Chapter 7. Fighting Silicosis

The dust that kills

Many readers will have visited Edinburgh and admired the city's fine sandstone buildings, and perhaps even thought about the dust exposure implications. Ken Donaldson, who has contributed a lot to the understanding of the pathology of occupational lung disease, and from that the causes and dose-response, extended his interests and wrote a paper on the epidemic of silicosis amongst the quarries and stone-masons who built Edinburgh New Town in the 18th and 19th centuries.[1] The buildings still look good, but the stone masons often showed signs of lung disease by the age of 40, and many died before they were 50 (Fig 7.1a).

In East Anglia, a completely different part of the country, for hundreds of years masons were working with flint, almost pure silica like the Edinburgh sandstone, and were skillfully breaking it to produce flat surfaces (Fig 7.1b). There are hundreds of medieval churches built with this material in Norfolk, and Norwich city walls alone could have had 25,000 square metres of worked flint facing. A Factory Inspectorate survey in the 1920s found that the few remaining knappers working flints to produce gun flints died on average 30 years younger than their wives,[2] and we will see in this article that their exposure may have been several times the current exposure limit. Like the Edinburgh New Town, Norfolk's much-admired buildings would have been deadly for many of their masons.

The previous chapter looked at the revolution in British occupational hygiene in the early 1900s, and all that happened to eventually eliminate lead poisoning as it was then defined. In this article we look at how the revolution worked out for silicosis, where it has not been quite as successful.

In Chapter 2 we saw that 200 years ago people suspected that the wasting diseases and early deaths in potteries, needle grinding, and stone working were all linked, and in the first two at least there were attempts to control the dust. Before the mid 1800s such wasting disease of the lungs was called consumption, or its Latin-derived equivalent phthisis. People who were not exposed to dust could get a very similar disease called phthisis pulmonalis, which might be accompanied by high temperature. From the 1840s, the term tuberculosis came into use, and in 1882, Robert Koch identified an infective bacterium as a cause. There followed several decades of confusion about whether the dust could cause the disease on its own, or whether silica inhalation might cause lung damage which predisposed to tuberculosis. Some employers overseas exploited the confusion, because it is easier and cheaper to treat disease than control dust adequately, and if dust is not the primary cause then the employers are relieved of compensation. As late as 1934, the ILO Convention on compensation for occupational disease clearly identified a dozen agents which led to compensable disease, but attributed silicosis not to silica, but to "Industries or processes recognised by national law or regulations as involving exposure to the risk of silicosis".[3,4]

Fig 7. 1. Two very different structures which would have exposed their masons to high-silica content dust. (a) The Old College, University of Edinburgh, built about 1820 of Craigleith sandstone, whose effects were recently described by Ken Donaldson and colleagues.[5] (b) A wall of an old house in Norwich, believed to be 14th Century, one of many East Anglian buildings made of shaped flints. The carefully-fitted blocks average about 8 cm square. [(a) LW Yang in Wikipedia, Creative Commons https://creativecommons.org/licenses/by/2.0/legalcode , (b) T Ogden]

In Britain, however, the cause had been clearly stated in 1915 by the Senior Medical Inspector Edgar Collis in the Milroy Lectures to the Royal College of Physicians.[6] He published an extensive review of British and other data and experience, and concluded that "A study of phthisis mortality rates in dusty occupations has, then, established the fact that when phthisis occurs in excess it is always found associated with exposure to dust containing crystalline silica." This provided a clear target for occupational hygiene control.

The start of respirable dust measurement

If people are getting ill and there is a cloud of dust which "rolls slowly about the rooms, till the atmosphere has become so fully impregnated that the men are scarcely visible", as in the grinding shops of Sheffield[7] and in the stonemason's sheds of Edinburgh,[8] then we do not need to wait for measurements before beginning control. But if conditions improve but dust cannot be completely eliminated at acceptable cost, then we need to target control at the components and exposures which cause disease. Dusts can differ in various characteristics, and in some workplaces are too fine to be visible in normal light. In the early 1900s the Factory Acts could require control of material "injurious to health", but it was a problem for inspectors in the early 1900s to know what characteristics and how much dust were "injurious".

In 1922, EL Middleton, like Collis a Medical Inspector of Factories, contributed a chapter to the Chief Inspector of Factories Report, on an "Investigation into the dust content of the atmosphere of work-places in the grinding of metals and cleaning of castings". In discussing what component of the dust should be targeted, he explained that silica particles in a diseased lung rarely exceed 2 µm in size. If larger particles are inhaled, they are caught in the upper airways are "discharged in the mucous secretions". However, some of the very small particles "are deposited in or near the alveoli, that is, beyond the action of the cilia which line all the bronchial tubes except those closest to the alveoli", so that they are not cleared. He then made an important statement fundamental to the measurements of dust causing silicosis and similar diseases: "The only measure of the injuriousness of an atmosphere containing an injurious insoluble dust is the number of particles, say, below 2µ in a given volume of air at breathing level." Middleton pointed out that a few large particles can outweigh thousands < 2µm, so a simple method of weighing the dust cannot measure risk from finer particles.[9]

The exact definitions have changed, but Middleton was arguing for the modern approach of measuring respirable dust. At that time, the only way of discriminating by particle size was to count the particles under a microscope, so Middleton looked for a method which would enable him to disregard the particles with maximum dimension >2 µm. For microscope counting, this meant taking only very brief samples, so the particles did not overlap. (Size-selective personal respirable-dust samplers, which permit longer sampling periods and the weighing of the samples, were not developed until the 1960s.) Without going into details, the shape and density of quartz particles means that Middleton's limit of 2 µm maximum dimension corresponds to an aerodynamic diameter of about 1.4 µm (see Appendix to this chapter). Modern gravimetric respirable samplers accept larger sizes than this, because they are designed to match the sizes which penetrate to the alveoli; Middleton was trying to measure the particles which not only reach the alveoli, but deposit and are retained.

