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Estimating Historical Respirable Crystalline Silica Exposures for Chinese Pottery Workers and Iron/Copper, Tin, and Tungsten Miners

Z. ZHUANG†*, F. J. HEARL†, J. ODENCRANTZ†, W. CHEN‡, B. T. CHEN†, J. Q. CHEN‡, M. A. MCCAWLEY†, P. GAO† and S. C. SODERHOLM†

†*National Institute for Occupational Safety and Health, Centers for Disease Control and Prevention, US Department of Health and Human Services,* 1095 *Willowdale Road, Morgantown, WV* 26505, *USA*; ‡*Department of Labor Health and Occupational Diseases, Tongji Medical University,* 13 *Hangkong Road, Wuhan, Hubei, People's Republic of China*

Collaborative studies of Chinese workers, using over four decades of dust monitoring data, are being conducted by the National Institute for Occupational Safety and Health (NIOSH) and Tongji Medical University in China. The goal of these projects is to establish exposure–response relationships for the development of diseases such as silicosis or lung cancer in cohorts of pottery and mine workers. It is necessary to convert Chinese dust measurements to respirable silica measurements in order to make results from the Chinese data comparable to other results in the literature.

This article describes the development of conversion factors and estimates of historical respirable crystalline silica exposure for Chinese workers. Ambient total dust concentrations (n>17 000) and crystalline silica concentrations (n=347) in bulk dust were first gathered from historical industrial hygiene records. Analysis of the silica content in historical bulk samples revealed no trend from 1950 up to the present. During 1988–1989, side-by-side airborne dust samples (n=143 pairs) were collected using nylon cyclones and traditional Chinese samplers in 20 metal mines and nine pottery factories in China. These data were used to establish conversion factors between respirable crystalline silica concentrations and Chinese total dust concentrations. Based on the analysis of the available evidence, conversion factors derived from the 1988–1989 sampling campaign are assumed to apply to other time periods in this paper. The conversion factors were estimated to be 0.0143 for iron/copper, 0.0355 for pottery factories, 0.0429 for tin mines, and 0.0861 for tungsten mines. Conversion factors for individual facilities within each industry were also calculated. Analysis of variance revealed that mean conversion factors are significantly different among facilities within the iron/copper industry and within the pottery industry. The relative merits of using facility-specific conversion factors, industry-wide conversion factors, or a weighted average of the two are discussed. The exposure matrix of the historical Chinese total dust concentrations was multiplied by these conversion factors to obtain an exposure matrix of historical respirable crystalline silica concentrations. Published by Elsevier Science Ltd on behalf of British Occupational Hygiene Society

Keywords: crystalline silica content; respirable crystalline silica exposure; silicosis; mining dust; exposure estimates

INTRODUCTION

According to a 1988 report from the Chinese Academy of Preventive Medicine, the prevalence of pneumoconiosis in China was over 314 000 in 1986 (Ministry of Public Health, 1989). Over 400 000 additional cases were diagnosed as 'suspect' pneumoconiosis. Pneumoconiosis prevention programs were

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*Author to whom correspondence should be addressed. Tel.: +1-304-285-6167; Fax: +1-304-285-6321; E-mail: zaz3@cdc.gov

developed in response to reports of wide-scale problems in Chinese industries. Since the 1950s, the government of the People's Republic of China has enforced systematic dust sampling regulations. These regulations require mines and companies in the dusty trades to measure the total dust level monthly in dusty work areas wherever silica exposure is expected, to measure the size distribution of total dust particles and crystalline silica content in bulk dust if deemed necessary, and to report the results to higher administrative levels quarterly. Collaborative studies using over four decades of dust monitoring data and silicosis diagnosis records are being conducted by the National Institute for Occupational Safety and Health (NIOSH) in the United States (US) and Tongji Medical University (TMU) in China. The goal of these projects is to establish exposure–response relationships for the development of diseases such as silicosis or lung cancer in cohorts of pottery and mine workers.

Dust monitoring records at 20 metal mines and nine pottery factories in five provinces of south central China were collated and merged into a Chinese total dust exposure matrix based on facility, job title, and calendar year. Approximately 70 000 workers were employed between January 1, 1972 and December 31, 1974 at these mines and factories. The exposure matrix was so constructed that the lifetime dust exposures for workers who ever worked in these facilities can be determined if their work histories are known. This matrix was based on the original job–exposure matrix developed by Dosemeci *et al*. (1993) and used for a nested case-control study of lung cancer among pottery and mine workers exposed to silica-containing dust (Chen *et al*., 1992). The Chinese dust monitoring system is based on a gravimetric method for assessment of airborne concentration, a particle count method for size distribution, and phosphoric acid gravimetry for silica content of settled dust. These dust monitoring strategies are quite different from the sampling strategies used in the United States. The Chinese respirable fraction based on particle count (<5 μm based on physical rather than aerodynamic diameter), using light microscopy, is quantitatively different from the US respirable fraction based on mass. One cannot simply multiply the Chinese total dust concentration by the Chinese respirable fraction to obtain respirable dust concentration in terms relevant to the US respirable crystalline silica dust exposure standards.

US standards were promulgated after studies, conducted by the US Public Health Service in 1929 among granite workers, established a connection between dust exposure and the debilitating dust disease known as silicosis (Russell *et al*., 1929). Further analysis of this cohort in 1941 led to the establishment of a quantitative exposure limit (10 million particles per cubic foot) based on dust concentration and the crystalline silica content of the dust (Russell, 1941). Air sampling during this period was done using impinger samplers, with analysis of dust concentration completed by microscopic particle counting techniques (Greenburg and Bloomfield, 1935; Ayer, 1966). With the recognition that respirable-size particles were most responsible for development of silicosis, respirable mass samplers, such as the cyclone with filter apparatus, were introduced and particle size-selective sampling criteria were defined (Soderholm, 1989; Anon, 1999). In 1970, the American Conference of Governmental Industrial Hygienists (ACGIH) established a respirable mass exposure limit based on the combined exposure to respirable crystalline silica and overall respirable mass. They accomplished this by setting the respirable dust Threshold Limit Value (TLV®) defined by a formula that included the percent respirable silica (%Silica):

$$\text{TLV}^{®} = 10\ \text{mg/m}^3/(\%\text{Silica} + 2)$$

If the dust is 100% crystalline silica, the TLV® is computed at 0.1 mg/m^3, and if the respirable dust contains no silica, the formula produces a TLV® of 5 mg/m^3 (Anon, 1999).

