Measurement of human exposure to biologically relevant fractions of inhaled aerosols

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Abstract

Aerosol sampling has evolved with changes in technology and our understanding of the importance of assessing the biologically relevant fractions of the total aerosol. During the past decade there has been international agreement on the definitions for the inhalable, thoracic, and respirable fractions and instruments have been developed to collect samples according to these conventions. These measurement techniques are now well established in the workplace and are increasingly being applied to assessments of non-occupational exposure, as the practical difficulties in obtaining samples over 24 hours have been solved. It is argued that multiple aerosol size fractions should be measured for hazardous substances—such as inorganic lead—in which inhaled material may either be absorbed in the alveoli (respirable) or cleared from the ciliated airways in the lung to the gut and then absorbed (thoracic and extrathoracic). Such measurements should improve the evaluation of the risk for inhaled lead, particularly for non-occupational exposure of children. Also, passive aerosol sampling techniques may enable measurements of non-occupational exposure to be made over several weeks and this would also help improve the reliability of the risk evaluation for aerosol toxicants with a long biological half life. However, further development of these techniques is required if they are to be practicable and biologically relevant.

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It has long been recognised that the presence of particles in the air may adversely affect the health of exposed people. However, it is only in the past 100 years that serious attempts have been made to measure the size, shape, and concentration of airborne particles and to then relate these measures to human health. Inevitably, almost all of the early work was involved with measurement of aerosols in occupational settings such as mining, as it was in these situations that effects on health were most obvious.

Most of the early aerosol monitoring devices were designed to be placed in one place while the sample was collected. However, the advent of small and light battery operated pumps in the 1960s meant that personal exposure measurements became possible. Early personal samplers comprised simple open faced filter

holders that were thought to collect "total dust". More complex devices, such as cyclones designed to collect only the finer fractions of airborne dust, became available only later. However, these improvements in technology enabled the links to be identified between occupational exposure to a range of aerosols and lung disease.

Developments in monitoring of exposure to aerosols have generally been focused towards sampling of occupational exposure, although the principles are equally applicable to non-occupational exposures. In the past, limitations on analytical sensitivity have dictated that environmental sampling of aerosols should be undertaken with high volume fixed location monitoring equipment. This is no longer the case and personal exposure measurements in non-occupational situations should help improve risk assessments.

In this paper we briefly review some of the recent developments in sampling methods used to assess human exposure to aerosols, and in particular, we look at issues of transferring experience from workplace sampling to non-occupational exposure monitoring. We use non-occupational exposure to inorganic lead aerosol as an example of the issues involved in developing an appropriate measurement strategy.

Early developments in aerosol sampling

Up to the mid-1920s the principal method of sampling aerosols was the sugar tube, later described by Walton.¹ This device comprised a 32 mm diameter tube filled to a depth of about 100 mm with 10–20 mesh sugar granules. Air was drawn through the granules by a hand operated pump and the collected dust was analysed by dissolving the sugar, and filtering the residue, which was then weighed to provide an estimate of the airborne mass. As well as being difficult to use, this method was criticised because reductions in the measured dust concentration in mines and other dusty environments were not matched by corresponding falls in dust related diseases.

More complex instruments soon became available. One such device was the konimeter, which was developed in 1916 by Kotze.² In this device, a small sample of air was drawn through a jet nozzle and directed towards a plate covered with petroleum jelly where the particles deposited by impaction. The plate was removed and the particles counted with a microscope. The operating principles of this device recognised that both the particle numbers and the particle size could be important determinants of risk. However, it was only able to collect a sample over a very short duration (about 1 second).

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By the mid-1930s there were several alternatives to the konimeter, the most important being the thermal precipitator.³ This overcame the limitations of the konimeter to collect very small particles (<1 µm) and samples could be collected over much longer periods. The thermal precipitator had an inlet leading to a narrow channel where there was a heated wire oriented perpendicular to the direction of flow. As the contaminated air passed the wire, thermal gradients caused the particles to be deflected towards the walls of the sampler where two glass disks were located. After sampling, the glass disks were removed and the particles counted as with the konimeter. The thermal precipitator was a heavy bulky instrument that could only be used for fixed location measurements.