Middleton's measurements

After considering alternatives, Middleton made his measurements with a device developed by J.S.Owens of the Meteorological Office.[10] This instrument, the Owens Jet (Fig 7.2), sucked the dusty air through a narrow orifice, impacting the particles onto a glass coverslip, which could be removed and the particles sized and counted. Owens observed that in foggy weather the moistened particles adhered much better to the coverslip. He therefore introduced a chamber in which the sampled air was humidified. Water condensed on the particles in the expansion of air through the jet, so they adhered, but the water evaporated before the particles were counted.

Middleton took samples at the breathing level of the worker, with the orifice facing in the direction the worker faced, and as close to his face as was consistent with his continuance at work. Middleton counted the particles with a microscope with an oil-immersion objective which enabled him to include particles down to about 0.1 µm, about the limit his impactor would have collected (see Appendix to this chapter).

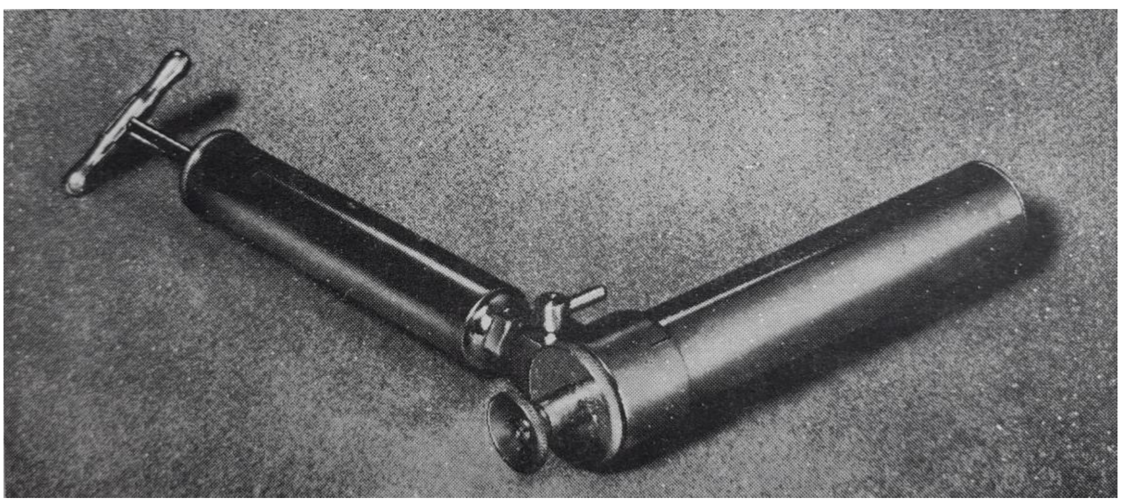

Fig 7.2. The commercial version of the Owens Jet sampler. The humidifying chamber, which was lined with wet blotting paper, is on the right, open to the air at the far end. The hand pump is on the left. The jet and coverslip are accessed using the knurled knob in the middle. (Photo courtesy of Casella UK)

Middleton first reported about 120 samples from several dozen grinding and cleaning processes in various parts of the country, recording in each case the number concentration and the percent >2 µm. For each sample, his report summarised other processes in the same room, and gave a description of the particles on the sample.[11] [12] Some years later, at a silicosis conference in Johannesburg in 1930, he presented a 95-page paper, with details of much more extensive surveys, including measurements and silicosis incidence in 13 industries. For three of them, potteries, and sandstone and granite quarrying and use, he gave detailed tables of results, totalling about 240 samples.[13]

