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Exposure to silica dust in the Danish stone industry

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GUÉNEL P, BREUM NO, LYNGE E. Exposure to silica dust in the Danish stone industry. Scand J Work Environ Health 1989;15:147—53. Exposure to silica dust among Danish stone workers was assessed from data collected in 1948—1980. After 1970, the exposure level was given in milligrams per cubic meter and an exposure index (concentration of respirable dust divided by the threshold limit value for quartz) was calculated. The median index was 2.1 for the road and building material industry and 0.6 for the stoneworking industry. Crushing showed a median index of 2.6, compared to 1.0 for drilling, 0.9 for sieving, and 0.5 for cutting. The median index for the road and building material industry was higher for Bornholm than for other parts of Denmark. The median index of the stoneworking industry was 1.0 for Copenhagen and 0.5 for other parts of Denmark, and no data were available for Bornholm. Before 1970, the exposure level was given in particles per cubic meter, and few data were available on quartz content. Crushing showed the highest exposure before 1970.

Key terms: occupational4 air pollutants, silica.

Crystalline silica is a well-known agent of pneumoconiosis, and in recent years investigations on humans and animals have strengthened the hypothesis that silica may also be a carcinogen. The International Agency for Research on Cancer (IARC) recently concluded that there is sufficient evidence for the carcinogenicity of crystalline silica in experimental animals and limited evidence for its carcinogenicity in humans (1).

In order to provide further information on the possible carcinogenicity of silica in humans, we identified a cohort of stone workers in Denmark. An assessment of the exposure to silica dust was carried out from data retrieved on all measurements from the stone industry available in the archives of the Danish National Institute of Occupational Health. The analysis of these exposure data is presented in this paper.

The stone industry in Denmark can be divided into two main branches: the stoneworking industry and the road and building material industry. The work processes in the stoneworking industry include sawing, edging, surface finishing by grinding, hammering, or polishing, and carving. In the road material industry stones are crushed by machines, and the gravel is sieved through metal screens to be sorted by size. The island Bornholm, located south of Sweden, is the only place in Denmark where granite rocks are found, and granite is won in open-cast quarries. Granite has been the main material in the Danish stone industry, but sandstone has also been used in the stoneworking industry, and flint has been used in the road material industry. Flint is 100% quartz (2). Sandstone consists of grains of quartz cemented together with clay (3). Granite is a mixture of quartz, feldspar, and mica (3). The quartz content in Bornholm granite varies between 15 and 35% (4); table 1 gives the number of workers in the Danish stone industry in 1930—1980 (5—10).

Material and methods

The data set included all measurements made in the Danish stone industry in Denmark by the Danish National Institute of Occupational Health from 1948 to 1980. These measurements were made at the request of the local labor inspection officers and thus reflect either areas where a special effort for regulation has been made or areas where complaints had been made about work conditions. The records were retrieved manually from the archives of the Institute. No record was available before 1948, as the Institute was founded in that year. The available information was the date of the investigation, the name of the company, the name of the employee when a personal air sample was collected.

Table 1. Number of workers in the Danish road material industry and stoneworking industry in 1930—1980.

| Year | Road material industry | Stonecutting industry |
|------|------------------------|-----------------------|
| 1930 | 708                    | 1 701                 |
| 1940 | 2 186*                 | 1 251                 |
| 1950 | 2 156                  |                       |
| 1960 | 2 774*                 | 731                   |
| 1970 | 2 236*                 | 548                   |
| 1980 | 1 864*                 |                       |

* Includes limestone quarry, mojer quarry, salt mining, and oil and gas well drilling.
pler was used, the work process or the place of work involved during the sampling, and, in some cases, the type of stone material processed. Details on the sampling and analytical methods were also given.

The data set can be divided into two subsets according to the date of investigation. Before 1970 most of the air samples were taken in the general environment at a fixed location with an impinger or an electrostatic precipitator or by filter sampling, and with sampling periods generally of less than 1 h. The results were expressed in number of respirable particles per cubic meter (diameter range 0.5—5 μm), counted by optical microscopy. After 1970 most of the samples were taken in the breathing zone with personal air samplers. The sampler met the requirements of the Johannesburg pneumoconiosis conference (11). The sampling period was generally 6—8 h, and the time-weighted average concentration of respirable dust was expressed in milligrams per cubic meter. The content of crystalline silica in both the general environment and the personal samples was determined by X-ray diffraction. No attempt has been made to convert the count of particles into weight or vice versa, and the two subsets of data have been analyzed separately.