Conversion factors were also developed to convert the earlier particle-count exposure limits to exposure limits based on respirable mass (Sutton and Reno, 1968; Ayer, 1995). In 1986, the ACGIH adopted a revised TLV that required direct measurement of respirable crystalline silica. The revised TLV was set at 0.1 mg/m^3 for α-quartz, and 0.05 mg/m^3 for cristobalite and tridymite (Hearl, 1996). In 2000, the ACGIH further reduced the TLV for α-quartz to 0.05 mg/m^3. The current NIOSH recommended Exposure Limit (REL) is 0.05 for all crystalline forms of silica.

To interpret the Chinese total dust exposure measurements in terms of respirable crystalline silica concentrations as measured in US workplaces, conversion factors are needed. The exposure–response relationship should be established in terms of respirable crystalline silica, so the results of this study can be compared to the results of other studies. It is also necessary to express the exposure variable in terms of respirable crystalline silica concentrations in order to use the results of Chinese-based studies to judge the appropriateness of current respirable crystalline silica standards in the United States.

During 1988–1989, airborne dust samples were collected on the same work shifts using nylon cyclones and the traditional Chinese samplers at selected locations in 20 metal mines and nine pottery factories in China. Both types of samples were analyzed gravimetrically. The cyclone samples were also analyzed for crystalline silica by X-ray diffraction (XRD) (Anon, 1994). Bulk dust samples were also collected at various stations and analyzed for crystalline silica content by phosphoric acid gravimetry (Talvitie, 1951). This survey was conducted to develop valid conversion factors which take into account differences in respirable crystalline silica between settled

dust and airborne respirable dust and between XRD and phosphoric acid gravimetry.

In applying conversion factors derived from the 1988–1989 measurements to other time periods, it would be helpful if historical data on the crystalline silica content and size distribution of airborne particles were available for trend analysis. But they are not. However, data on the crystalline silica content of bulk dust are available for tin mining, tungsten mining, and pottery industries. Therefore, historical percent crystalline silica contents in bulk samples from these sources were collated and analyzed by industry, facility, job title, and time period. These analyses were conducted to help answer questions such as whether job title, facility, industry, and/or time-specific conversion factors should be developed.

This paper describes our proposed conversion factors between Chinese traditional dust samples and current respirable crystalline silica measurements, based on the special 1988–1989 sampling survey. The conversion factor is the ratio of respirable crystalline silica concentrations obtained from each cyclone sample to the paired total dust concentrations measured by the Chinese airborne dust sampler during the special 1988–1989 sampling survey. The conversion factor may be estimated for a given level of specificity, either by industry or by facility within industry. More specific estimates are based on smaller sample sizes and hence are associated with more random error while less specific estimates are associated with more systematic error. We chose a method to determine the best level of specificity for the conversion factor, based on minimizing the estimated mean square error.

In related work, factors to convert Chinese total dust to respirable dust without regard to chemical (crystalline silica) content are described in a separate paper (Gao *et al*., 2000).

MATERIALS AND METHODS

Collation of historical total dust and crystalline silica content data

Chinese industrial hygienists have been measuring and recording workers' environmental exposures to airborne dust and silica since the 1950s. The total dust level was measured in dusty work areas wherever silica exposure was expected. Each facility kept good records of the dust monitoring data, worker employment history, and medical information. All available industrial hygiene records starting in the 1950s were reviewed for total dust concentrations. From 1950 to 1986, concentrations of total dust were averaged over three year intervals for each job title. Exposure levels for total dust were summarized every year from 1986 to 1992. Exposure levels before 1950 were estimated from the monitoring data for 1950. Exposure indices were then created based on facility, job title, and calendar year. Approximately 50% of the facility/job title/calendar year combinations of the job–exposure matrix were estimated based on direct monitoring data. The others were estimated by using monitoring data for similar jobs, or data for the same job at different time periods, with adjustment for other historical exposure information and task description of the job title. To do this, consensus estimates were made by industrial hygiene experts, public health doctors, safety engineers, samplers, and local supervisors, based on the history of control measures, major changes in technical processes in the mines, and comparisons with the other jobs. This matrix is a further development of the job–exposure matrix developed by Dosemeci *et al*. (1993). Dust data from 1990 to 1992 were added.

Although the number of measurements was limited, crystalline silica content was measured for various jobs in most of the facilities as early as 1959 and up to 1993. To determine historical crystalline silica content, bulk (settled) dust samples were collected by brushing the dust into a small polyethylene bottle and analyzed by phosphoric acid gravimetry as will be described later.

Selection of sampling sites

During the special 1988–1989 sampling survey, three sampling sites were chosen in each of the 29 mines or facilities. Each station was sampled for two days. First, each of the three sampling sites was chosen to be representative of a distinct exposure zone. For example, the sites chosen in mining might represent ore extraction, ore transportation, and crushing or bagging. In pottery factories, the three chosen sites were usually allocated to raw material preparations, molding and forming, and glazing final finish. Second, preference was given to sampling sites that were previously sampled under the Chinese national dust monitoring program and that were at locations which were representative of high, medium, and low dust exposures.

Collection and analysis of samples

During the special 1988–1989 sampling survey, the dust levels were measured at each sampling site during the same shift using two instruments: the 10-mm nylon cyclone and the Chinese dust sampler. The cyclone was operated at 1.7 l./min to collect full-shift time-weighted average respirable dust levels (Anon, 1984). Actual work-shift times varied from 2.3 to 7.5 h. The cyclone samples were then analyzed gravimetrically to determine respirable dust concentrations. Respirable crystalline silica mass was determined from the cyclone samples using XRD and percent crystalline silica was calculated. The X-ray diffraction method involves dissolution of the polyvinyl chloride (PVC) filter using tetrohydrofuran, suspension of the silica dust in isopropanol, deposition of the dust on a silver filter, and analysis of the contents of this filter by XRD (Anon, 1994).