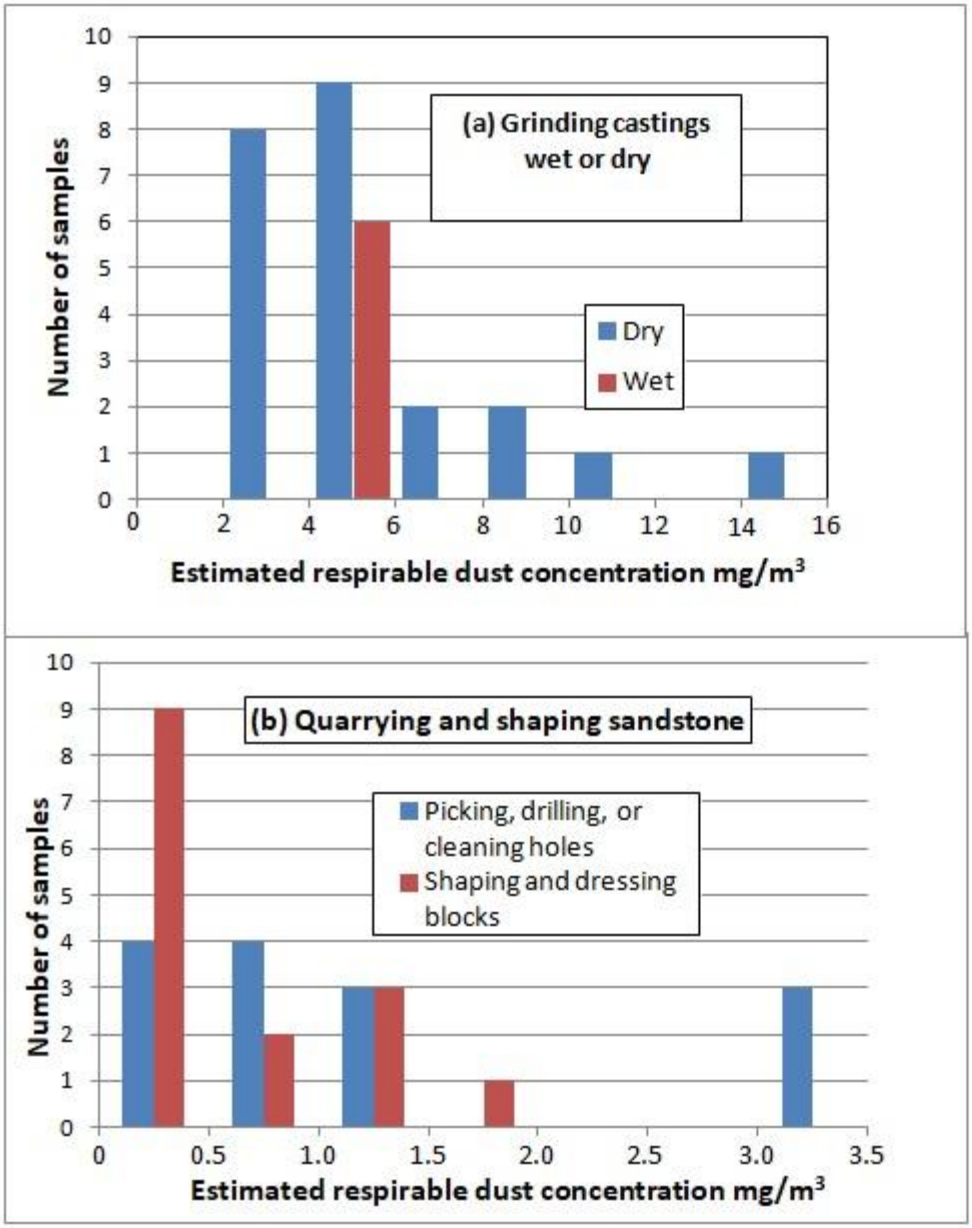

Fig.7.3. Rough estimates of gravimetric respirable concentrations from Middleton's particle counts. Uncertainties in the conversion process mean that the concentrations may be in error by a factor of two or more, and are more likely to be underestimates than overestimates. Samples where Middleton noted a preponderance of soot or other non-quartz particles are excluded.

It is possible to make a very rough estimate of the respirable gravimetric concentrations that correspond to Middleton's number counts, by assuming that the number of particles is lognormally distributed, as often found, and using what is known of the aerodynamic and shape properties of quartz particles. The

Appendix to this chapter gives details, and comes up with a very rough conversion factor of Middleton's count results to modern respirable dust concentrations. Some results are shown in Fig 7.3. The uncertainties mean that the gravimetric estimates could be in error by a factor of two or more, and are more likely to be on the low side than the high. Also, as already mentioned, these are very short term measurements of concentration.

Fig. 7.4. Table blade grinding, one of the many processes illustrated in the Factory Inspectorate's 1923 report. This process took place almost entirely in Sheffield, and "involved great physical effort", in a "cramped position…most trying to the body". Middleton's results suggest breathing zone respirable dust concentrations of roughly 3 to 6 mg/m^3 .Wear reduced the grindstone diameter by about 60cm in 4 to 6 weeks. Of grinders employed for more than 20 years, over 90% were reported to show signs of fibrosis. Their mortality rate attributed to "tuberculosis" was about 15 times the average rate for Sheffield. [Crown Copyright (expired)]

Despite the uncertainties, the results in Fig 7.3 are interesting. Concentrations using grindstones are much higher than in quarrying and shaping sandstone, which is not surprising. For comparison, the exposure limit applied to shift-length gravimetric samples in Britain is 0.1 mg/m^3, and in some countries is lower. We cannot get a realistic estimate of risk, because these are very short samples, and we do not know how long the operator did the task for, or how frequently. However, with this kind of concentration it is not surprising that some people got silicosis. Referring back to Donaldson and colleagues' study of Edinburgh New Town, four of Middleton's measurements in Edinburgh quarries equate to very roughly 1 mg/m^3, and the fifth, noted as being taken "sheltered from the wind", was roughly 7 mg/m^3. The two samples of flint knappers in East Anglia, mentioned above but not included in Fig 7.3, also gave concentrations of about 1 mg/m^3.

The grinding and cleaning of castings report

In 1923, the year after Middleton's chapter in the Chief Inspector's Annual Report, the Factory Inspectorate issued a major "Report on the Grinding of Metals and Cleaning of Castings with Special Reference to the Effects of Dust Inhalation upon the Workers".[14] This is a very impressive piece of work, about 50,000 words, with over 50 illustrations of a wide range of processes and control methods (eg, Fig 7.4). Part 1, by EL Macklin, an engineering inspector, surveyed the processes, and Part 2, by Middleton, reported medical examinations of 1153 workers, with their illnesses broken down by process.

The part of the report by Macklin devotes four pages to dust measurements. Despite his co-author's statement in the previous year's Chief Inspector's report that "mere mass is no measure of the injuriousness of dust", Macklin only reports "total" mass concentrations measured by Duckering's equipment (see Chapter 5), which are therefore quantitatively useless for assessing risk from silica. Middleton's 120 respirable results are in an Appendix without discussion in the text – we must go to the previous year's Annual Report for Middletons' review of these processes and risks and his conclusions on control in the light of the results. Apparently Middleton had failed to convince his engineering colleagues that respirable dust measurements were preferable, and the otherwise admirable report concentrated on the familiar, but misleading, gravimetric measurements!

However, if conditions are obviously bad, measurements are less important. The report has a lot of information and advice about control at particular processes and methods of work, local exhaust ventilation, and dust arrestment. Sand blasting should be enclosed and carried out under negative pressure (a suitable arrangement was illustrated). Exhaust ventilation "has been a fruitful field for the ignorant tradesman", and the report gives a lot of attention to good design of equipment and workplaces. Bag filters are commended. Cyclones can be quickly abraded by the dust. Chambers with baffles could be used, or the dusty air could be filtered through a layer of coke. Effective maintenance programmes were a problem as they are now. Substitution of sandstone wheels by those manufactured from emery or carborundum is favoured. Wet grinding is not necessarily an answer, because the water must be clean and applied at the right point, and it can give a false sense of assurance. Preparation of the wheels, which involves removing the excess stone at high points due to imperfect manufacture or mounting, is obviously a dusty process, but segregation was generally impracticable.

