Respiratory Toxicity of Raw Materials Used in Ceramic Production

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Abstract: The main purpose of this study was to assess the pulmonary reactions associated with exposure to raw materials used in ceramic production (RMCP). This was a cross sectional study in which 33 male workers with current exposure to RMCP and 20 healthy male unexposed workers (referent group) were interviewed and respiratory symptom questionnaires were administered to them. Furthermore, they underwent chest X-ray and lung function tests. Additionally, personal dust monitoring for airborne inhalable and respirable dust was carried out at dusty areas of the industry. To determine the chemical composition, possible silica phases and SiO₂ contents of dust samples, they were analyzed by both X-ray diffraction (XRD) and X-ray fluorescence (XRF) techniques. Demographic and socioeconomic variables of both groups were similar, except that referent individuals were, to some extent, older and heavier than their exposed counterparts. Personal dust monitoring showed that the concentrations of inhalable and respirable dust were very high and dust contained large amounts of crystalline silica. Additionally, respiratory symptom questionnaires revealed that exposed workers, compared to their unexposed counterparts, had higher prevalences of cough, wheezing, phlegm and shortness of breath. Likewise, significant decrements in some parameters of pulmonary function were noted and most of the exposed subjects showed abnormalities in their chest radiographs. These data provide further evidence in favor of the notion that exposure to RMCP, probably due to their silica contents, is associated with respiratory symptoms, radiographic abnormalities and functional impairments.

Key words: Ceramic dust, Occupational exposure, Respiratory toxicity

Introduction

Employees of ceramic production, manufacturing of wall tiles and sanitary-ware potteries (i.e., manufacturer of china plumbing fixtures) are occupationally exposed to crystalline silica^{1–3}. Long term inhalation exposure to dust during production of ceramic has been associated with elevated risk of pneumoconiosis, chronic bronchitis and ventilatory disorders among both male and female workers^{4–6}. For instance, dust exposure and pneumoconiosis were investigated in a South African pottery. X-ray diffraction showed that the quartz content of the clays used by the pottery ranged 23% to 58% and 80% of the workers employed were potentially at risk of occupational lung disease². Similarly, based on radiographic evi-

dence, cases of silicosis with a clear dose-response gradient for years of exposure as well as a significant reduction in Forced Vital Capacity (FVC) and Forced Expiratory Volume in the first second FEV₁ have been reported among ceramic and pottery workers in Italy^{4, 7}. However, in other studies no significant decrements have been detected in the parameters of pulmonary function of both ceramic and pottery workers with less than 21 yr of exposure history⁸.

Recently concern was raised about workers employed in a local ceramic production plant in Shiraz, South of Iran, over their exposure to dust. The exposed workers in the current study, during their periodic health surveillance, were found to complain from respiratory symptoms. Given their short length of service and employment of about 6.5 yr and the fact that to the best of authors' knowledge, to date, the effects of inhalation

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exposure to raw materials used in ceramic production had not been previously studied among employees of the local industries in Shiraz, this peculiar observation prompted an investigation of the potential toxic respiratory effects of the inhaled dust. The aims of the present cross sectional study were, therefore;

- 1- To assess the extent to which workers in the industry were exposed to respirable and inhalable mixed dust and quartz;
- 2-To determine whether (or not) despite the short length of exposure, there exists any abnormalities in the chest radiographs of exposed workers;
- 3- To find out whether (or not) there exists any significant differences in the prevalence of respiratory symptoms between dust-exposed workers and a non-exposed referent group;
- 4-To find out whether (or not) exposure to ceramic dust has resulted in significant decrements in the parameters of pulmonary functions of exposed employees.

Materials and Methods

Studied population

This cross-sectional study was carried out at a local ceramic producing factory in Shiraz, South of Iran. A total of 33 male workers with past history and current exposure to ceramic dust were included in the study. They were working in two major dusty areas, as detailed in the measurement of atmospheric dust section. Similarly, 20 healthy males from the same industry with almost identical socioeconomic and demographic status (sex, ethnic background, education, smoking habits, income as well as family size) served as the referent group.