Of the 23 cyclone samples taken from iron/copper mines, 20 samples did not have detectable respirable crystalline silica mass on the filters, i.e., the silica mass was less than the limit of detection (LOD=0.015 mg) reported by the laboratory where the samples were analyzed. About 40% of the cyclone samples collected at pottery factories, tin and tungsten mines had silica mass less than the LOD. Hornung and Reed (1990) proposed several techniques for estimation of the average concentration from data containing nondetectable values which cannot be measured but are known to be below some threshold. One of the techniques is that all samples determined to be nondetectable are assigned the value of one-half the LOD. This method was used to estimate the average concentrations of respirable crystalline silica because the percentage of nondetectable values is high and the data are highly skewed.

The Chinese dust sampler (Model FC-2, Wuhan Analytical Instrument Company, Wuhan, China) has an open face 40-mm polytetra-chloroethylene filter (pore size 1.2–1.5 μm, thickness of 0.1 mm) and is battery operated (Ministry of Public Health, 1986). The Chinese samplers collect total dust directly onto the exposed pre-weighed filter. Unlike the cyclones which sample continuously during the work shift, the Chinese sampler operated only while the tasks were in progress. Samplers were typically run during active working periods at a flow rate of 25 l./min, for approximately 15–20 min. After sampling, the filters were brought back to the Tongji Medical University laboratories where they were post-weighed to determine the total airborne dust concentration.

During the special 1988–1989 sampling survey, bulk dust samples were also collected at each sampling site and analyzed by phosphoric acid gravimetry. The percent crystalline silica was estimated using a form of the Talvitie method (Talvitie, 1951), which had also been used for analyzing historical samples. For each percent crystalline silica measurement, a 0.1 g settled dust sample was carefully weighed into a flask, to which was added 15 ml phosphoric acid. Then the dust was stirred at a temperature of 245–250°C for 15 min. The residue was neutralized and washed with distilled water through a residue-free filter. The filter was placed in a tared crucible, dried, ashed, and weighed. The percent of crystalline silica in the sample was the mass gain divided by the initial mass of dust (0.1 g).

Conversion factors

The conversion factor between Chinese traditional dust samples and current respirable crystalline silica measurements was the ratio of respirable crystalline silica concentration (CS) obtained from each NIOSH cyclone sample to the paired total dust concentration (CTD) measured by the Chinese airborne dust sampler during the special 1988–1989 sampling survey.

$$\text{Conversion Factor} = \text{CS}/\text{CTD} \tag{1}$$

The respirable crystalline silica concentration converted from the Chinese total dust concentration was equivalent to a full-shift, time-weighted-average concentration for the actual work-shift duration on that day. The concentration in the historical total dust exposure matrix was multiplied by the conversion factor to obtain the concentrations in the historical respirable crystalline silica exposure matrix. Since most work shifts in these industries in China were shorter than 8 h, these concentrations will have to be adjusted, when 8-h time-weighted-average concentrations are needed for comparison with US standards.

Selection of specificity level by mean square error

We considered the calculation of conversion factors at two levels of specificity, industry-wide factors and facility-specific factors. They are not equally accurate at any given facility. Assuming that there are differences in silica dust composition between the various facilities within an industry, the accuracy for a given facility, resulting from a given level of specificity, will be a combination of the bias resulting from the use of data that are not reflective of the silica dust composition at that facility and random sampling error (which is larger for smaller samples). Generally speaking, it would be expected that a facility-specific conversion factor would have smaller bias but greater variability, due to its smaller sample size, than an industry-wide factor.

The development of facility-specific and industry-wide conversion factors was somewhat similar to the approach used by Seixas *et al*. (1991). Each facility-specific conversion factor was obtained by averaging the ratios of respirable crystalline silica concentration to the Chinese total dust concentration over sampling sites within the facility:

$$\text{CFF}_i = \sum_{j=1}^{n_i} (\text{CS}_{ij}/\text{CTD}_{ij})/n_i, \tag{2}$$

where CFF_i is the conversion factor of the ith facility, CS_{ij} is the respirable crystalline silica concentration of the jth cyclone sample at the ith facility, CTD_{ij} is the Chinese total dust concentration of the jth total dust sample at the ith facility, and n_i is the number of samples taken at the ith facility. Industry-wide conversion factors were obtained by taking the averages of all facility-specific conversion factors within each industry:

$$\text{CFI} = \sum_{i=1}^{N} \text{CFF}_i/N \tag{3}$$

where N is the number of facilities.

We calculated the variances of the facility-specific and industry-wide conversion factors using formulas similar to those that had been used by Seixas *et al*. (1991) for exposures:

$$\mathrm{Var}(\mathrm{CFF}_i) = \sum_{j=1}^{n_i} (\mathrm{CS}_{ij}/\mathrm{CTD}_{ij} - \mathrm{CFF}_i)^2/[n_i(n_i-1)] \quad (4)$$

and

$$\mathrm{Var}(\mathrm{CFI}) = \sum_{i=1}^{N} (\mathrm{Var}(\mathrm{CFF}_i))/N^2 \quad (5)$$

Like Seixas *et al*., we used the mean square error (MSE) of a conversion factor as the measure of its accuracy. However, our use of the MSE, although similar to that of Seixas *et al*., is not exactly the same, as we assign a distinct MSE to each facility, and we include a bias term. Formally, the MSE of an estimate is the average squared distance from each observation to that estimate. If the estimate is simply the average of the observations, the MSE and the variance are the same thing. In the case of the facility-specific conversion factor, $\mathrm{MSE}(\mathrm{CFF}_i)$ is equal to $\mathrm{Var}(\mathrm{CFF}_i)$. In the case of the industry-wide conversion factor, however, the factor is the average of the facility-specific conversion factors, so the MSE is proportional to that facility's weighted-by-facility sum of squares around the industry-wide conversion factor. This is mathematically equivalent to a variance term plus a squared bias term. Although Seixas *et al*. did not consider bias in their definition of mean square error, presumably because between-facility differences may be regarded as systematic rather than random error, we feel that they are part of the inaccuracy inherent to industry-wide conversion factors and should be considered when deciding what estimate is best for a given facility's conversion factor. The industry-wide conversion factor accuracy measure for the ith facility is

$$\mathrm{MSE}(\mathrm{CFI}_i) = \mathrm{Var}(\mathrm{CFI}) + (\mathrm{CFF}_i - \mathrm{CFI})^2 n_i/[(n_i-1)N] \quad (6)$$

The second term is the squared bias term, weighted so that the accuracy measure is proportional to the contribution of the ith facility to the overall mean square error about the CFI.