But there is still silicosis

There is a lot more to say about silicosis and other progressive lung diseases, but that will wait for the next chapter. But, unlike deadly lead poisoning discussed in Chapter 6, silicosis is not just history. Health and Safety Executive statistics show that in Britain there are 10 to 20 cases of silicosis a year. Donaldson and colleagues conclude their paper:

> Unfortunately silicosis still occurs among stonemasons in Scotland and of course elsewhere. Scotland … still sees the occurrence of early stage silicosis in stonemasons. A recent report of six cases of silicosis in individuals referred to specialist respiratory clinics with minimal respiratory symptoms found that all were stonemasons. According to the International Labour Organization, silicosis afflicts tens of millions of workers in hazardous occupations and kills thousands of people globally every year.

There is a strong risk that consumers are contributing to the "tens of millions" through choosing things like granite worktops and sandstone garden paving, which may have been quarried in a country where controls are fewer, and can be hazardous if they are cut. Unlike some industrial hazards, silica cannot be legislated away, because it is a natural material, and young men will continue to rot and die until everyone is suspicious of working with stony materials, or of buying them.

Appendix: Conversion of Middleton's number concentrations of quartz particles to respirable mass equivalents

Summary

As detailed above, in the 1920s, EL Middleton of the British Factories Inspectorate took hundreds of breathing-zone samples of silica in a wide range of industries, using an Owens Jet impactor, and counting and sizing the particles by oil-immersion microscopy, attempting to take account of the finding that only the finest particles are retained in the lung. For each sample he reported the total number concentration, and the percentage of particles with a maximum dimension > 2µm. Consideration of what is known of the shape and aerodynamic behaviour of quartz particles in particular, and fitting lognormal distributions to Middleton's counts, leads to the following formula.

$$G = \frac{N}{1000}(0.146\,P + 0..47) \quad mg/m^3$$

Where G is the respirable mass concentration by the definition in European Standard EN 481, N is the count obtained by Middleton in particles/cm^3, and P is the percentage of those particles with maximum dimension > 2 µm. There are major sources of uncertainty in this formula, the most important being the efficiency of the sampler: there are contradictory reports in the literature about this, and the sampler design may have been changed during its history. These problems mean that the formula may give results in error by a factor or two or three. Also, it must be born in mind that the sampling method gave only spot samples, not shift average exposures.

A1. Middleton's surveys

In the 1920s, EL Middleton, a Medical Inspector in the British Factories Inspectorate, made several hundred measurements of what we would now call respirable dust concentrations in a wide range of workplaces and industries using silica. He gave a justification for concentrating on particles < 2 µm particle diameter in the Chief Inspector of Factories Report for 1922, because these were the sizes predominantly found in lungs post mortem .[15] His results for metal grinding and cleaning of castings were published in a report in 1923,[16] and for other industries in a 95-page paper in the proceedings of the 1930 Johannesburg silicosis conference.[17] At the time, the only way available for discriminating by size was to size and count the particles by optical microscopy, and Middleton reported the number concentration, and the percentage of particles with a maximum diameter > 2 µm. This Appendix gives a method for obtaining a rough estimate of the corresponding gravimetric respirable mass concentrations.

A2 Sampling and microscopy efficiencies

A2.1 The Owens jet

Middleton chose the Owens Jet impactor [18] to take his samples, and was an early user of this instrument, saying that "an effort made to obtain a replica of Dr Owens' apparatus which was not then available was eventually successful".[19] It sounds from this that Middleton used a prototype rather than the version which soon became available from Casella Ltd (Fig 7.2). As explained below, this may be significant in considering the collection efficiency of the instrument.

Impactors were not then in wide use. Owens designed his to impact onto a glass microscope coverslip, and in using prototypes to study air pollution Owens observed that in foggy weather wet particles adhered better to the glass. He therefore made his instrument with a humidifying chamber so that water condensed on the particles in the expanding air downstream of the jet. They then adhered to the coverslip, and the water then evaporated before counting began.

Referring to Fig 7.2, the humidifying chamber was lined with wet blotting paper. Middleton took samples at the breathing level of the worker, with the orifice facing in the direction he faced, and as close to his face as was consistent with his continuance at work. The humidifying chamber was first filled with the air being studied by two or three strokes of the air pump. The knurled knob was then undone, a coverslip placed under the jet, and the knob replaced. 50 ml of air was then sucked through the jet with the pump.

In Owens' paper he describes making the jet in the form of a slot: "…the hole for the jet was produced by cementing the two halves of a cover-glass, the edges of which had been ground smooth, over a hole formed either in a brass disc or in another coverslip". As used by Middleton, the hole was apparently 10 mm in diameter and the two coverslip halves were 0.1 mm apart, so they formed a rectangular slot 10 mm long[20] and 0.1 mm wide. From the duration of the stroke of the airpump, Owens estimated the velocity through the jet as about 250 m/sec (compare the velocity of sound, 330 m/sec).

A2.2 Impaction and visibility size limit

Below a certain size, particles do not impact efficiently, but are carried away in the air. The efficiency of impaction has been extensively studied since Owens' time, and from the formulae given by Hinds,[21] the impaction efficiency of the Owens jet would be expected to fall below 50% for quartz particles less than about 0.15 µm diameter. The Reynolds number of the slot would be in the range recommended by Hines, but the jet to coverslip distance would be about 10 times the slot width, compared to a recommended ratio of 1.5 to 5. This would be expected to reduce the impact efficiency.

To count the particles, Middleton used a 1.9 mm oil immersion objective[22]. This would be expected to have a resolution of about 0.2 µm.[23] Particles below this size could be seen, but could not be sized: as the particles get smaller below this, the images appear to stay the same size, but become fainter.

