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Inhalation of china stone and china clay dusts: relationship between the mineralogy of dust retained in the lungs and pathological changes

JC WAGNER, FD POOLEY, A GIBBS, J LYONS, G SHEERS, CB MONCRIEFF

From the Medical Research Council External Staff Team on Occupational Lung Diseases, Llandough Hospital, Penarth; the Institute of Materials, University of Wales; the Department of Pathology, Llandough Hospital, Penarth; the Medical Boarding Centre (Respiratory Diseases), Cardiff; the Chest Clinic, Plymouth; and the National Institute of Agricultural Engineering, Silsoe, Bedford

ABSTRACT A combined clinical, radiological, pathological, and mineralogical study was undertaken on 62 cases referred to the Medical Research Council Pneumoconiosis Unit by the Cardiff Pneumoconiosis Panel as Cornish china clay workers. Considerable pathological lesions were found in the lungs, both nodular and interstitial fibroses being present. Some men had worked with china stone but others had worked entirely with china clay. Nodular fibrosis appeared to be related to a high quartz content of the dust recovered from the lung, whereas among those with a high content of kaolinite dust in the lungs interstitial fibrosis was observed.

Reports of pneumoconiosis in Cornish china clay workers have been made by Hale *et al*,[1] Sheers,[2] and Oldham.[3] It has not been clear, however, which minerals are responsible for the pulmonary lesions. Some cases have been complicated by possible exposure to other dusts and by tuberculosis. The results of an initial study of the mineral content of the lungs of one Cornish china clay worker showed an excess of silica,[1] but the analytical technique is open to criticism.

The china clay and china stone deposits in Cornwall were discovered in the eighteenth century and were initially worked as a source of raw material for the then expanding British pottery industry. Since that time the production of china clay has grown, mainly as a result of its use as a filler in paper, while china stone production has ceased. China clay consists mainly of the mineral kaolinite and in most other countries it is referred to as kaolin. The main ancillary minerals present in china clay are quartz, micas, and feldspars; commercial samples contain over 90% kaolinite. China stone, if hand picked, consists essentially of a mixture of quartz, feldspars, micas, and amorphous silicon dioxide.

It was considered that if the disease among these workers had been due to exposure to excessive amounts of silica dusts the most likely source was the grinding of china stone. Some of the lungs sent to the pathology section of the Medical Research Council Pneumoconiosis Unit, however, were from workers who appear to have been employed exclusively in the preparation of china clay. It was important to confirm this finding and consider what implications there might be for the industry.

Address for reprint requests: Dr AR Gibbs, Department of Pathology, Llandough Hospital, Penarth, S Glam CF6 1XW.

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Methods

CLINICAL INFORMATION

From 1968 to 1981 the lungs of 62 men who had recently died and who had apparently been employed in the production of china clay or china stone were received at the MRC Pneumoconiosis Unit. They had all been referred by the Cardiff Pneumoconiosis Medical Panel for an assessment of the severity of lung disease. In all cases the coroner had been informed that exposure to china clay dust might have contributed to the cause of death, this being the essential criterion for inclusion in the study. Most of these men had retired from the industry. All cases in the study were male. Smoking histories were not available and this aspect was ignored.

Of these 62 workers, three had worked with ball clay and one with slate; in three cases there was no histological evidence of pneumoconiosis and in one no adequate evidence of employment in the china clay industry. The remaining 54 workers, all from Cornwall, had all worked with china clay or china stone.

Occupational histories were derived from several sources, including the Pneumoconiosis Medical Panel, hospital and clinical records, and the china clay industry. Information from these sources was combined into a working history, and cross checked so far as possible. For a few cases this history lacked detail—for example, "clay worker at location *x*," where the works might have closed down some years ago and could have been a drier or pit. The best that could then be done was to make an enlightened guess of the actual occupation. In many cases the number of years of employment shown is less than the total number of years worked.

The available information was broken down as follows: O1—dusty china clay, assigned to those who had worked in drying kilns or mechanical dryers; O2—wet, non-dusty china clay, assigned to those who had worked in the clay pits (wet) or wet processing plants; O3—china stone, assigned to those who had worked in the mining, crushing, and grinding of china stone (usually a dry process); O4—other dusty environments, assigned to those who had been employed in a different extraction industry, such as tin or coal, or in hard rock quarrying or as farmers.