In addition to conversion factors based on facility-specific or industry-wide samples, intermediate estimates based on some combination of those may also be formed. The simplest of such combinations are linear combinations, in effect weighted averages, where the conversion factor has the following form:

$$\mathrm{CFL}_i = p\mathrm{CFI} + (1-p)\mathrm{CFF}_i \quad (7)$$

where p is a number between 0 and 1; CFI and CFF_i are industry-wide and facility-specific conversion factors. The number p can be chosen to minimize the mean square error by taking the derivative of the mean square error with respect to p and setting it to zero in an approach similar to that of Seixas *et al*. (1991). The MSE of the *i*th linear conversion factor ($\mathrm{MSE}(\mathrm{CFL}_i)$) can be given by the following formula:

$$\mathrm{MSE}(\mathrm{CFL}_i) = p^2\mathrm{MSE}(\mathrm{CFI}_i) + [(1-p)^2 + 2p(1-p)/N]\mathrm{Var}(\mathrm{CFF}_i) \quad (8)$$

This is essentially an application, with the addition of a bias term, of the expression for the variance of a sum of two variables X and Y:

$$\mathrm{var}(X + Y) = \mathrm{var}(X) + \mathrm{var}(Y) + 2\mathrm{cov}(X,Y) \quad (9)$$

where var(X) is the variance of X and cov(X,Y) is the covariance between X and Y. In this expression, X corresponds to the quantity p CFI and Y corresponds to the quantity $(1-p)$ CFF_i. The final term, the covariance, can be obtained by expanding the product of the means and then taking the covariance of each term; this reduces to variance of CFF_i, multiplied by the quantity $2p(1-p)/N$. The resulting optimal value of p, obtained by taking the derivative of the MSE with respect to p and setting the resulting expression to zero, is the following:

$$p = (1-1/N)\mathrm{Var}(\mathrm{CFF}_i)/(\mathrm{MSE}(\mathrm{CFI}_i) + (1-2/N)\mathrm{Var}(\mathrm{CFF}_i)) \quad (10)$$

Statistical analyses

Each industry's percent of crystalline silica in bulk samples was tested for unspecified differences between years, for a historical trend, and for a difference between the years 1988 and 1989 and all earlier years. The models used fixed-effect predictors for the year the data were collected: a class variable for year in the first model, a numerical variable for year in the second model, and an indicator variable for whether or not the data had been collected in 1988–1989 for the third model. Facility (the specific mine or pottery factory at which the measurements were made) and job title by facility were included as random effects. The response variable was percent crystalline silica in bulk dust.

A comparison was made between the Talvitie method applied to bulk dust and X-ray diffraction applied to airborne dust collected in the cyclone. The comparisons were carried out by a paired *t*-test on silica percent estimated by the two measures.

Conversion factors were tested for differences between industries and between facilities within industry. The predictors for this model were class variables for industry and facility. Industry was taken to be a main fixed effect and facility was a random effect nested within industry. For the purposes of this test an individual conversion factor was calculated from each observation. Because the data were markedly right-skewed, they were log-transformed for the analysis. Tukey's test (Montgomery, 1984) was used to determine which means were significantly different, with a 0.05 significance level. The linear mixed models for silica concentration and conversion factor differences were implemented using SAS PROC MIXED (Anon, 1996).

RESULTS

Exposure matrix of historical Chinese total dust

An exposure matrix based on facility, job title, and calendar year was constructed using historical Chinese total dust data from ongoing joint NIOSH–Chinese epidemiological studies to quantitatively assess the exposure–response relationship for the development of silicosis. The matrix has a total of 17 632 historical Chinese total dust estimates and consists of 19 columns (calendar-year periods) and 928 rows (facility–job titles combinations). Some of the cells were estimated based on as many as 36 direct measurements (monthly monitoring data for three-year intervals). The numbers of facility–job title combinations were 172, 124, 96, and 263 for iron/copper mines, pottery factories, tin mines, and tungsten mines, respectively.

Historical crystalline silica data

The historical percent crystalline silica contents in bulk samples were first analyzed by industry and facility and are summarized in Table 1. Only the mean crystalline silica (28.2%) for pottery factory No. 29 was significantly smaller than a number of the other pottery facilities (21, 22, 23, 26, and 27). No statistically significant difference in mean crystalline silica was found among different tin mine facilities. Thus, the variability was relatively small among tin mine and pottery factory facilities. However, the variability of crystalline silica was large among tungsten mine facilities. The difference between the largest (facility 14) and smallest mean percent crystalline silica in tungsten bulk dust (facility 12) was 33%. The mean crystalline silica for facility 12 was significantly lower than that for most of the other tungsten facilities (10, 11, 13, 14, 15, and 17); additionally, the mean crystalline silica content for facility 11 was significantly larger than that of two others (16 and 18).