Measurements of respirable dust were carried out in nine stonecutting companies and in 11 road and building material companies. Some companies owned different plants throughout Denmark. The numbers of measurements, specified by sampling method and type of industry, are shown in Table 2.

The results of the personal sampler measurements were compared to the threshold limit value (TLV) for respirable dust as established by the American Conference of Governmental Industrial Hygienists (12):

$$\text{TLV (mg/m}^3\text{)} = \frac{10}{2 + \text{quartz (\%)} + 2 \times \text{cristoballite (\%)} + 2 \times \text{tridymite (\%)}}$$

The comparison with the TLV was expressed by a well-established index, which is the ratio of the dust concentration (in mg/m\(^3\)) to the TLV. With no cristoballite or tridymite, the TLV is 0.1 mg/m\(^3\) for exposure to pure quartz and 1.0 mg/m\(^3\) for exposure to 8 % quartz. As is a common practice in industrial hygiene (13), the median exposure level was used in the analysis.

**Results**

Comparison of exposure levels associated with the use of granite and flint

Cristoballite was detected in eight measurements from two road material industries and ranged between 2 and 11 % of the respirable dust. Tridymite was not present in any of the measurements from the Danish stone industry. Since the quartz content of flint is higher than that of granite, an attempt was first made to assess the exposure levels to crystalline silica associated with each of them. The results are shown in Table 3 for the personal sampling measurements. The respirable dust concentrations were 1.3 and 1.0 mg/m\(^3\) for flint and granite, respectively. The median quartz content in the dust was 23 % for flint and 13 % for granite. These values lead to a median exposure index of 4.5 for the...
work operations in which flint was involved, compared to an index of 2.0 for work operations with granite.

**Comparison of exposure levels in the road material industry and in the stonecutting industry**

From the subset of measurements carried out in the general environment, the median of the respirable dust concentration was 125 particles/cm³ in the road material industry and 205 particles/cm³ in the stonecutting industry. The quartz content of the dust was not stated in any of the measurements from the stonecutting industry and in only half of the measurements from the road material industry, with a median of 23% quartz.

The subset of personal sampling data was more informative. The results are summarized in Table 4 and Figure 1. As can be seen from Table 4, the respirable dust concentration was higher in the road material industry than in the stonecutting industry. The median quartz content of the dust was 13% in the road material industry and 9% in the stonecutting industry for a median exposure index of 2.1 in the road material industry and of 0.6 in the stonecutting industry for all of Denmark. Figure 1 shows that approximately 75 and 45% of the measurements exceeded the TLV in the road material industry and the stonecutting industry, respectively. The analysis by region shows that the median exposure index was the highest in Bornholm for the road material industry (Table 4). No data were available in Bornholm for the stonecutting industry. The median exposure index was twice as high in Copenhagen as in other parts of Denmark (1.0 versus 0.5), although few measurements were available.

 Flint was the most frequently used material in the road material industry, and granite was the main material in the stonecutting industry. The differences between the two industries could not be ascribed to type of mineral. A comparison of work processes in which only granite was used showed the same median quartz content of respirable dust for the two industries, 13 and 15%, respectively. The median exposure index was, however, 3.0 in the road material industry and 1.0 in the stonecutting industry.