Both groups were volunteer subjects. No selected subjects refused to participate in the study. Additionally, the study was conducted in accordance with the Helsinki Declaration of 1964 as revised in 2000⁹⁾. All participants signed an informed consent form before commencement of the study. None of the exposed subjects had past medical or family history of respiratory illnesses or any other chest operations or injuries. Similarly, none of the referent subjects had been exposed to ceramic or other chemicals known to cause respiratory symptoms, ventilatory disorders or abnormal radiographic changes in the lungs during their employment with the plant or prior to it. The prevalence of respiratory problems and changes in the parameters of lung function were studied among the above- mentioned exposed and unexposed groups.

Measurements of Study Variables

Respiratory Illness

Subjects were interviewed and respiratory symptom

questionnaire, as suggested by the American Thoracic Society (ATS 1978)¹⁰⁾, with a few modifications, were administered to all of them. This standardized questionnaire included questions on respiratory (presence or absence of regular dry and/or productive cough, wheezing, shortness of breath, etc), nasal and eye symptoms, smoking habits, medical and family history of each subject. Additionally, it contained detailed occupational history and specific questions concerning all jobs held before employment at the plant under study, particularly those associated with the risk of respiratory morbidity. These, were then used to obtain symptom prevalence data among exposed and unexposed groups.

Measurement of Atmospheric Dust

To assess the extent to which workers had been exposed to ceramic dust, personal dust monitoring for airborne inhalable (particle size $\geq 5 \mu$) and respirable fractions (particle size $< 5 \mu$) was carried out in different dusty worksites. Dusty worksites included two major areas; (1) manual blending of raw materials, (2) grinding and crushing of the coarse materials. For estimation of the atmospheric dust concentration in these units, a personal dust sampler (Casella, London-LTD), calibrated by a digital automatic calibrator connected to a filter holder equipped with a 25 mm membrane filter (pore size 0.45μ) through which air was pumped by a battery-powered motor at a constant flow rate of 2 l/min was used. Based on a few preliminary tests, the optimum sampling time, to avoid overloading of the filters, was evaluated to be 80 min.

Dust concentration expressed in mg/m^3 was calculated from the changes in the dried filter (for respirable fraction) or cyclone collector weight (for inhalable fraction), as measured by a digital scale at a sensitivity of 0.1 mg, before and after sampling, divided by the volume of air sampled.

Pulmonary Function Tests

Pulmonary function tests (PFTs), including mean percentage predicted Vital Capacity (VC), (FVC), (FEV_1), Peak Expiratory Flow (PEF), Forced Expiratory Flow between 25% and 75% of the FVC ($\text{FEF}_{25-75\%}$), followed guidelines given by the ATS (1979)¹¹⁾ and measured with a portable calibrated vitalograph spirometer (Vitalograph-COMPACT, Buckingham-England) on site. The spirometer was calibrated twice a day with a 1-liter syringe in accordance to the standard protocol for the instrument used. The mean percentage predicted value was based on a subject age, weight, standing height, sex and ethnic group as calculated and adjusted by spirometer device. Subjects were requested not to take shower or smoke for at least two hours prior to the test. Additionally, they were trained to become familiar with the maneuvers. The

standing height and weight of each subject were measured in his normal working clothes. Before the test, they rested in a sitting position for about 5 min. They were then asked to stand in front of the spirometer, as comfortably as possible, and a nose clip was put on. At least, three acceptable maneuvers were performed. If subject showed great variability among the various FVC volumes, up to eight maneuvers were obtained. The largest volumes (as percentage predicted lung function) were selected for analysis. The percentage predicted lung values were observed capacities as measured by spirometer device divided by predicted or expected capacities (based on gender, age, weight, height, ethnic background, etc, as calculated and adjusted by spirometer device) multiple by 100.

% Predicted lung value = (observed capacities/expected capacities) $\times$ 100

Chest Radiography

Subjects were invited to a medical center and underwent Posterior-Anterior (PA) chest x-ray, using a Siemens instrument. Standard PA chest X-rays were read by a radiologist. The size of the film (Agfa Company) was 35 $\times$ 35 cm, the distance of the subject with X-ray tube was about six feet, the electrical voltage was 100 kv and the intensity was about 500 mA.

Chemical Composition of Dust Samples

XRD and XRF techniques were performed to determine the chemical composition, the silica phases and the SiO₂ contents of the dust samples.