Table 1. Summary of historical crystalline silica in bulk samples (analysis by phosphoric acid gravimetry) by industry and facility

Industry	Facility no.	*n*	Mean crystalline silica (%)	Standard deviation
Pottery	22	15	43.3	15.6
Pottery	26	8	41.6	11.1
Pottery	27	13	41.5	9.3
Pottery	21	14	40.1	9.5
Pottery	24	5	39.2	12.9
Pottery	23	11	38.5	8.2
Pottery	25	13	34.0	12.9
Pottery	29	55	28.2	11.7
Tin	42	7	38.9	11.7
Tin	43	16	35.8	9.2
Tin	44	11	34.6	27.3
Tin	41	14	31.8	13.5
Tungsten	14	4	61.5	18
Tungsten	11	10	58.9	14.5
Tungsten	13	11	52.9	16.4
Tungsten	15	8	51.4	20.4
Tungsten	17	11	47.9	16.9
Tungsten	10	34	47.4	16.6
Tungsten	19	6	41.3	21.8
Tungsten	16	16	39.0	8.1
Tungsten	18	8	36.4	22.0
Tungsten	12	57	27.8	10.7

Table 2 shows the variations of the historical crystalline silica data in time. In the pottery industry, the largest mean silica content (52.8%) was observed in 1993. The smallest mean percent silica contents were measured in 1986 (21.0%) and 1991 (22.0%). No statistically significant between-year differences up to 1989 for free silica concentration were found by the mixed-effect linear models used for that purpose. The three types of differences tested for were nonspecific differences among the calendar years, linear trends, and differences between the silica concentration in 1988–1989 and in earlier years. Significance levels for nonspecific differences were $P = 0.168$ for tungsten mines, $P = 0.138$ for pottery factories, and $P = 0.344$ for tin mines. Significance levels for linear trends over time were 0.068 for tungsten mines, 0.524 for pottery factories, and 0.067 for tin mines. Significance levels for a difference between 1988–1989 and earlier years were 0.696 for tungsten mines, 0.091 for pottery factories, and 0.052 for tin mines.

Effect of sample type and analytical method on crystalline silica content

Table 3 shows the mean percent crystalline silica in the 1988–1989 bulk dust samples (analyzed by phosphoric acid gravimetry) by industry as compared to the mean percent crystalline silica in the corresponding cyclone samples XRD. Percent crystalline silica in bulk dust samples was significantly larger than the percent crystalline silica in cyclone samples in all industries (Overall: $P<0.0001$, Pottery: $P<0.0001$, Tin: $P = 0.014$, Tungsten: $P = 0.003$) except iron/copper in which there was only one paired sample and no *t*-test was performed. Thus, sample type and analytical method had significant effect on crystalline silica content. Respirable crystalline silica exposure cannot be reasonably estimated by multiplying the respirable fraction of the Chinese total dust by the percent crystalline silica in bulk dust.

Concentrations of respirable crystalline silica dust and Chinese total dust

Table 4 summarizes the respirable crystalline silica dust and Chinese total dust concentrations by each facility and across all facilities in each industry using data from the special 1988–1989 sampling survey. Figure 1 is a scatter plot of respirable crystalline silica concentrations against the paired Chinese total dust concentrations by industry. One data point was excluded from the data analysis because the respirable crystalline silica dust concentration (0.3 mg/m^3) was

Table 2. Summary of historical crystalline silica in bulk samples (analysis by phosphoric acid gravimetry) by industry and year

Year	Pottery			Tin			Tungsten		
	n	Mean crystalline silica (%)	Standard deviation	*n*	Mean crystalline silica (%)	Standard deviation	*n*	Mean crystalline silica (%)	Standard deviation
1959	2	28	0	2	17	0	4	57.8	14
1962	2	41	0	3	45.0	1.7	7	61.6	12.1
1965	0	–	–	4	44	24.2	4	37.3	4.3
1968	0	–	–	3	36.3	33.6	0	–	–
1974	8	26.8	11.9	8	30.8	18	1	40.0	–
1977	18	34.4	8.7	3	35.7	12.5	27	36.0	10.3
1980	3	27.0	11.5	2	35.5	14.8	1	28.0	–
1983	10	34.5	9.1	0	–	–	11	47.7	22.4
1986	1	21	–	2	35.0	1.4	5	23.0	26.1
1987	2	24.5	9.2	0	–	–	3	43.3	28.2
1988	33	35.8	8.9	7	29.3	16.4	32	39.3	15.8
1989	8	28.4	11.7	0	–	–	24	39.1	17.5
1990	9	24.3	9.7	0	–	–	19	38.5	19.8
1991	7	22	9.4	0	–	–	0	–	–
1992	15	40.3	16.7	7	39.1	12.7	16	31.2	12.7
1993	16	52.8	7.9	7	34.7	12.9	11	60.7	13.3
All	134	35.1	13	48	34.8	16.1	165	40.7	17.9

Table 3. Comparison of 1988–1989 percent crystalline silica by sample type and analytical method

Industry	*n*	Bulk samples by phosphoric acid gravimetry		Cyclone samples by X-ray diffractometry		*P*-value[a]
		Mean % crystalline silica	Standard deviation	Mean % crystalline silica	Standard deviation	
Iron/copper	1	12.5	–	15.8	–	–
Pottery	19	37.4	9.1	21.4	11.0	<0.01
Tin	5	29.5	13.4	6.4	3.8	<0.05
Tungsten	17	50.4	18.3	29.2	17.6	<0.01

[a]P-values are calculated for *t*-test of paired samples to see if the mean % crystalline silica in bulk dust by phosphoric acid gravimetry is significantly different from that in respirable dust by X-ray diffractometry.

higher than the Chinese total dust concentration (0.2 mg/m^3). The average respirable crystalline silica concentrations were 0.017, 0.116, 0.097, and 0.101 mg/m^3 for iron/copper mines, pottery factories, tin mines, and tungsten mines, respectively. The mean respirable crystalline silica concentrations were found to exceed the ACGIH TLV and NIOSH REL of 0.05 mg/m^3 in none of the five iron/copper mines, eight of the nine pottery factories, one of the three tin mines, and seven of the ten tungsten mines.

The average Chinese total dust concentrations were 6.1, 5.1, 2.9, and 1.9 mg/m^3 for iron copper mines, pottery factories, tin mines, and tungsten mines, respectively. The Chinese total dust concentrations were found to exceed the Chinese total dust standard of 2 mg/m^3 in three of the four industries. The mean Chinese total dust concentrations were found to exceed the Chinese total dust standard of 2 mg/m^3 in two of the five iron/copper mines, all nine pottery factories, one of the three tin mines, and five of the ten tungsten mines.