It seems from this that particles down to about 0.1 or 0.2 µm diameter could be collected and counted.

A2.3 Overall collection efficiency

Large particles can fracture on impact, or get swept away. Middleton's results in grinding and cleaning especially have comments such as "a considerable proportion of quartz 1 to 6 µ" and from "under 1 µ to 15 µ" with mentions of clumps, which indicates that the impactor is collecting larger particles. (The conventional abbreviation for micrometres was then µ, now µm.) Fuchs quotes some Russian work from 1933 which found that the maximum diameter of carborundum particles collected by an Owens instrument with moistening was 7.4 µm, corresponding to an aerodynamic diameter of perhaps 10 to 12 µm, depending on the particle shape.[24] In his original work on his instrument, Owens put two impaction stages in series and used the results to calculate jet efficiencies ranging from 53 to 83% in different trials, but these were without humidification, so should be underestimate.

However, in 1935 Green and Watson published measurements of the efficiency of the Owens sampler. They generated clouds of fine flint dust in a chamber and compared the results of the sampler with those obtained with a sedimentation cell. They found that efficiency decreased quickly as particle size increased, falling below 25% for particle diameters above 1.5 µm.[25] For their test cloud, the overall efficiency was about 42%. A similar size-dependence was obtained by CN Davies and colleagues by comparing the Owens sampler results with a thermal precipitator with coal dust in about 1950.[26] Davies and colleagues attributed these low efficiencies to shattering of the particles because of the high impact velocities, in the accelerating airstream and on the jet walls, as well as on the impaction stage.

It is difficult to reconcile these later results with the earlier ones of Owens, and with Middleton's observations of larger particles in his samples. It raises the question of whether the shape of the impaction jet had been changed between the 1920s and the later tests. As quoted above (section A2.1), Owens described forming the slot with the ground edges of two halves of a cover slip, and then later as "a diaphragm of hard brass or copper sheet in two half-discs, the straight edges of which form between them the slot." The natural interpretation of these descriptions is of a straight slot with sides parallel in both dimensions. However, the profile of the slot illustrated in Davies' paper and Fuchs' book is sharply curved, narrowest as the air exited the jet. It would have been very difficult for Owens to grind the edges of a glass coverslip to this shape. There have been cases in the past where a manufacturer changed the form of an aerosol instrument to ease production problems, and perhaps this was done with or without Owens' consent, and without realising the possible effects.

If the slot design was changed, it seems that Middleton's results which he published in 1923 were with the original form, but we cannot be sure whether he continued to use this for the results published in 1930. Even if he did not go back to the same process, it is hard to believe that Middleton would not have noticed a threefold reduction in the density of his samples in similar environments. However, we cannot be sure, and the best option seems to be to assume an efficiency in the middle of the range and accept the uncertainty. I have therefore assumed it to be 50%, and accept that the final calculated concentrations may be in error by a factor of two from this cause. The supposed 50% efficiency has been taken into account by multiplying the counts by two as part of the calculations.

A3 Converting to respirable gravimetric concentrations

A3.1 Particle size distributions

In order to work out the mass of the particles which Middleton's counts correspond to, we need to make assumptions about size and shape, because all Middleton tells us is the total number and the percent > 2 µm maximum dimension. For example, if we assume that the particles are all quartz spheres 1 µm in diameter, then 1000 particles/cm^3 (at 50% collection efficiency) would correspond to 2.72 mg/m^3. We can however refine this assumption and estimate. (Here and in the rest of this Appendix I have assume a quartz density of 2.6 g/cm^3 (2.6 kg/dm^3).)

In the past, various particle size distributions have been found or assumed in particular situations, but the lognormal distribution "has been found to apply to most single-source aerosols".[27] Dust particles are often irregular in shape, to a greater or lesser degree, but provided the shape does not vary with size, then any measure of size should be lognormally distributed. A common measure when particles are examined under the microscope is the equal-area diameter d_p, defined as the diameter of a circle with area equal to that of the particle as seen in the microscope. We will work with this measure, and assume that it is lognormally distributed.

Even if the size distribution is not exactly lognormal, then experience shows that it is likely to be positively skewed, so the assumption will give roughly-right results.

A3.2 The shape and volume of quartz particles

Quartz particles are not spherical, but tend to be flattened, as are larger fragments of flint, and Cartwright[28] found that aerodynamically and in some other respects they behave like oblate spheroids, ie spheres flattened at the poles.

Their cross-section through the poles is then an ellipse, with an equatorial diameter x and a polar diameter z. The volume of a oblate spheroid is $\pi x^2 z/6$; a sphere has a volume $\pi d^3/6$, so the diameter of a sphere equal in volume to the spheroid is

$$d_v = (x^2 z)^{1/3} \qquad (1)$$

Cartwright also found that quartz particles behave generally as if they had an axial ratio x:z of 2.5:1, so the spheroid volume is $0.4\pi x^3/6$, or 0.4 of the volume of sphere with diameter equal to the equatorial diameter x of the spheroid. If the spheroid settled flat, with its equatorial diameter horizontal, then its

equal area diameter d_p would be equal to its equatorial diameter x. However, small particles may settle randomly oriented, and Cartwright showed that in this case on average
$d_p = 0.8x$, or $x = d_p/0.8$.

A3.3 Mass concentrations

To convert to mass concentrations, we need to calculate the probable mass of a particle from its d_p , which we can measure under the microscope, and we do this by assuming that the quartz particles are oblate spheroids with an axial ratio of 0.4, and that they are randomly oriented on the slide.