Partly because of ease and reliability of analysis, gravimetric sampling with filters to collect the dust from the air largely superseded particle counting methods. An early example of such an instrument is the Mine Research Establishment (MRE) type 113A gravimetric sampler⁴ which has been used in the mining industry in the United Kingdom to collect the British Medical Research Council (BMRC) respirable dust fraction.⁵ The respirable fraction was defined qualitatively as the fraction of the airborne dust that approximates to that which would reach and deposit in the alveoli, reflecting the belief that these particles were of greater importance in causing occupational lung disease, especially pneumoconiosis. With this device a close relation was established between the prevalence of pneumoconiosis and the mass concentration of respirable coal mine dust,⁶ whereas earlier attempts to correlate disease with the number of particles of dust had been less successful.⁷ The development of the BMRC respirable fraction was the first of what have become known as the “biologically relevant” aerosol size fractions that now form the basis of modern sampling methods.

Sampling of biologically relevant and other aerosol size fractions

Over the past 10 years substantial progress has been made towards standardising the sampling of aerosols to ensure that the fractions sampled are biologically relevant. In 1993 a new European standard for the definition of size fractions for measuring airborne particles was approved by the European Committee for Standardisation.⁸ This standard defines three sampling conventions: inhalable fraction (the mass fraction of airborne particles which is inhaled into the nose or mouth); thoracic fraction (the mass fraction of inhaled particles penetrating beyond the larynx) and respirable fraction (the mass fraction of inhaled particles penetrating to the unciliated airways). These three criteria are shown in figure 1. The same definitions, for workplace atmospheres, have also been adopted by the International Standards Organisation⁹ and The American Conference of Governmental Industrial Hygienists (ACGIH).¹⁰

These conventions recognise that only part of the total aerosol is inhaled into the nose or

mouth (the inhalable fraction). This concept was based on measurements of the aspiration efficiency of life sized human models reported by Ogden and his co-workers^{11,12} and subsequently by Vincent and his colleagues.¹³⁻¹⁵ For large particles (>50 µm, aerodynamic diameter) only about 50% of the total aerosol enters the respiratory tract according to the inhalable criteria, and this increases to almost 100% for small (~1 µm diameter) particles. The thoracic and respirable conventions are subfractions of the inhalable aerosol and have their basis in a wide range of human volunteer deposition and clearance studies.¹⁶ However, the final forms of the definitions were a compromise in the case of the respirable convention between the BMRC curve and the previous ACGIH definition.¹⁷

The development of size conventions for measurement of ambient aerosol has been through a separate process to that of the workplace definitions and has been driven primarily by the deliberations of the United States Environmental Protection Agency. Greater emphasis in these considerations has been placed on the size distribution of aerosol in the ambient air but potential penetration into the human respiratory tract was also considered. The two commonly used environmental sampling conventions are PM₁₀ and PM_{2.5}, both of which are curves falling from unity to zero with, in the case of PM₁₀, a 50% (0.5) value at 10 µm and, for PM_{2.5}, a 0.5 value at 2.5 µm. Although not specifically designed to be so, the thoracic convention is almost identical to the PM₁₀ size selection curve. There is, however, a significant difference between the respirable convention and the PM_{2.5} curve.

It is now considered that personal samplers are much more effective tools for assessing exposure to aerosols in the workplace than static samplers. The types and performance of these devices have been described by other authors.¹⁸ In the main, these samplers have been targeted to meet either the inhalable convention—for example, the Institute of Occupational Medicine personal inhalable sampler¹⁹—or the respirable convention—for example, the Higgins cyclone.²⁰ As yet there are no commercial samplers for the thoracic convention although several research devices have or are being developed.^{21,22} The clear rationale is that sampling personal exposure provides a better evaluation of potential risks, as it has also been shown that personal

Figure 1 Inhalable, thoracic, and respirable sampling criteria.

exposures are almost invariably higher than those found with simultaneously operating fixed location monitors.²³

Measurement of exposure in non-occupational groups

Procedures for monitoring personal exposure to airborne particles in non-occupational situations have been adapted from those used in the workplace. The equipment generally comprises a small battery operated pump connected to a size selective sampling head containing a filter paper or some other particle collection system. The main problems associated with these techniques arise because of the low airborne mass concentrations in non-occupational situations and the necessity to sample over relatively long periods, typically 24 hours or more. As the person should wear the pump throughout the sampling period, except during sleep, it must be lightweight and quiet and this limits the maximum airflow. As a consequence, the minimum detectable mass concentration is generally about 10 µg/m³ over 24 hours.²⁴ The detection limit for components of the aerosol, such as lead, will be determined by the sensitivity of the analytical techniques used, but could be much less than for gravimetric assessments, perhaps about 10 ng/m³ for lead.²⁵

During personal sampling the entry into the sampling head is normally located close to the person's nose and mouth, typically on the outside of their clothing on a lapel to give an estimate of the concentration of particles inhaled. During the hours of sleep the sampler is generally placed in the individual's bedroom. To ensure that the noise of the sampler does not disturb the person during sleep some investigators have devised sound insulated boxes to muffle the pump noise.²⁴ These systems may also be used at other times when the noise would be distracting—for example, while watching television or reading. It is necessary to replace the pump battery about every 8 hours or to use mains electricity overnight.