RADIOLOGY

Chest radiographs were available for 39 of the cases and they were read by three readers using the 1971 ILO classification.[4] This provides separate readings for the profusion of rounded and irregular opacities in addition to the assessment of the combined profusion of small opacities. Also provided is an assessment of progressive massive fibrosis, given by the size of large opacities. The median readings were used in the analysis.

PATHOLOGY

The lungs were described macroscopically and representative blocks taken. From four to 13 blocks per case were examined histologically for nodular and interstitial fibrosis and an overall grade, based on all the sections, was obtained for each case (this grading was modified from that previously described for coal worker's pneumoconiosis[5 6]). Both fibroses were graded on ordinal scales as follows:

Nodular fibrosis 0 (none)—no nodules seen in any of the sections; 1 (slight)—up to two nodules per section, all of less than 3 mm diameter; 2 (moderate)—up to seven nodules per section or nodules of 3–6 mm diameter; 3 (severe)—more than seven nodules per section or nodules of 6–10 mm diameter. Nodular lesions greater than 1 cm in diameter were recorded separately as progressive massive fibrosis (PMF).

Interstitial fibrosis 0 (none)—interstitial fibrosis absent; 1 (slight)—fibrosis located around respiratory bronchioles, which may extend into alveolar ducts, atria, and adjacent alveoli but with areas remaining free of fibrosis between adjacent respiratory bronchioles; 2 (moderate)—early widespread interstitial fibrosis linking adjacent respiratory bronchioles, with some obliteration of alveolar sacs; 3 (severe)—widespread diffuse fibrosis with few recognisable alveoli: honeycombing may or may not be present.

MINERALOGICAL EVALUATION

Samples of lung tissue from 42 cases were submitted for determination of mineral content and for mineralogical analysis by electron microprobe (not here reported) and by *x* ray diffraction techniques (special procedures were developed to permit quantitative analysis of the necessarily small samples, the mass in some cases being only 0·3 mg).[7] Replicate analyses were made on some of the specimens and in general showed good agreement. The cases were divided into three groups as follows: (*a*) *China clay* (16 cases)—kaolinite $\geqslant$90% by mass; subsidiary components: quartz $\leqslant$1.1% by mass, feldspars $\leqslant$1.0% by mass. (*b*) *China clay and stone* (16 cases)—kaolinite <90% by mass; subsidiary components: quartz $\geqslant$0.9% by mass, feldspars $\geqslant$1.0% by mass. (*c*) *Miscellaneous* (10 cases)—The "miscellaneous" cases were fairly diverse in their mineral compositions, being defined by their failure to meet appropriate subsidiary conditions for either group *(a)* or group *(b)*; exposure to other materials was suggested.

The numbers of cases with and without radiological and mineralogical information are given in table 1. The nine cases with radiological but not mineralogical information were accommodated into the exposure groupings by taking account of their occupational records. Thus an additional six cases were assigned to the "china clay" group and three to the "china clay and stone" group. Except when specifically mentioned, these nine cases are not included in the results.

The data obtained, for all except the three individuals in whom neither mineralogical nor radiological information was available, are given in table 2; the cases without mineralogical information are listed at the end of each section of the table.

Table 1 *Numbers of china clay cases with and without radiology or mineralogy*

		Mineralogy		
		Yes	*No*	*Total*
Radiology	*Yes*	30	9	39
	No	12	3	15
	Total	42	12	54

Table 2 *Information obtained on china clay workers*
Group (a): "china clay" group

Case No	Age at death (y)	Years in occupation 01	02	03	04	Dust concentration (mg/g)	Mineral concentration (%) Kaolinite	Quartz	Pathology N	I	PMF	Radiological opacities* Small	Large
1	84	55				26.1	95	0.7	2	1	p=3	qt 2/2	B
2	63	31				160.6	95	0.4	2	2		tq 3/3	A
3	61	15	10			39.8	96	0.5	2	2	p=4	rt 2/2	A
4	65	38	11			39.1	96	0.3	1	2		qt 2/2	
5	67	40				40.9	95	0.2	2	3		tq 3/3	
6	69	35			4	104.3	93	1.0	0	3		qt 3/3	A
7	59	20	8			21.6	96	0.2	0	3		qt 3/3	
8	82	20	27			7.6	92	0.5	2	2	p=3	qt 2/1	C
9	81	48				289.3	96	0.2	2	2	p=3	qt 2/1	C
10	64	46				19.7	94	0.1	1	3		tq 3/3	
11	76	26	25			26.1	95	0.8	1	2	p=2	qt 2/2	A
12	76	1	16			54.4	94	0.5	1	2		qr 1/1	A
13	66		5		40	8.5	90	0.9	0	1		tt 1/0	
14	84		47			57.5	93	0.9	2	2		tq 2/2	
15	54	41				53.0	95	0.6	1	2		qt 1/0	
16	69	20				40.2	96	0.2	1	1			
43	64	20	20						2	2		qt 3/2	A
44	50	20	6						0	3		ps 3/3	
45	65	39							0	1		qs 3/2	
46	64	30	8						2	1		qq 2/1	
47	57	13			17				0	2	p	qt 2/2	C
48	63	43	2		5				2	1		qq 3/+	