**Table 4. Levels of exposure to respirable dust and to crystalline silica in the road material industry and in the stonecutting industry — personal air samples.**

| Period          | Number of measurements | Median | Range       |
|-----------------|------------------------|--------|------------|
| **Road material industry** |                        |        |            |
| Respirable dust concentration (mg/m³) |                        |        |            |
| Copenhagen      | 1968—1974              | 47     | 1.1        | 0.1—39.7   |
| Bornholm        | 1972—1977              | 24     | 1.9        | 0.3—9.4    |
| Other Denmark   | 1972—1977              | 16     | 0.8        | 0.1—7.5    |
| All Denmark     | 1968—1977              | 87     | 1.1        | 0.1—39.7   |
| Quartz content in the respirable fraction (%) |                        |        |            |
| Copenhagen      | 1968—1974              | 45     | 12         | 4—35       |
| Bornholm        | 1972—1974              | 24     | 13         | 3—25       |
| Other Denmark   | 1972—1977              | 11     | 15         | 3—27       |
| All Denmark     | 1968—1977              | 80     | 13         | 3—35       |
| Exposure index<sup>b</sup> |                        |        |            |
| Copenhagen      | 1968—1974              | 45     | 1.5        | 0.2—135.0  |
| Bornholm        | 1972—1974              | 24     | 3.3        | 0.5—18.8   |
| Other Denmark   | 1972—1977              | 11     | 2.3        | 0.7—9.7    |
| All Denmark     | 1968—1977              | 80     | 2.1        | 0.2—135.0  |
| Stonecutting industry<sup>c</sup> |                        |        |            |
| Respirable dust concentration (mg/m³) |                        |        |            |
| Copenhagen      | 1977—1978              | 13     | 0.7        | 0.2—1.6    |
| Other Denmark   | 1980                   | 8      | 0.7        | 0.4—5.4    |
| All Denmark     | 1977—1980              | 21     | 0.7        | 0.2—5.4    |
| Quartz content in the fraction (%) |                        |        |            |
| Copenhagen      | 1977—1978              | 13     | 15         | 3—30       |
| Other Denmark   | 1980                   | 8      | 6          | 3—19       |
| All Denmark     | 1977—1980              | 21     | 9          | 3—30       |
| Exposure index<sup>b</sup> |                        |        |            |
| Copenhagen      | 1977—1978              | 13     | 1.0        | 0.4—3.2    |
| Other Denmark   | 1980                   | 8      | 0.5        | 0.3—6.3    |
| All Denmark     | 1977—1980              | 21     | 0.6        | 0.3—6.3    |

<sup>a</sup> In addition to quartz, two samples contained 2% and one sample each contained 4, 5, and 7% cristoballite. Cristoballite has been taken into account in the calculation of the exposure index.

<sup>b</sup> Concentration of respirable dust divided by the threshold limit value (12) for quartz.

<sup>c</sup> No data available for Bornholm from the stonecutting industry.
Figure 1. Cumulative frequency distributions of the exposure indices (concentration of respirable dust divided by the threshold limit value for quartz) for crystalline silica calculated from personal air samples from the stonecutting (1) and the road and building material (2) industries. The broken lines represent the estimated cumulative log-normal frequency distributions.

Table 5. Levels of exposure to respirable dust associated with different work processes — general environment air samples.

| Industry   | Material            | Period      | Number of measure- | Median | Range     |
|------------|---------------------|-------------|---------------------|--------|-----------|
|            |                     |             | measurements       |        |           |
| Respirable dust concentration (particles/cm³) |                     |             |                     |        |           |
| Crushing   | Road and building material | 1955—1966 | 7                   | 725    | 90—3,000 |
| Sieving    | Road and building material | 1951—1968 | 49                  | 105    | 45—1,980 |
| Cutting    | Stonecutting        | 1950        | 12                  | 205    | 56—1,010 |
| Quartz content in the respirable fraction (%) |                     |             |                     |        |           |
| Crushing   | Road and building material | 1955—1966 | 4                   | 10     | 4—20     |
| Sieving    | Road and building material | 1951—1966 | 18                  | 25     | 8—100    |
| Cutting    | Stonecutting        |             |                     |        |           |

No data available for drilling
In addition to quartz, one sample contained 11% cristoballite.

Figure 2. Cumulative frequency distributions of the respirable dust concentrations measured from the general environment samples collected during the following work conditions: sieving (1), stonecutting or surface finishing (2), and crushing (3). The broken lines represent the estimated log-normal cumulative frequency distributions.
Comparison of exposure levels in different work processes

The work processes in the road material industry were divided into drilling, crushing, and sieving. The number of measurements from the stonecutting industry was small, and hand cutting and surface finishing were therefore grouped into a single category.