Data Analysis and Statistical Procedures

The data were statistically analyzed using student's *t*-test (or Welch's alternate *t*-test, when the SD of two com-

parable variables with the F-test were significantly different) and χ^2 or Fisher exact test, where applicable (with a preset probability of $p < 0.05$). When the detection of an effect caused by an independent variable in any particular field was not predictable, statistical analysis was conducted using a two-side *p*-Value. Experimental results are presented as arithmetic Mean $\pm$ SD. Statistical tests were conducted using GraphPad InStat tm, V204a, 1990–1993 on a personal computer. Additionally, using multiple linear or logistic regression analysis, the simultaneous effects of confounding variables such as smoking, age, weight, height, etc, on the prevalence of respiratory symptoms and changes in the parameters of pulmonary function were evaluated.

Results

Subjects' physical characteristics, their duration of exposure to ceramic dust, number of smokers and length of smoking in both groups, major components of dust samples as well as concentrations and SiO₂ contents of inhalable and respirable dusts are shown in Table 1. As seen, the average (Mean $\pm$ SD) age (yr), weight (kg), height (cm), duration of exposure to dust and length of smoking (yr) for the exposed group were 31.3 $\pm$ 4.2, 68.8 $\pm$ 10.2, 175.7 $\pm$ 7.6, 6.5 $\pm$ 2.0 and 4.5 $\pm$ 6.3, respectively. The corresponding values for the referent group were 39.6 $\pm$ 6, 74.9 $\pm$ 9.1, 173.6 $\pm$ 5.3, 0 $\pm$ 0 and 6.8 $\pm$ 4.9, respectively. Additionally, XRD and XRF revealed that the crystalline silica phase of the samples was Quartz and the sample contained 69% SiO₂. Similarly, levels of exposures to inhalable and respirable dust were estimated to be 71.7 $\pm$ 16.2 and 26.7 $\pm$ 19.2 mg/m³, respectively. These data indicate that unexposed subjects are, to some extent, older and heavier than exposed individuals and the

Table 1. The physical characteristics and exposure data (Mean $\pm$ SD) and smoking habits of study subjects

Parameter	Exposed (n=33)	Unexposed (n=20)	<i>p</i> -value ¹
Age (yr)	31.3 $\pm$ 4.2	39.6 $\pm$ 6	<0.0001
Weight (kg)	68.8 $\pm$ 10.2	74.9 $\pm$ 9.1	0.03
Height (cm)	175.7 $\pm$ 7.6	173.6 $\pm$ 5.3	0.28
Years smoked (yr)	4.5 $\pm$ 6.3	6.8 $\pm$ 4.9	0.16
Number of smokers	7	6	0.69
Duration of exposure to dust (yr)	6.5 $\pm$ 2.0	N/A	N/A
Inhalable dust concentration (mg/m ³)	71.7 $\pm$ 16.2 ^a	N/A	N/A
Respirable dust concentration (mg/m ³)	26.7 $\pm$ 19.2 ^a	N/A	N/A
Type of the crystalline silica phase	Quartz ^b	N/A	N/A
SiO ₂ content (%)	69 ^b	N/A	N/A
Al ₂ O ₃ content (%)	18 ^b	N/A	N/A
Loss of Ignition (LOI) (%)	6 ^b	N/A	N/A

¹There was no statistically significant difference between any of these means, except for age and weight (student's *t*-test or χ^2 statistics with Yates correction, $p < 0.05$). a: n = 12 ; b: n = 3 N/A: Not applicable.

Table 2. Frequency (%) of abnormal clinical findings in exposed and control subjects

Parameter	Exposed (n=33)	Unexposed (n=20)	p-Value	χ^2	Relative risk
Cough	18.2	5	0.170	0.9128	3.636
Phlegm	39.4	5	0.007*	5.912	7.879
Wheezing	21.2	5	0.110	1.440	4.240
Breathlessness	30.3	5	0.030*	3.430	6.060

*Significantly different from control (χ^2 statistics with Yates correction, $p < 0.05$).