Group (b): "china clay and china stone" group

Case No	Age at death (y)	Years in occupation 01	02	03	04	Dust concentration (mg/g)	Mineral concentration (%) Kaolinite	Quartz	Pathology N	I	PMF	Radiological opacities* Small	Large
17	60	38				16.0	80	1.1	0	1		st 0/1	
18	74	15	23	7		4.1	75	7	3	2		rt 1/1	A
19	71	8		21		18.8	64	6	0	1		0/0	
20	79	21	5	18		35.8	77	6	3	2		qt 2/3	
21	79			26	21	10.4	20	15	3	2	p	tq 2/2	B
22	69			24	26	41.0	30	20	3	1		rr 3/3	
23	59	1		34	4	27.5	25	13	3	2	p		
24	73		41			8.1	89	0.9	0	1		0/0	
25	65		3	21		4.4	30	8	1	2			
26	66	5		26		32.3	26	20	3	2	p	rt 3/3	B
27	62	7		30		16.1	33	18	3	2		rt 2/2	
28	80		55	4		13.9	71	1.5	0	2			
29	74					5.4	15	19	2	2			
30	57					7.6	75	1.5	0	1			
31	68			14		13.9	25	14	2	1		qq 1/1	
32	65		4	30	7	44.8	41	17	3	1			
49	65			17					2	3		sq 3/3	
50	58	32		4					1	2		tt 1/1	
51	70	25			25				2	2		qr 2/2	

Group (c): "miscellaneous" group

Case No	Age at death (y)	Years in occupation 01	02	03	04	Dust concentration (mg/g)	Mineral concentration (%) Kaolinite	Quartz	Pathology N	I	PMF	Radiological opacities* Small	Large
33	61	10	31			28.7	91	3.0	2	2		tt 0/1	
34	65		15			4.7	59	3.0	1	0			
35	74					2.0	79	1.1	1	0		0/0	
36	75					11.6	87	1.4	0	1		0/0	
37	73		20			6.9	67	1.3	0	1		tq 1/1	
38	79		26			1.6	80	1.1	0	1		tt 1/1	
39	68		41		11	5.2	85	1.2	0	1			
40	62					6.5	33	5.0	0	1			
41	69	17	4			10.8	89	1.3	2	1			
42	78	10				8.0	73	0.4	1	1			

*ILO classification.[4]
N—nodular; I—interstitial; PMF—progressive massive fibrosis.

Results

OCCUPATIONAL HISTORY AND AGE

The ages at death ranged from 50 to 84 years, the mean ages being 67, 68, and 68 in the extended groups *(a)*, *(b)* and *(c)* respectively. The maximum total period of employment for any one individual was 59 years.

A history of working with china stone was recorded for none of the 16 cases in the "china clay" group, whereas it was recorded in 12 of the 16 cases in the "china clay and stone" group. It follows that results based on the mineralogically defined groups should correspond closely to those based on occupational histories.

RADIOLOGICAL DATA

Radiographs taken during life were available for 39 of the cases. These included 13 cases of progressive massive fibrosis and 22 of simple pneumoconiosis and four cases with no radiological evidence of the disease. The radiological appearances tended to reflect the dual nature of the pathological condition since nodular and irregular opacities were present concomitantly in most cases—28 out of a total of 35. Those lungs with interstitial changes and a high kaolinite content tended to show predominantly irregular radiological changes, while nodular opacities were more common in those with a high quartz content.

MINERALOGICAL DATA

The kaolinite and quartz concentrations are shown in figure 1, where they have been plotted on logarithmic scales. Cases from the three groups used in the tabulations are differentiated in this figure. Cases from the first two groups fall in separate regions; this accords with the mineral composition defining the groups and implies that a line corresponding with a kaolinite:quartz ratio of 90:1 would roughly divide these two groups.