Volume of the spheroid $= 0.4\pi x^3/6$
and $x^3 = (d_p/0.8)^3$
so volume $= (0.4/0.8^3)\,\pi\, d_p^3/6 = 0.78\,\pi\, d_p^3/6$ approx. (2)
We have therefore assumed the volume of the quartz particle to be 0.78 times the volume of a sphere with a diameter equal to the equal-area diameter of the particle. The mass of the particle will be this multiplied by the density of quartz, which we assume to be 2.6 g/cm^3. If there are $n(d_p)$ particles of this size present, then their contribution to the total mass will be about

$$(0.78\, n(d_p)\, \pi\, d_p^3/6) \times 2.6 \text{ grams} \qquad (3)$$

if d_p is in cm.

A3.4 Respirable mass concentrations

Finally, to calculate the contribution of these particles to the respirable mass concentration we must mutiply each $n(d_p)$, and therefore each mass given by (3), by the respirable mass factor for d_p given in EN 481, which in µm is given by a culmulative lognormal distribution with a median diameter 4.25 µm and a GSD of 1.5.[29] This calculation is conveniently done in a spreadsheet. (We have assumed that d_p is equal to the aerodynamic diameter of the particle, which experimentally is found to be approximately true.[30 31]) As already mentioned, it is necessary to multiply the results by two, to allow for the assumed sampler efficiency of 50%.

A3.5 Summary

We can use a spreadsheet function to calculate, for a chosen lognormal size distribution (Section A3.1), the fraction $f(d_p)$ of the total number of particles with diameter d_p . If we multiply this fraction by the total number of particles present, we obtain $n(d_p)$. For example, if the overall number concentration is 1000 particles/cm^3, then $n(d_p) = 1000\, f(d_p)$ particles/cm^3.

We can then calculate the mass concentration contributed by the number $n(d_p)$ of particles with a projected area diameter d_p, using expression (3) in Section 3.3, and then adjust that to the contribution of those particles to the respirable mass concentration by multiplying by the respirable mass factor at that size (Section A3.4), and also by a factor 2 to allow for the assumed 50% effiency of the collector. The total respirable mass concentration of the whole aerosol is then the sum of this over all particle sizes.

The next section looks at the best lognormal distribution to choose for a particular sample.

A4. Fitting a distribution

Fig A1 shows a lognormal distribution that might be fitted to one of Middleton's measurements. This distribution has M = 1 µm and GSD = 1.5, and calculating on a spreadsheet shows that it has 4.37% of particles > 2 µm. It might at first sight therefore to be a candidate for fitting to any sample that has that percent above that size. This distribution could then be used to calculate the respirable mass distribution for that sample, using the procedure in Section A3. Unfortunately, the distribution is not uniquely specified by the percent above 2 µm – distributions with smaller M and larger GSD (or larger M and smaller GSD) could give the same percentage. To what extent does this affect the calculated respirable gravimetric concentration?

Before answering that we have to deal with another unknown. We are working with the projected area diameter d_p , but Middleton specified the percent greater than 2 µm maximum dimension. We therefore need to know how the maximum dimension of quartz particles relates to d_p. The overall modelling of the aerodynamic behaviour of quartz particles as prolate spheroids is too approximate for this purpose. Unfortunately, maximum dimension of quartz particles does not seem to be a variable that has been investigated in the literature. However, Davies' 1979 paper on particle-fluid interaction[32] includes two electron micrographs of particles, one by Kotrappa of 41 particles,[33] and another by Cartwright of three particles.[34] (A few silhouettes which could have been overlapping pairs were omitted from the following analysis.) The maximum dimension and d_p for each of these 44 particles was measured, and the distribution of the ratio is shown in Fig A2.

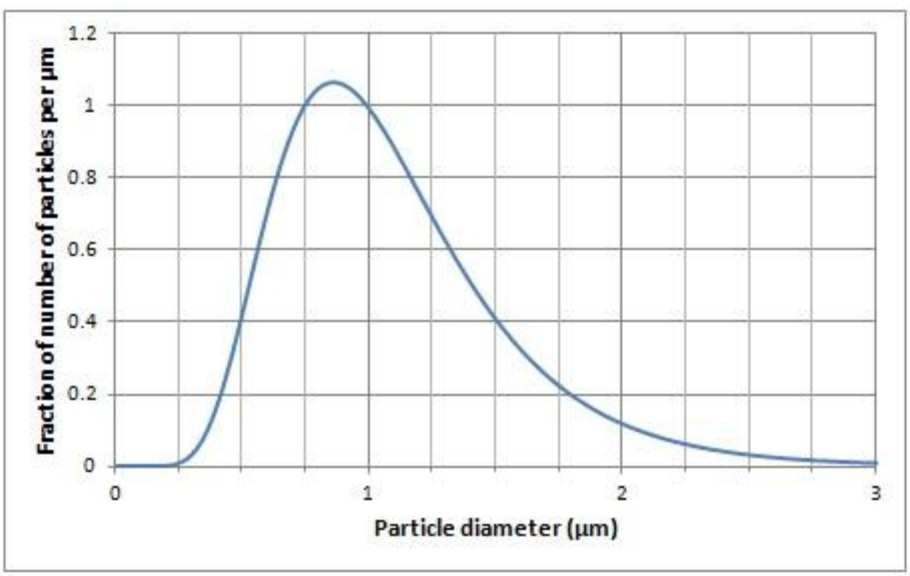

Fig A1. A lognormal particle-size distribution, in this case with M = 1 µm and GSD = 1.5. 4.37% of the particles in this distribution have diameter > 2 µm