Table 3. Frequency (%) of abnormal findings in chest radiographs of exposed and control subjects

Abnormality	Exposed (n=33)	Unexposed (n=20)	p-Value	χ^2
Reticulonodular Shadows	76*	0	<0.0001	25.72
Reticulonodular Shadows associated with Prebronchial Thickening	9.1	0	0.23	0.6008

*Significantly different from control (Fisher's exact test, $p < 0.05$).

differences were statistically significant. However, apart from these differences, as far as the other variables are concerned, no significant differences exist between exposed and unexposed employees. Similarly, other socioeconomic and demographic factors (ethnic background, education, income and family size), their data for the sake of clarity are not shown, of both groups were identical and none of the exposed or referent individuals had past medical or family history of respiratory illnesses, chest operations or injuries.

Table 2 illustrates the frequency of abnormal clinical findings among exposed and referent subjects. As shown, data extracted from respiratory symptom questionnaires revealed that exposed workers, compared to their unexposed counterparts, regardless of their smoking habits, had higher prevalence of regular cough (18.2% vs. 5.0%), phlegm (39.4% vs. 5%), wheezing (21.2% vs. 5.0%) and shortness of breath (30.3% vs. 5.0%). Table 3 depicts the frequency of abnormal findings in chest radiographs. As shown, while a majority (76%) of radiographs of exposed workers showed abnormality (reticulonodular shadows), no significant changes were noted in the radiographs of unexposed subjects.

The results of lung function tests are presented in Table 4. As shown, exposure to ceramic dust was associated with a significant reduction in VC, FVC and FEV₁ parameters. However, no significant differences were noted between PEF and FEV₁/FVC ratios of both groups.

Association between exposure to ceramic dust and changes in the parameters of pulmonary function is shown in Table 5. Multiple Linear Regression Analysis, including independent variables of age, weight, height, length of smoking and length of exposure to ceramic dust in the model showed that after adjusting for important confounders, there was statistically significant associations between length of exposure to ceramic dust with VC, FVC

Table 4. Percentage predicted lung function among exposed and control subjects

Parameter	Exposed (n=33)	Unexposed (n=20)	p Value
VC	92 ± 27.5 ^a	105.6 ± 17.8	0.016
FVC	92.9 ± 27.5 ^a	107.6 ± 15.7	0.008
FEV ₁	95.0 ± 30.2 ^a	108.9 ± 16.7	0.018
PEF	88.1 ± 41.765 ^a	103.7 ± 14.9	0.06
FEV ₁ /FVC	107.0 ± 35.5 ^a	101.4 ± 8.5	0.39

* Significantly different from control value (student's or Welch's alternate *t* test, $p < 0.05$).

^a% predicted lung function = % observed/predicted.

Table 5. Association between exposure to ceramic dust and changes in the parameters of pulmonary function (n=33)

Parameter (dependent variable)	Regression coefficient (β)	SE	p-value ^a
VC	-17.93	7.8	0.03
FVC	-26.58	7.08	<0.0001
FEV ₁	-28.16	7.4	<0.0001
FEV ₁ /FVC	-6.84	8.98	0.45
PEF	-9.6	12.38	0.44

^aMultiple Linear Regression Analysis.

and FEV₁ values; in that, there was a general tendency for these values to become smaller as estimated cumulative exposure increased (Table 5).

Similarly, the association between exposure to ceramic dust and the prevalence of respiratory symptoms is presented in Table 6. Logistic Linear Regression Analysis including independent variables of age, weight, height, length of smoking and length of exposure to ceramic dust in the model showed that after adjusting for important confounders, there was statistically significant associations between length of exposure to ceramic dust with the prevalence of cough, phlegm and shortness of breath.

Table 6. Association between exposure to ceramic dust and the prevalence of respiratory symptoms

Respiratory symptoms (Parameter)	Regression coefficient (β)	SE	Odd Ratio	<i>p</i> -value ^a
Cough	2.60	1.34	13.50	0.05
Phlegm	2.51	1.08	12.35	0.02
Wheezing	1.63	1.11	5.11	0.14
Shortness of breath	2.11	1.03	8.26	0.05

^aLogistic Regression Analysis.

Although this association did not reach statistical significance for wheezing, the odd ratio of this symptom was quite remarkable (Table 6).

