)	−	+	+ (fume and particles)	+ (fume and particles)
Al rich	−	−	(−)	−	(−)	−
Fibers	+ Gypsum	(−) gypsum and other fibers	Organic	+ Fibrous silicates	Short	Organic
Others mix	Ca, Si, Al	Ca, Si, Al; Si, Ca	Ca, Si, Fe; Si, Ca, Al, Fe	Cu, Zn, Ca, Si, Al	Si, S, Ca, Al, Cl, Si, S, Cu	Ca, Si, Al

Notes: − = No particles; (−) = Few particles; +, ++, and +++ = Relative amount of particles.

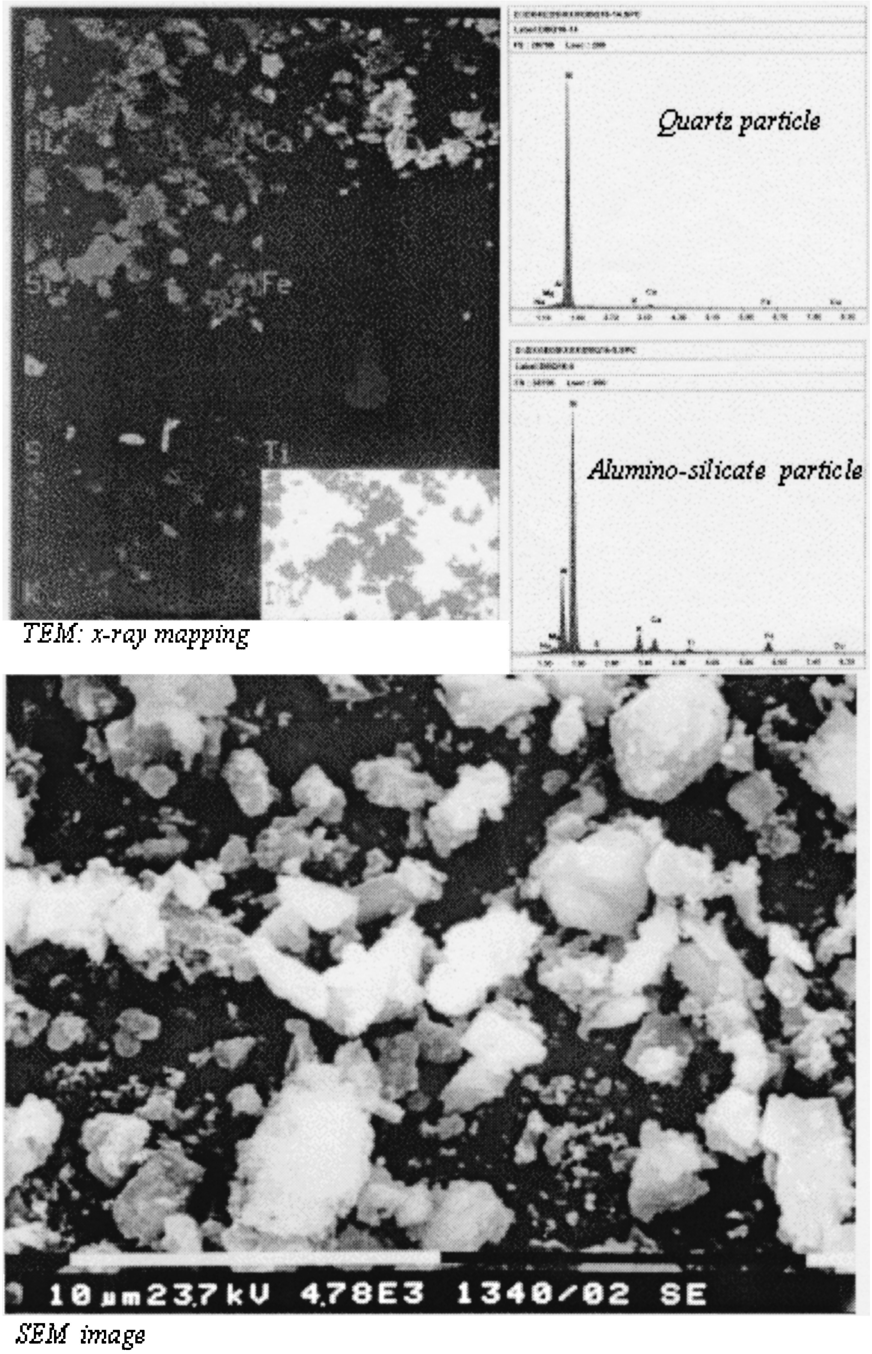

FIGURE 2. SEM and TEM images from dust generated during demolition and element analysis of two of the particles

determining the biological activity of quartz, but the relation between *in vivo* and *in vitro* activity is not clear.(15) It is recommended to use toxicological tests parallel to classic exposure assessment in relation to incidence of biological adverse effects in workers exposed to mixed quartz dusts.

Determining accurate exposure-response relations for quartz-exposed construction workers is likely to be obscured by the heterogeneity in dust characteristics, but to which extend is unknown. When interpreting the results of the our study, one has to keep in mind that the measurements in this study were performed on a limited number of workers and exposed groups performing a wide range of tasks.

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