The lung dust concentrations estimated from the specimens of tissue found within the three groups are briefly summarised in table 3. The lung dust concentrations were highest in the china clay group, were lower by a factor of about 0.4 in the china clay and china stone group, and were lower in the miscellaneous group by a further factor of 0.4. Although the dust concentrations of the samples of tissue could be obtained with high precision there may be considerable variation in different areas of the lung.[7 8] The sensitivity and precision of the x ray diffraction analyses are dependent on instrument parameters, the mass of the specimen analysed, and the individual mineral concentrations. For example, the lower limits of detection for quartz and for potash feldspar in a specimen of 4 mg mass are less than 0.1% and 0.4% respectively. Sixty one specimens (including replicates) were examined by x ray diffraction and the mean mass of the samples analysed was 4.42 mg. The precision associated with this mass of specimen has been investigated by the use of a reference sample and the results are given in table 4.

In the china clay and china stone group the percentage of kaolinite ranged from a minimum of 15% to a maximum of 89%. The maximum absolute kaolinite loading in this group was 27.5 mg/g, which was comparable with the lower levels of kaolinite in the china clay group.

PATHOLOGY AND DUST LOADING

In the china clay group (*a*) the grades of nodular

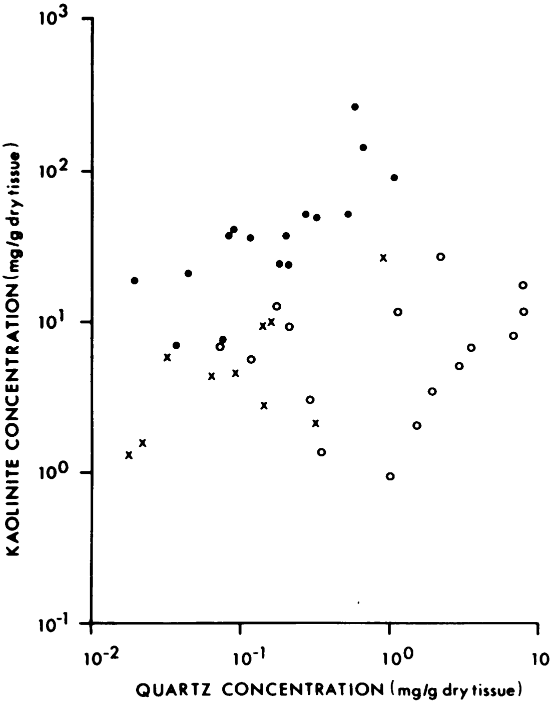

Fig 1 *Distribution of kaolinite and quartz concentrations in 42 cases with mineralogical analysis.* ●*—group (a) china clay exposure;* ○*—group (b) china clay and china stone exposure;* ×*—group (c) miscellaneous exposure.*

Table 3 *Lung dust concentrations found in three defined groups*

Mineralogical groups	*Lung dust concentrations (mg/g)*		
	Min	*Max*	*Median*
a	7.6	289.3	40.0
b	4.1	44.8	15.0
c	1.6	28.7	6.5

Table 4 *Precision of x ray diffraction for "thin" specimens prepared on silver filters*

Replicate No	*Mass on filter (mg)*	*Kaolinite*	*Mica*	*Quartz*	*Feldspar*
			(% by mass)		
1	3.995	93.8	2.7	1.5	0.9
2	3.879	95.6	3.2	1.4	1.5
3	4.320	95.6	3.2	1.4	1.5
4	3.971	94.1	3.6	1.4	0.9
5	3.998	94.0	3.3	1.5	1.2
Mean	4.033	94.5	3.1	1.4	1.0
(SD)	(0.168)	(0.8)	(0.4)	(0.2)	(0.3)
Analysis of an "infinitely" thick randomly oriented specimen gave:		96	2	1	1

fibrosis ranged from 0 (none) to 2 (moderate), but in the china clay and china stone group *(b)* half of the cases (8 out of 16) had grade 3 (severe). The nodular fibrosis grades and their relationship to the loadings of kaolinite and quartz are shown in figure 2, where the symbols used in figure 1 have been replaced by the nodular fibrosis grades. Nodular fibrosis appears to be related to the quartz concentration.