Conversion factors

Table 5 compares three approaches to the conversion factor: industry-wide, facility-specific, and the best linear combination of those two. For each method, the estimate and the mean square error of the estimate are given, the latter in order to allow a comparison between the methods. Using the nested mixed model on log-transformed conversion factors, a significant ($P = 0.001$) difference was found among industries. The conversion factors for tungsten were found to be significantly larger than those for iron ($P = 0.001$) and pottery ($P = 0.036$) using Tukey's multiple hypothesis test. Analysis of the log-transformed data showed significant differences among facilities both for iron/copper ($P<0.0001$) and for pottery ($P = 0.033$). The conversion factors within the iron

Table 4. Summary of 1988–1989 respirable crystalline silica dust concentration, and Chinese total dust concentration by each facility and over all facilities in each industry

Industry	Facility no.	*n*	Respirable crystalline silica concentration (mg/m^3)		Chinese total dust concentration (mg/m^3)	
			Mean	Standard deviation	Mean	Standard Deviation
Iron/copper	31	3	0.015	0.002	1.67	0.88
Iron/copper	32	6	0.017	0.002	4.11	2.46
Iron/copper	33	3	0.016	0.0004	1.42	1.16
Iron/copper	35	5	0.014	0.002	20.2	14.9
Iron/copper	36	6	0.021	0.006	0.91	0.59
Iron/copper	All	23	0.017	0.004	6.11	10.1
Pottery	21	6	0.153	0.208	2.78	3.61
Pottery	22	6	0.167	0.298	10.8	20.8
Pottery	23	6	0.119	0.165	3.08	2.13
Pottery	24	6	0.169	0.234	4.58	3.35
Pottery	25	6	0.173	0.392	9.74	15.6
Pottery	26	6	0.056	0.040	2.41	1.86
Pottery	27	12	0.085	0.101	3.11	3.25
Pottery	28	3	0.017	0.006	2.00	1.59
Pottery	29	3	0.051	0.057	11.2	11.1
Pottery	All	54	0.116	0.199	5.14	9.23
Tin	41	1	0.036	.	0.60	.
Tin	43	4	0.199	0.262	4.80	6.08
Tin	44	5	0.028	0.018	1.81	1.91
Tin	All	10	0.097	0.175	2.89	4.10
Tungsten	10	3	0.420	0.278	2.97	1.08
Tungsten	11	6	0.065	0.078	3.02	4.09
Tungsten	12	6	0.246	0.130	3.13	3.33
Tungsten	13	6	0.081	0.086	2.13	2.41
Tungsten	14	6	0.093	0.121	1.54	1.47
Tungsten	15	6	0.046	0.024	2.56	2.75
Tungsten	16	6	0.053	0.059	1.28	1.68
Tungsten	17	5	0.061	0.044	1.05	0.82
Tungsten	18	6	0.049	0.061	0.78	0.74
Tungsten	19	6	0.044	0.026	1.11	0.40
Tungsten	All	56	0.101	0.131	1.92	2.26

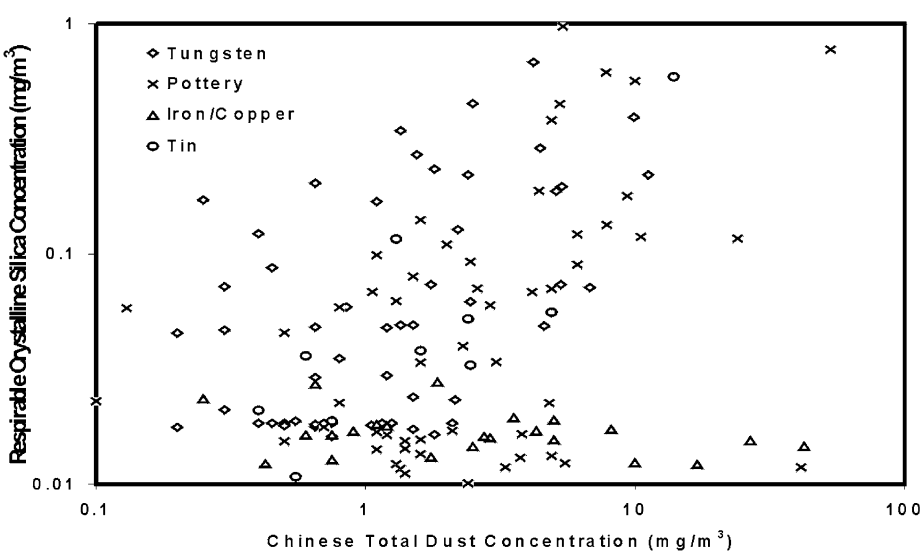

Fig. 1. Scatter plot of respirable crystalline silica concentrations against the paired Chinese total dust concentrations by industry.

Table 5. Summary of conversion factors by each facility: estimate and mean square error (MSE) of the estimate