Discussion

Given the data provided, there should be very few, if any, socioeconomic and demographic differences between ceramic-exposed and unexposed subjects; as they were from the same industry with almost identical annual income, education, sex, ethnic background, etc. Additionally, none of the subjects had past medical or family history of respiratory illnesses or any other chest operations or injuries. Since there were no significant differences in the major confounding variables of cigarette smoking, past history of medical illnesses, family history, socioeconomic and ethnic factors, the decrements in the parameters of pulmonary function, increased prevalence of respiratory symptoms and abnormal radiographic findings are likely to be the direct results of exposure to ceramic dust.

Similarly, as dust exposed and unexposed subjects had smoked the same length of time, it is unlikely that cigarette smoking accounts for the differences in symptoms between the two groups.

Interestingly, while the average length of exposure to ceramic dust was as small as 6.5 yr, prevalence of respiratory symptoms among exposed individuals who were also younger than referent group was higher than their unexposed counterparts, significant reductions in lung function parameters of exposed subjects were noted and the majority of radiographs of exposed subjects showed some degree of abnormality. However, no significant changes were noted in the radiographs of unexposed employees.

Compared to referent subjects, workers at the plant were exposed to significant concentrations of (Table 1) ceramic dust (several times higher than the current standard¹²) of 0.1 mg/m³ and 0.2 mg/m³ for respirable and inhalable fractions, respectively). This heavy exposure scenario may well explain the cause of an increased prevalence of respiratory symptoms (cough, phlegm, dys-

pnea and wheezing) observed in this study (Table 2) as well as the significant reductions in some parameters of lung function such as VC, FVC and FEV₁ (Table 4). Similar findings have been reported by other investigators^{4-7, 13}).

Our findings are also in line with studies where silica dust has been introduced as one of the most toxic mineral dusts, causing fatal disease after exposure for only a few months to concentrations not perceptible to the naked eye^{1, 14}).

Conversely, in some studies⁸) no significant decrements in FEV₁ and FVC values of subjects with less than 21 yr of exposure to low concentrations of ceramic dust has been observed. This apparent discrepancy may well be explained by the fact that the exposure scenario of the above-mentioned study totally differs from that of our study; in that, the concentration of respirable dust fraction was 0.08 mg/m³, whereas, its corresponding value in our study was more than 300 times higher (26.7 mg/m³).

Given the above, one would expect, in our study, cases of subacute (accelerated) silicosis, which generally follows, compared to chronic silicosis, shorter (i.e., 2–5 yr), heavier exposures to silica dust¹⁵) and chronic silicosis in the exposed population described in reference 8, which usually follows greater than 10 yr of exposure to respirable dust with less than 30% quartz¹⁵). Infact, this was the case, as in our study ventilatory disorders and radiographic abnormalities in individuals with less than 10 yr of exposure were shown. In contrast, pulmonary function tests of subjects with less than 21 yr of exposure to ceramic dust, as reported by Bahrami and Mahjub⁸) were similar to those of control subjects. However, for subjects with more than 21 yr of exposure history, FEV₁ and FVC were significantly lower than those of unexposed population.

The absence of any significant difference in the FEV₁/FVC ratios of exposed and referent subjects, deserves comment. This observation may indicate that occupational inhalation exposure to raw materials used in ceramic production, probably due to their silica content, is likely to induce restrictive ventilatory impairment. This proposition is in full agreement with the views expressed by Kumar *et al.*¹⁶) indicating that in restrictive pulmonary diseases, FVC is reduced but the expiratory flow rate, usually measured by FEV₁, is normal or reduced proportionately. Hence, the ratio of FEV₁/FVC is near normal.

In this study, exposed workers had a significantly higher rate of radiographic abnormalities than their unexposed counterparts. This finding is also in line with other studies where higher rates of chronic bronchitis and pulmonary emphysema have been reported in workers exposed to cement dust containing silica when compared to normal population^{17, 18}).

Conclusions

In conclusion, our data provide corroborative evidence to further substantiate the contention that occupational inhalation exposure to raw materials used in ceramic production, most probably due to their silica content, even after a short period of exposure, is associated with respiratory symptoms, radiographic changes and functional impairments. Given the high silica content of raw materials used in ceramic production, these results further justify the use of proper personal protective respirators, while at work, and the reduction or elimination of dust, using appropriate ventilation system, to protect the workers from developing more severe chronic respiratory diseases in the future.

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