The grades of interstitial fibrosis ranged from 1 (slight) to 3 (severe) in group (*a*) and from 1 (slight) to 2 (moderate) in group (*b*). The interstitial fibrosis

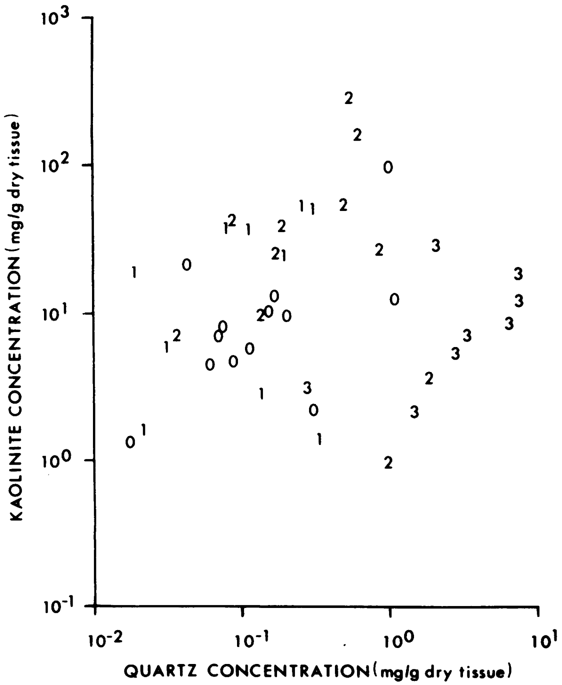

Fig 2 *Relationship of nodular fibrosis to kaolinite and quartz concentrations. Numbers indicate the degree of nodular fibrosis. 0—none; 1—slight; 2—moderate; 3—severe.*

grades are shown in figure 3, in which they replace the symbols of figure 1. Here interstitial fibrosis seems to be related to the kaolinite concentration.

In table 5 analyses of variance of the logged concentrations of quartz and kaolinite are presented. These take account of nodular and interstitial fibrosis in the 32 cases belonging to the first two groups. Clearly there is a significant relationship between quartz concentration and nodular fibrosis, but there appears to be no relationship between kaolinite concentration and interstitial fibrosis grades. The 16 cases of the first group can have had little exposure to anything but china clay and the high levels of interstitial disease in this group could plausibly be attributed to china clay.

In all five cases with large opacities diagnosed radiologically as category B or C, and in three of the six cases diagnosed as category A, progressive massive fibrosis was found at necropsy. There were cases found at necropsy to have progressive massive fibrosis in both group (*a*) (five cases) and group (*b*) (three cases).

In the miscellaneous group the grades of both nodular and interstitial fibroses were lower; out of 10 cases, there were only two with levels other than 0 (none) or 1 (slight), including one case that had grade 2 (moderate) for both fibroses.

Exploration of the loadings for kaolinite, quartz, and the four other mineral components specified in table 2 suggested that mica, feldspars, albite, and talc behaved in a manner similarly to quartz, so that the results for kaolinite would be mainly unaffected by inclusion of these additional minerals.

Discussion

The results of this study indicate that excessive exposure to china clay dust, which is largely composed of kaolinite, may cause pneumoconiosis. Pulmonary fibrosis was found at necropsy in the 16 men of group (*a*), who had no known occupational exposure to dust other than china clay. Varying degrees of fibrosis of both nodular and interstitial type were found in these

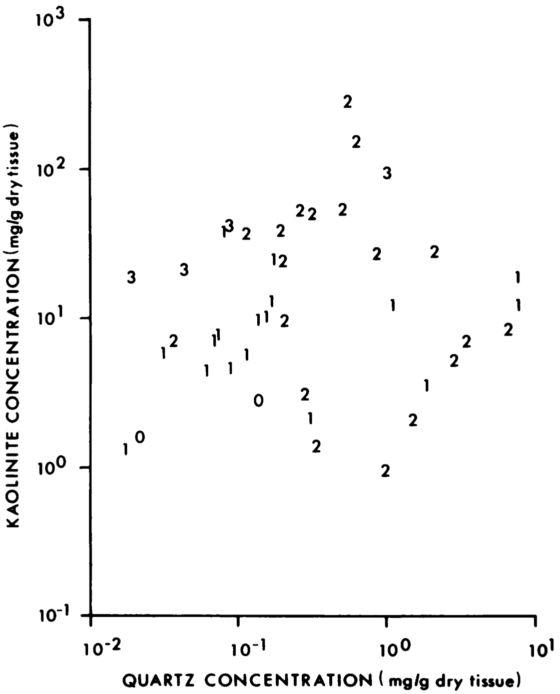

Fig 3 *Relationship of interstitial fibrosis to kaolinite and quartz concentrations. Numbers indicate the degree of interstitial fibrosis. 0—none; 1—slight; 2—moderate; 3—severe.*