Industry	Facility no.	*n*	Industry-wide		Facility-specific		Best linear combination	
			Mean	MSE ($\times 10^4$)	Mean	MSE ($\times 10^4$)	Mean	MSE ($\times 10^4$)
Iron/copper	31	3	0.0143	0.485	0.0117	0.257	0.0125	0.191
Iron/copper	32	6	0.0143	0.619	0.0063	0.048	0.0068	0.046
Iron/copper	33	3	0.0143	0.491	0.0173	0.387	0.0160	0.254
Iron/copper	35	5	0.0143	0.895	0.0012	0.003	0.0012	0.003
Iron/copper	36	6	0.0143	1.503	0.0351	1.562	0.0245	0.922
Pottery	21	6	0.0355	7.085	0.0868	9.713	0.0565	4.621
Pottery	22	6	0.0355	3.732	0.0246	1.019	0.0268	0.838
Pottery	23	6	0.0355	3.583	0.0332	1.377	0.0338	1.055
Pottery	24	6	0.0355	3.671	0.0270	1.398	0.0292	1.073
Pottery	25	6	0.0355	3.577	0.0349	8.501	0.0353	2.897
Pottery	26	6	0.0355	3.641	0.0285	1.008	0.0299	0.826
Pottery	27	12	0.0355	4.751	0.0666	12.49	0.0427	3.969
Pottery	28	3	0.0355	4.382	0.0135	0.549	0.0157	0.499
Pottery	29	3	0.0355	5.217	0.0041	0.005	0.0041	0.005
Tin	41	1	0.0429	–	0.0602	–	–	–
Tin	43	4	0.0429	3.162	0.0443	2.468	0.0437	1.986
Tin	44	5	0.0429	5.309	0.0243	0.545	0.0253	0.531
Tungsten	10	3	0.0861	16.12	0.1334	14.51	0.1111	8.359
Tungsten	11	6	0.0861	16.77	0.0283	0.228	0.0290	0.225
Tungsten	12	6	0.0861	13.86	0.1164	13.19	0.1017	7.419
Tungsten	13	6	0.0861	14.85	0.0444	0.294	0.0451	0.289
Tungsten	14	6	0.0861	12.90	0.0753	23.04	0.0824	9.316
Tungsten	15	6	0.0861	16.30	0.0318	0.892	0.0343	0.855
Tungsten	16	6	0.0861	12.77	0.0886	12.37	0.0874	6.902
Tungsten	17	5	0.0861	14.62	0.1247	38.14	0.0953	12.03
Tungsten	18	6	0.0861	19.23	0.1595	115.7	0.0911	18.71
Tungsten	19	6	0.0861	13.66	0.0587	7.937	0.0685	5.387

industry ranged from 0.001 in facility 35 to 0.035 in facility 36; those in the pottery industry ranged from 0.004 for facility 29 to 0.087 for facility 21.

For most of the facilities the use of a facility-specific conversion factor resulted in a smaller mean squared error than the use of an industry-wide conversion factor, but for seven facilities the mean square error was somewhat smaller when the industry-wide conversion factor was used. They were facility 36 of the iron/copper mining industry, facilities 21, 25, and 27 of the pottery industry, and facilities 14, 17, and 18 of the tungsten mining industry. For these seven facilities, the ratio of the facility-specific mean square error to the industry-wide mean square error ranged from 1.04 to 6.01. It was greater than 2 for four facilities: pottery factory number 25 (2.38), pottery factory number 27 (2.63), tungsten mine number 17 (2.61) and tungsten mine number 18 (6.01). Of the seven facilities, copper mine number 36 had the lowest ratio.

Estimates of historical respirable crystalline silica exposure

The historical total dust exposure matrix was multiplied by the best-linear-combination conversion factors to obtain the respirable crystalline silica concentration matrix for all facilities except for those where the MSE could not be estimated. For those facilities (i.e., tin mines 41 and 42) the industry-wide conversion factor was used instead. The average respirable crystalline silica concentration of all facility–job title combinations was calculated for each calendar-year period by industry and is plotted against the ending year of each period (Fig. 2) to show the trend of the respirable crystalline silica concentrations over time.

DISCUSSION

In applying conversion factors, derived from the 1988–1989 measurements to other time periods, it would be helpful if historical data on the crystalline silica content and size distribution of airborne particles were available for trend analysis. But they were not. As a surrogate, available historical data on the crystalline silica content of bulk dust were analyzed and discussed. Possibly due to small sample sizes and large variability in some calendar years, there is no statistical indication of the need for factors based on time-specific percent crystalline silica content. Based on this analysis of the available evidence, conversion factors derived from the 1988–1989 sampling campaign are assumed to apply to other time periods in this paper.

In deriving conversion factors, distribution of particle size is an important factor to be considered. The mechanisms of dust generation producing a wide range of particle sizes may change over time due to new methods of production or installation of venti-

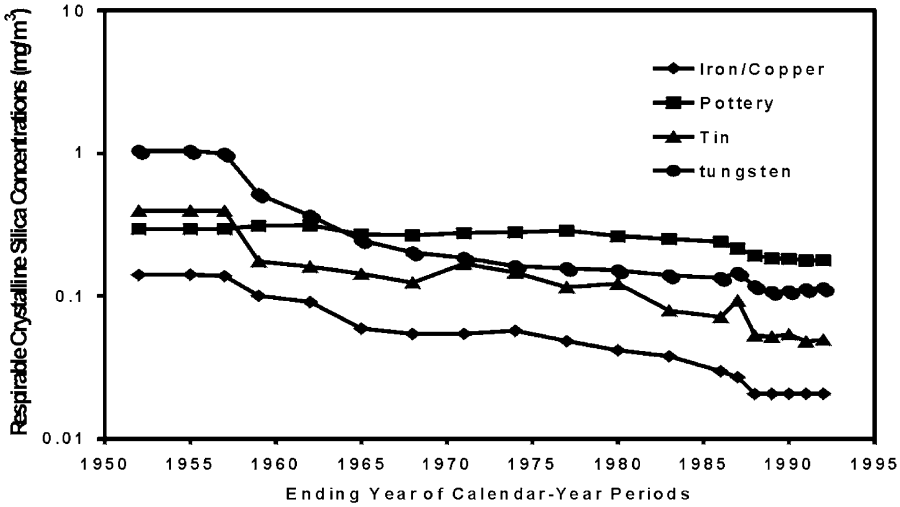

Fig. 2. Trend of average respirable crystalline silica concentrations by industry.

lation controls. The distribution of particle size is also expected to vary among job tasks, facilities, and industries. The conversion factors only took into account the variation among job tasks, facilities, and industries because cyclone (i.e., respirable) samples were collected at three work stations at each facility. There are insufficient historical particle size data to allow an analysis of whether size distributions changed from the 1950s to the 1990s. Those using these conversion factors in epidemiological or other studies should evaluate information about changes in work practices and conditions before deciding whether to apply the conversion factors from the 1988–1989 measurements to other time periods.