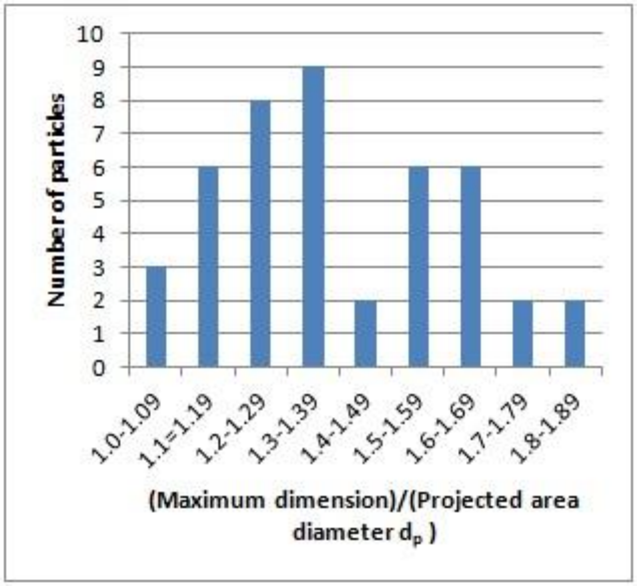

Fig A2. Ratio of maximum dimension to d_p for the 44 respirable quartz particles illustrated by Davies.[35]

Clearly there is plenty of variation from particle to particle, so this gives another area of uncertainty. The mean of the ratios shown in Fig A2 is 1.39, so the d_p corresponding to a maximum diameter of 2 µm is 2/1.39 = 1.44 µm on average. In interpreting the modelled distributions of d_p, therefore, we will assume that Middleton's percentages above 2 µm equate to percents for d_p > 1.44 µm .

We can now return to the distributions and see what range of possible lognormal distributions (ie, ranges of M and GSD) give the percentages > 2 µm (max dimension) observed by Middleton, and what respirable mass concentrations these correspond to. After initial trials, the percentages were calculated for M between 0.2 µm and 1.0 µm, and GSD between 1.5 and 3, and for each distribution the respirable mass concentration corresponding to 1000 particles/cm^3 was calculated. The procedure is illustrated in the section of a Microsoft Excel 2007 spreadsheet in Fig A3.

At B1 and B2 are the values of M and GSD respectively

Column A shows d_p in µm

B gives the culmulative lognormal distribution, by using the LOGNORMDIST function

C gives the differential distribution by subtracting successive values of cells in col B. Each cell in this column therefore gives the fraction of the total number of particles at that d_p

D gives the respirable factor for each d_p, calculated as explained in Section A3.4

E applies D to C to get the respirable fraction at that size

F multiplies E by 10^9, to give the number of respirable particles at d_p in one cubic metre if the total concentration is 1000 particles/cm^3

G uses expression (2) above to calculate the respirable mass of a particle at that d_p

H multiplies F and G and adjusts units to get the respirable mass concentration for that d_p in mg/m^3 .

The spreadsheet was run up to d_p = 3 µm: higher values made no significant contribution. H was then summed to give the total respirable concentration. It was assumed that Middleton did not include particles with $d_p < 0.1$ µm (see Section A2.2 above), so the fraction (and hence the percent) of particle numbers for $d_p > 1.44$ µm for comparison with Middleton's percent figure was obtained by subtracting the column B value at $d_p = 1.44$ µm from 1.0, and expressing this as a percent of the number > 0.1 µm.

	A	B	C	D	E	F	G	H
1		0.6	(M)					
2		1.5	(GSD)					
3	Particle equal-area diam dp	Cumulative lognormal distribution	fraction of total number of particles	Respirable fraction at dp from EN481	Col C x Col D	Number of ptls if 10^9 total	Mass per particle (g)	Contribution to the mass of the 10^9 (mg/m3)
4	0.02	2.46433E-17	2.46433E-17	0.9994004	2.46E-17	2.46E-08	1.69811E-17	4.1822E-22
5	0.04	1.20393E-11	1.20393E-11	0.9988014	1.2E-11	0.012025	1.35849E-16	1.6336E-15
6	0.06	6.77923E-09	6.76719E-09	0.9982032	6.76E-09	6.755033	4.5849E-16	3.0971E-12
7	0.08	3.35867E-07	3.29088E-07	0.9976058	3.28E-07	328.3003	1.08679E-15	3.5679E-10
8	0.1	4.95741E-06	4.62155E-06	0.997009	4.61E-06	4607.724	2.12264E-15	9.7805E-09
9	0.12	3.60326E-05	3.10752E-05	0.9964129	3.1E-05	30963.74	3.66792E-15	1.1357E-07
10	0.14	0.00016586	0.000129827	0.9958176	0.000129	129284.3	5.82452E-15	7.5302E-07
11	0.16	0.000557354	0.000391494	0.995223	0.00039	389623.7	8.69433E-15	3.3875E-06
12	0.18	0.001492093	0.000934739	0.9946291	0.00093	929718.4	1.23792E-14	1.1509E-05
13	0.2	0.003369121	0.001877028	0.9940359	0.001866	1865833	1.69811E-14	3.1684E-05
14	0.22	0.006672122	0.003303002	0.9934434	0.003281	3281345	2.26019E-14	7.4165E-05
15	0.24	0.01191525	0.005243128	0.9928516	0.005206	5205648	2.93434E-14	0.00015275
16	0.26	0.019582866	0.007667616	0.9922605	0.007608	7608273	3.73075E-14	0.00028385
17	0.28	0.030076625	0.01049376	0.9916702	0.010406	10406348	4.65962E-14	0.0004849
18	0.3	0.043678141	0.013601516	0.9910805	0.01348	13480197	5.73113E-14	0.00077257
19	0.32	0.06053	0.016851859	0.9904916	0.016692	16691624	6.95547E-14	0.00116098
20	0.34	0.080633804	0.020103804	0.9899033	0.019901	19900822	8.34282E-14	0.00166029
21	0.36	0.103861558	0.023227754	0.9893158	0.02298	22979584	9.90339E-14	0.00227576
22	0.38	0.129975949	0.026114391	0.988729	0.02582	25820055	1.16474E-13	0.00300735
23	0.4	0.158655254	0.028679305	0.9881429	0.028339	28339250	1.35849E-13	0.00384986

Fig A3. Example of spreadsheet calculation

As shown in Fig A4, the respirable mass concentration corresponding to 1000 particles/cm^3 correlates well with the percent of particles with maximum dimension > 2 µm, at least for percents > 1 . The assumed M and GSD have some effect on the relationship, but as we do not have any other grounds to choose the values, we have to accept this as another uncertainty in the conversion. The fact that M and GSD have limited effect indicates that modest departure from a lognormal distribution would not have too much effect either: other positively skew distributions are likely to give similar relationships between percent and gravimetric concentration.