16 subjects. The degree of interstitial rather than nodular fibrosis appeared to relate more closely to lung dust concentrations, although the results failed to reach statistical significance. We could speculate that a mineral in the china clay other than kaolinite might be responsible for pulmonary fibrosis. Micas have been implicated in the development of pneumoconiosis and the pathological features described appear similar to our china clay cases.[9] This explanation appears unlikely in these lungs from Cornish china clay workers, however, since most have less than 5% mica within the dust. Furthermore, pulmonary fibrosis has been found in American kaolinite workers, where the mica content is negligible.[10-15]

By contrast, in the groups exposed to china stone and china clay dust (group *b*) quartz concentrations correlated more closely with the degree of nodular fibrosis than with the degree of interstitial fibrosis. Indeed, the lung lesions in group (*b*) resembled fibrosis caused by mixed dust, as seen in slate workers. In most cases it was relatively easy histologically to separate the pulmonary lesions seen in group (*a*) from those of group (*b*).

Previous reports of the pathology of kaolin pneumoconiosis are sparse and describe few cases.[1 11-13 15] Most have tended to emphasise nodular rather than interstitial change, although occasional cases of progressive massive fibrosis have also been described.

Five group (*a*) cases showed pulmonary massive fibrosis but this did not correlate with lung dust concentrations, which ranged from 7.6 to 289.3 mg/g. This could be explained by clearance of dust from the lungs after retirement from the industry, sampling problems, or a secondary factor in causation.

This study supports previous epidemiological and radiological studies, which have indicated that the risk of developing kaolinite pneumoconiosis increases with the dryness of the material handled and cumu-

Table 5 *Analyses of variance (group (c), miscellaneous, excluded)*

	Analysis of variance for $\log_{10}$ (estimated quartz loading)					*Analysis of variance for $\log_{10}$ (estimated kaolinite loading)*			
	DF	*SS*	*MS*	*F ratio*	*p*	*DF*	*SS*	*MS*	*F ratio*
Nodular, interstitial ignored	3	8.18				3	1.17		
Interstitial, adjusted for nodular	2	0.56	0.28	1.20		2	0.47	0.24	0.65
Interstitial, nodular ignored	2	1.94				2	0.68		
Nodular, adjusted for interstitial	3	6.80	2.27	9.50	0.0	3	0.97	0.32	0.89
Interstitial and nodular	5	8.74				5	1.64		
Interstitial-nodular interaction	5	1.54	0.31	1.30		5	0.99	0.20	0.55
Residual	21	5.01	0.24			21	7.62	0.36	
Total	31	15.29				31	10.25		

Table 6 *Relationship between radiological and histological findings*

Profusion of small opacities	*Nodular fibrosis*		*Interstitial fibrosis*	
	0, 1	*2, 3*	*0, 1*	*2, 3*
0/– to 1/2	11	3	9	5
2/1 to 3/2	8	17	5	20
Significance of association	p = 0.01		p = 0.02	

lative years of exposure.[2 3 13 14] The major drawback of this study is that the group investigated was small and not representative; only a few of these workers were known in their lifetime to the Pneumoconiosis Panel. All the men were elderly and the findings reflect the much greater exposures of several decades ago. Another difficulty with this type of study is that occupational records for a person's exposure to various types of dust are often inadequate. We were encouraged to find that the occupational histories obtained in this study related well to the mineralogical findings.

Further clinical, pathological, and mineralogical studies should be carried out in cases of kaolin pneumoconiosis and should include a control group of people not employed in the china clay industry. This would provide the information necessary to evaluate the efficiency of the current precautions within the industry. It will take a further 10–20 years to accumulate such data. Meanwhile surveillance must depend on radiological examination.

We thank RA Comyns, WB Jepson, and PD Salt of English Clays Lovering and Pochin, who carried out the mineralogical analyses of all the specimens and who provided much of the information on occupational histories. We also thank Dr JC Gilson for assistance in reading the radiographs, DM Griffiths for preparing the figures and RM Mitha for preparing tissue and dust specimens.

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