There are several sampling heads that have been used to measure personal exposure to airborne particles. Measurements made more than 15 years ago may have been made as total suspended particulate (TSP), although this is a misnomer and these instruments generally had an undefined sampling characteristic which would have included some, but not all, of the larger particles suspended in the air. More recently measurements have been made with

the size selective sampling criteria described earlier. Even with such systems there is still the possibility that wearing a noisy pump may alter the person's behaviour and bias the measured exposure level.^{26–27}

There are a few studies, mostly carried out over the past 10 years, which provide reliable measurements of personal exposure to airborne particles. These studies, which have been carried out in North America and more recently in Europe, do not generally measure the metal content of the aerosol. Most of the data refer to urban or suburban populations, generally excluding workers in occupations where there is known exposure to aerosols, and most measurements have been made over 24 hours. In total, there are about 2000 published measurements from various subgroups within society and only about 1200 measurements made in relation to currently accepted biologically relevant sampling conventions—such as thoracic aerosol (table).

In these studies the mean thoracic aerosol concentration ranged from 42 µg/m³ for the study of Seaton *et al*²⁸ of elderly volunteers in Belfast to 105 µg/m³ for children in The Netherlands.²⁹ Most of these measurements were made over 24 hour periods and where this was not the case (Birmingham, UK) the data have been adjusted to estimate 24 hour exposure by using the average fixed point monitoring data to represent the unmeasured time. Other studies have shown that during the night indoor, outdoor, and personal concentrations agree quite well and so this adjustment seems justifiable.³⁰ The maximum individual thoracic concentration was 971 µg/m³ (24 hour average), measured by Loiy *et al* in Phillipsburg, NJ.³¹

Sampling human exposure to lead aerosol

Sampling of lead aerosol in occupational situations is one of the few cases where current sampling practice does not require biologically relevant size selective sampling, although the method has been standardised. There is no agreement about how lead should be sampled outside the workplace.

There is also very little information about the size range of lead aerosols in ambient air in the United Kingdom. From the Quality of Urban Air Review Group (QUARG) report on airborne particulate matter in the United Kingdom³² about 90% of the lead aerosol is in the fine fraction (aerodynamic diameter <2.5

Measurements of thoracic aerosol exposure in various groups of non-occupationally exposed subjects over 24 hours

Study	Location	Study group	Subjects (n)	Measurements (n)	Geometric mean exposure (µg/m ³)	Maximum exposure (µg/m ³)
Loiy <i>et al</i> ³¹	Phillipsburg, USA	Non-smoking volunteers	14	189	66	971
Pellizzari <i>et al</i> ³⁰	Riverside, USA	Representative selection of non-smokers	178	171	98*	285†
Janssen <i>et al</i> ²⁹	Amsterdam and Wageningen, Netherlands	Children aged 10–12 y	45	301	105‡	195
Mark <i>et al</i> ⁴²	Central Birmingham, UK	Volunteers, smokers, and non-smokers	22	127	70§	260
Mark <i>et al</i> ⁴²	East Birmingham, UK	Volunteers, smokers, and non-smokers	15	178	47§	120
Janssen <i>et al</i> ³³	Amsterdam, Netherlands	Non-smoking volunteers aged 50–70 y	37	262	59‡	113
Seaton <i>et al</i> ²⁸	Edinburgh, UK	Volunteers aged >60 y	57	57	57	418
Seaton <i>et al</i> ²⁸	Belfast, UK	Volunteers aged > 60 y	52	54	42	288

*Population weighted median exposure rather than geometric mean.

†Maximum is the estimated 98th percentile.

‡Geometric mean estimated from arithmetic mean (SD).

§Estimated geometric mean and maximum, see text for explanation.