The results of the measurements carried out in the general environment are presented in table 5 and figure 2. No data were available for drilling. From seven measurements only, crushing appeared to be the dustiest work operation. In figure 2 the distribution of the particle concentration is presented by work operation, and the median dust exposure levels of the work processes are readily estimated from the graphs. It should be noted that the quartz content of the respirable fraction has not been taken into account in that figure, since the percentage of quartz was not measured for cutting and was measured in four cases only for crushing.

The subset of personal air sampling data is presented in table 6. The respirable dust concentration associated with crushing was approximately twice as high as the concentration associated with the other work operations, for which the median dust concentration was below 1.0 mg/m³. The quartz content ranged from 7 to 14 %; the highest value was observed for crushing and the lowest for cutting. The median exposure index was 2.6 for crushing, and 80 % of the measurements were above the TLV. The median exposure index was 1.0, 0.9, and 0.5 for drilling, sieving, and cutting, respectively.

Finally, the data previously grouped in the cutting category were analyzed. A comparison was made between surface finishing (five measurements) and hand cutting (13 measurements). The median respirable dust concentration was 0.8 (range 0.4—3.0) mg/m³ for surface finishing and 0.6 (range 0.2—5.4) mg/m³ for hand cutting. Three measurements out of five exceeded the TLV for surface finishing, whereas the corresponding values for hand cutting were four out of thirteen.

Discussion

The exposure to siliceous dust in foundries, mines, and pottery manufacturing has been thoroughly investigated, and these environmental data have been reviewed by IARC (1). A study by Froines et al (14) also evaluated the exposure to silica in industries in the United States (US).

The exposure levels measured in foundries have been generally high. In ferrous foundries in the US 41 % of the observations exceeded the TLV for quartz, and the exposure index was 1.3 for the period 1979—1982 (14). Another US study of more than 10 000 samples analyzed by the National Institute for Occupational Safety and Health (NIOSH) and collected in foundries in 1972—1982 reported that 23 % of the samples exceeded twice the TLV (1). In pottery manufacturing, significant exposure to silica was also observed in the

| Table 6. Level of exposure to respirable dust associated with different work processes — personal air samples. |
|-------------------------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Respirable dust concentration (mg/m³)            | Industry        | Material        | Period           | Number of measures | Median          | Range           |
| Drilling                                        | Road and building material | Granite | 1974             | 6                | 0.9             | 0.5—7.1         |
| Crushing                                       | Road and building material | Granite, flint | 1968—1974       | 19               | 1.8             | 0.3—39.7        |
| Sieving                                        | Road and building material | Flint, no data | 1972—1974       | 11               | 0.5             | 0.1—4.5         |
| Cutting                                        | Stonecutting    | Granite, marble | 1977—1980       | 18               | 0.6             | 0.2—5.4         |
| Quartz content in the respirable fraction (%)   | Drilling        | Road and building material | Granite | 1974             | 6                | 10              | 7—25            |
| Crushing                                       | Road and building material | Granite, flint | 1968—1974       | 18               | 14              | 3—33            |
| Susician                                      | Road and building material | No data         | 1972—1974       | 5                | 12              | 3—16            |
| Cutting                                        | Stonecutting    | Granite, marble | 1977—1980       | 18               | 7               | 3—24            |
| Exposure index b                              | Drilling        | Road and building material | Granite | 1974             | 6                | 1.0             | 0.6—13.5        |
| Crushing                                       | Road and building material | Granite, flint | 1968—1974       | 18               | 2.6             | 0.6—135         |
| Sieving                                        | Road and building material | No data         | 1972—1974       | 5                | 0.9             | 0.4—5.8         |
| Cutting                                        | Stonecutting    | Granite, marble | 1977—1980       | 18               | 0.5             | 0.3—6.3         |

a In addition to quartz, one sample contained 4 % cristoballite. Cristoballite has been taken into account in the calculation of the exposure index.

b Concentration of respirable dust divided by the threshold limit value (12) for quartz.
US, and 73% of the samples exceeded the TLV (14). In addition, 23% of the samples analyzed in the pottery industry by NIOSH exceeded twice the TLV (1). An analysis has been made of 45,000 respirable dust samples collected by the US Mine Safety and Health Administration in 1977—1981. The geometric mean of the measurements of respirable silica dust was below 0.05 mg/m³ for most of the mining industries and work operations (1).