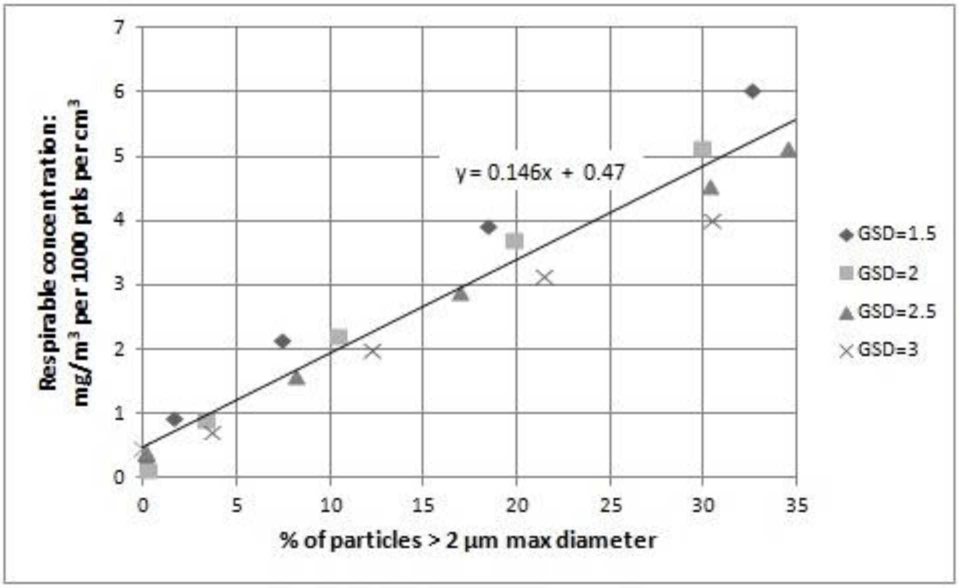

Fig A4. Relationship of the respirable mass concentration and the percent of particles > 2µm maximum diameter, for lognormal particle number distributions with combinations of M between 0.2 and 1 µm, and GSD between 1.5 and 3.

A5. Implications

Fig A4 shows that for a given number concentration the respirable mass concentration increases linearly with the percentage of particles with maximum dimension > 2 µm. At first sight, this contradicts Middleton's assumption – he recorded the percentage because he thought that particles > 2 µm were less dangerous. The reason is that the modern respirable convention includes a high percentage of these particles, and of course the mass contributed by each is proportional to the cube of its diameter. The underlying explanation for this difference is that the respirable convention aims to simulate the fraction *reaching* the alveoli, whereas Middleton aimed to measure those *retained* there. For comparison with modern standards, we must use the respirable convention, whether or not Middleton was right in terms of risk of silicosis.

The trend line for a number concentration of 1000 particles/cm^3 is shown on Fig A4. Our best estimate of the respirable mass concentration as defined by EN 481 corresponding to Middleton's number counts is therefore

$$G = \frac{N}{1000}(0.146\,P + 0.47) \quad mg/m^3 \qquad (4)$$

where N is the number of particles/cm^3 recorded by Middleton, and P is the percent of these that he reported > 2 µm maximum dimension.

A6. Uncertainties

A number of sources of uncertainty have been noted in the course of this report, and these will now be summarised. They must be considered in interpreting results obtained with equation (4) above.

(1) (Section A2.1) Middleton's samples took only a few seconds, so each gives the concentration at only one point in time. Modern gravimetric samples and standards are exposure averaged over some hours.

(2) (Section A2.3) The largest potential source of uncertainty is the overall efficiency of the Owens Jet impactor. The original studies gave high results. However, Green and Watson and Davies and colleagues later measured much lower efficiencies, possibly because the slot design had changed. Our conversion calculations therefore assume an efficiency in between these, at 50%, but it must be recognised that this introduces an uncertainly of a factor of about two. If Green and Watson's efficiencies are really applicable to Middleton's results, then the concentrations will be substantially higher than those calculated from (4).

(3) (Section A3.1) We have assumed that the size of the particles is lognormally distributed. Also (Section A4) there is uncertainty on the fitting because we rely on one data point per distribution: the percentage above a maximum dimension of 2 µm. However, it seems from Fig A4 that provided the distribution does not depart too far from lognormal, the uncertainty of the respirable concentration is only about 20% from this cause.

(4) (Section A3.2) We have assumed that for the purposes of calculating volume from d_p the particles can be treated as oblate spheroids with an axial ratio of 2.5:1, and that they lie randomly oriented on the slide, and (Section A3.4) that the aerodynamic diameter is equal to d_p. This is the best information we have for the behaviour of quartz particles on average, but is another source of uncertainly. Also, these approximations are based on studies on pure quartz particles, but some of the dusts measured by Middleton were not. However, the outcome is that we have assumed that the volume of the particles is 78% of the volume they would have if the particles were spheres, so if that were spheres we would have underestimated the respirable mass by about 30%.

(5) (Section A4) The ratio of maximum dimension to d_p was based on measurements of a relatively small number of quartz particles (Fig A2). The particle-to-particle variation introduces some uncertainty in the mean value, and we do not know how well it will apply to other minerals which contain silica.

To a certain extent, the random particle-to-particle component of uncertainties (4) and (5) will be smoothed out if a large number of particles contribute, which will usually be the case. However, the uncertainties introduced by (2) and (4) mean that the concentrations could wrong by a factor of two or three, and more likely to err on the low side than the high. Also, as most of the uncertainty was a fixed bias, the relative values at different processes should be valid.

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