It has been reported that the percentage of quartz in bulk or settled dust samples is usually greater than that in the smaller respirable-size dust samples (Ayer, 1969). The results of this study are consistent with Ayer's observation. The tendency for quartz to be lower in percentage in the finer dust can be explained by the fact that quartz is harder than most of the other minerals with which it occurs, and thus, more successfully resists comminution into finer particles.

An important issue that must be considered in developing and using measures such as percent crystalline silica in dust and conversion factors is the most appropriate level of specificity. This paper considers whether the percent crystalline silica for 1988–1989 can reasonably be applied to earlier years, or whether a time-varying percent crystalline silica measure would be preferable. It also considers two levels of specificity, facility-specific and industry-wide, for the conversion factor, plus a linear combination of the two. The level of specificity that is appropriate for percent crystalline silica or a conversion factor depends on a number of factors including the quantity and accuracy of the data, the reasonableness of assuming that applying a broader level of specificity will still result in a useful estimate, and the statistical properties of an epidemiological or other model that makes use of a given percent crystalline silica estimate or estimates or a given conversion factor or set of conversion factors.

Given the limited amounts of percent crystalline silica data available for the years other than 1988–1989, it is not feasible to make use of a year-by-year industry-specific percent crystalline silica content, so the questions considered in this regard were whether the 1988–1989 measurements could reasonably be considered typical for earlier data and whether a linear trend exists for any of the industries. Both of these questions were tested statistically, along with an additional test for unspecified differences between calendar years, for each industry for which such a test could be made. As there were no statistically significant differences, it was concluded that the 1988–1989 percent crystalline silica content could be considered typical for years prior to that.

Industry-wide conversion factors are averages of the facility-specific conversion factors for a given industry. The facility-specific factors may be averaged in a number of ways, one obvious choice being between a weighted or unweighted average. Seixas *et al*. (1991) used an unweighted average, and we have followed them in this. It would, however, have been feasible to weight each facility's conversion factor by the number of observations taken at that facility, which would correspond to an unweighted average over the observations. Averaging over the observations reduces the amount of sampling variability in the industry-wide conversion factor but has the effect of allowing facilities at which more observations were taken to exert a disproportionate influence on what is used as a normative conversion factor for the industry, and a case can be made for either kind of weighting. A weighted-by-observations approach would have resulted in the following industry-wide conversion factors: 0.015 for iron/copper mines,

0.042 for pottery shops, 0.036 for tin mines, and 0.086 for tungsten mines.

This paper proposes the use of the mean square error as one possible criterion for deciding between facility-specific and industry-wide conversion factors. The mean square error for a given facility is the mean squared difference between the individual measurements of the conversion factor at that facility and the final estimate of the conversion factor that is used for that facility. It can be decomposed into a term for the sample variance and a term for the squared bias, if any, that is associated with a particular estimate of the conversion factor. The motivation for its use is that, in choosing between facility-specific versus industry-wide conversion factors, we are in effect choosing between greater random error on the one hand and the addition of bias on the other. Although data from a particular facility may suggest that the facility is atypical for the industry, the data may be of such poor quality as a result of excessive variability and small sample size that that conclusion is suspect. Using the mean square error criterion, a facility-specific conversion factor will be used if it is similar to the conversion factors for other facilities in the same industry or if its difference from the industry norm is well supported by the data, but not if it is both atypical and unreliable. Of the facilities considered in this study, seven have mean square errors that are somewhat larger for facility-specific conversion factors than for industry-wide conversion factors. One of these, facility 18 of the tungsten mining industry, has a mean square error for the industry-wide conversion factor that is only a small fraction of its mean square errors for its facility-specific conversion factors. Additionally, three other facilities, 25 and 27 of the pottery industry and facility 17 of the tungsten mining industry, have industry-wide mean square errors that are less than half of their facility-specific mean square errors, and three others, facility 36 of the iron/copper mining industry, facility 21 of the pottery industry, and facility 14 of the tungsten mining industry, have industry-wide mean square errors that are less than their facility-specific mean square errors. Since their conversion factors are both atypical and unreliable, application of the mean square error criterion would result in using the industry-wide rather than the facility-specific conversion factor. In addition to the facility-specific and industry-wide conversion factors, a linear combination of the two was also proposed. Since the linear combination minimizes the mean square error, a researcher who was choosing conversion factors from these three on the basis of the smallest mean square error would always choose the best linear combination.

That is not to say that the mean square error is the only criterion that can be applied to choosing among different levels of specificity. Application of the mean square error principle results in more reliable conversion factor estimates at individual facilities, at least by one measure of reliability, but reliability may not be a researcher's only concern. Among the possible drawbacks to its use is the fact that statistical models using mixtures or linear combinations of industry-wide and facility-specific conversion factors have more complicated variance structure than statistical models that use either a common conversion factor for all facilities or else facility-specific conversion factors for all facilities. Then, too, some researchers may feel that the introduction of additional bias to the conversion factor estimate is unacceptable regardless of any accompanying reduction in the random sampling error.

The average respirable crystalline silica concentrations for iron/copper mines and pottery factories remained relatively constant over time as seen from Fig. 2. The average respirable crystalline silica concentrations for tungsten mines declined dramatically in the late 1950s. This trend can be attributed to the installation of wet dust suppression and ventilation systems in the mines and pottery factories as required by the Chinese government in the late 1950s.

The relationship between cumulative risk of silicosis among Chinese tin miners and cumulative Chinese total dust exposures are reported in another paper (Chen *et al*., 2001). Using the results reported here, the exposure–response relationship between cumulative risk and respirable crystalline silica exposure estimated with the US monitoring strategies was also established and reported in that paper. The results of that epidemiological study were found to be similar to the results from other studies which established similar relationships (Kreiss and Zhen, 1996; Steenland and Brown, 1995; Hnizdo and Sluis-Cremer, 1993). This finding indirectly validated the conversion factor. Work history data for pottery and tungsten workers are still being collected. When they become available, the results of this study will also be used to establish exposure–response relationships and assess the risk of developing silicosis with cumulative respirable crystalline silica exposure among pottery and tungsten workers.

DISCLAIMER

Mention of a product or company name does not constitute endorsement by the National Institute for Occupational Safety and Health.

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