The exposure to silica in these industries thus appears to be on the same order of magnitude as found around 1980 in the Danish stone industry. In Denmark 75% of the measurements exceeded the TLV in the road material industry, and the corresponding value for the stonemasonry industry was 45%. The median exposure to respirable silica was 0.16 mg/m³ for the road material industry and 0.05 mg/m³ for the stonemasonry industry.

Unlike the stone industry, however, the exposure to silica dust generally occurs in foundries, pottery manufacturing, and mines in association with other materials (1, 15—21). Studies of cancer incidence or mortality in groups of workers with pure exposure to silica, such as stone workers, are therefore important for assessment of the possible carcinogenic properties of silica dust.

Environmental data analyses incorporated into cancer mortality studies of granite industry workers are available from the US and Finland. In Vermont (US), a proportional mortality analysis among granite workers found a ratio for lung cancer of 1.2 [95% confidence interval (95% CI) 0.9—1.5] in comparison with the national figures (22). Exposures were estimated from six environmental surveys covering the period 1924—1977. The estimated average exposure to respirable silica was 0.07 mg/m³ for cutters after 1950. Other environmental studies (1) in five granite industries in Vermont, carried out in 1970, found the highest exposure levels to respirable silica among cutters (0.06 mg/m³) and sculptors and carvers (0.09 mg/m³), and data from Vermont and Georgia from 1973—1974 showed mean exposure levels to respirable silica of 0.10 and 0.06 mg/m³, respectively, for stonecutters. By comparison the exposure to respirable silica in the present study was 0.06 mg/m³ for the stonemasonry industry as a whole and 0.04 mg/m³ for cutting operations only.

A proportional mortality analysis among the members of the US Granite Cutters Union showed a slight excess of lung cancer (23) (proportional mortality ratio 1.19, 95% CI 0.97—1.46). From the 1,905 death certificates analyzed, about two-thirds came from Vermont, where the exposure levels to silica were documented. For the remaining third of the subjects, the study by Froines et al (14) provides relevant exposure data from the stone industries throughout the US. The percentage of inspections with test samples over the TLV was 40% in this study, and the median exposure index was 0.8. In the Danish stonemasonry industry, 45% of the measurements exceeded the TLV and the median exposure index was 0.6.

A Finnish study (24) among granite workers showed an overall standardized mortality ratio (SMR) of 129 for lung cancer (22 observed, 17.1 expected). The mortality from lung cancer was excessive for workers with at least 15 years since entry into granite work (SMR 221, 95% CI 137—338). Exposure data were available from a survey made in 1970—1972. The highest concentrations of silica dust were observed for drilling, the hygienic standard (0.2 mg/m³) for quartz being exceeded tenfold on the average and the range being 0.3—4.2 mg/m³. This value is higher than in the present study, in which the median exposure to quartz dust was 0.09 (range 0.03—1.20, mean 0.43) mg/m³ for drilling. The range of the quartz concentration was also high for block surfacing in Finland (0.2—4.9 mg/m³ of respirable quartz) compared to the findings in Denmark (0.03—0.57 mg/m³ of respirable quartz). Finally, the range of the respirable quartz concentration for other work processes was 0.02—3.6 mg/m³ in Finland, compared to 0.03—1.7 mg/m³ for other granite operations in Denmark.

The measurements from the stone industry available in the archives of the Danish National Institute of Occupational Health were relatively few, but they were coherent with the findings from larger samples available from other countries and collected in similar industries. In Denmark, differences in exposure levels appeared between the branches of activity, the materials processed, and the work operations. On the average, the road material and building industry was associated with exposure to crystalline silica that was three times higher than that found in the stonemasonry industry, and crushing was associated with higher exposure levels than drilling, sieving, and cutting. These environmental data provide a reference for the interpretation of epidemiologic results.

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