μm), probably associated with particles $<1\mu\text{m}$ in diameter.³³ It is possible that there may be greater concentrations of lead in the coarse fraction when lead contaminated dust, which is attached to other settled dust in the environment, is then resuspended. Because of the bonding of particles to surfaces, most dust which is resuspended will have a diameter $>1\text{--}5\mu\text{m}$.³⁴ Measurements of lead in household dust suggest that there may be substantial amounts of lead associated with larger particles.³⁵

When considering which size fraction or fractions are most appropriate for sampling human exposure to lead aerosol, we should consider the fate of the different size particles. Ideally, the measurement system should be capable of collecting the inhalable and respirable fractions. For lead particles which deposit in the alveolar region there is almost 100% absorption. However, only about 10% of the lead that is swallowed by adults is absorbed into the blood, although the proportion absorbed by children is higher, perhaps 30% to 40%.³⁶ The inhalable fraction less the respirable fraction represents that part of the aerosol which could potentially be absorbed through the gut and the respirable fraction that part which would be absorbed through the lung. These two measurements could then be combined as a weighted sum, based on the approximate efficiency of intake from each route of exposure. Alternatively, such data would provide reliable input data for developing the physiologically based pharmacokinetic models available for lead.³⁷

The United Kingdom Expert Panel on Air Quality Standards has recommended that there should be a standard of $0.25\mu\text{g}/\text{m}^3$ for lead, averaged over a year.³⁸ This standard is intended to protect the intellectual development of young people from inhaled lead aerosol. Unfortunately, the standard does not precisely specify the sampling procedure to be used and so it is unclear what size fraction it includes. For the fixed point measurements made by the United Kingdom Department of Environment, Transport, and the Regions this is probably unimportant because their samplers are generally located far from sources of coarse particles and most available samplers should give comparable results. However, differences in instrumentation would be much more important if the air quality standard were applied to personal exposure monitoring.

There is very little information on exposure to lead aerosol in the general population and the data that do exist have been obtained with samplers that do not conform to the agreed size selective sampling criteria.²⁵ Nevertheless, these data suggest that week long average lead exposures in Swedish adults range between 0.04 and $0.09\mu\text{g}/\text{m}^3$, as total dust. In the United Kingdom measurements of lead in the general atmosphere suggest that it makes up about 0.2% by mass of the thoracic (PM_{10}) aerosol. Applying this factor to the data shown in the table, assuming most of the variability in measured values is associated with the people

rather than the day of sampling, suggests that the average exposure level would be about $0.1\mu\text{g}/\text{m}^3$ and only about 5% of adults might exceed the United Kingdom air quality standard for lead. However, with the data from children there might be about half who could exceed the standard and it is this group who are especially at risk from lead exposure. Also, children living in homes where there was excessive lead contamination from flaking paint or from localised industrial sources might have personal exposures well above the standard.

Passive aerosol samplers

The appropriate averaging time for measurement should be dictated by the biological half life of the pollutant in the body with short term variations in exposure level being less relevant.³⁹ In the case of lead the biological half life is about 35 days³³ and this would be an appropriate averaging period. Clearly, measurement of personal lead exposure with conventional sampling strategies over such a long period is impracticable and the available data do not easily allow extrapolation to estimate such exposure. It might be possible to sample 24 hour average exposure on several occasions throughout a 5 week period and this would approximate to the long term exposure level, but this would still be costly and inconvenient for the subjects.

Sampling of gases and vapours has been revolutionised by the introduction of small lightweight samplers that do not have a pump but rely on the principle of diffusion to collect the material. Over the past 10 years there have been attempts to develop a similar approach for aerosols. The most promising of these, shown in figure 2, comprises a 25 mm diameter electret material held within an electrically conducting holder.⁴⁰ Electrets are polymers that have been treated to induce a permanent electrical charge within their structure, with the charge at the surface of the polymer as high as 1000 V. The sampler has a 1 cm gap between the surface of the electret and the front plate of the sampler and any particles that pass into this volume will drift towards the electret with a velocity proportional to their electrical mobility. The rate of collection of particles is relatively independent of their size, charge

Figure 2 (A) Passive aerosol sampler developed by the United Kingdom Health and Safety Laboratory.

distribution, or velocity, but this is highly dependent on the electrical mobility and this depends on the chemical composition of the aerosol.⁴¹

The sampler is small, lightweight (about 15 g), and not intrusive. It may be worn for extended periods and requires almost no operator maintenance over that period. Therefore it would be an excellent device to assess aerosol exposure of non-occupational groups and could be used to measure exposure to lead, although there are several limitations that make its use problematic. The rate of collection by the device is very low. In a study which is currently underway to assess the use of the device in assessing urban domestic aerosols, collected masses over 2 week periods were of the order of 50 µg. Also, in its present form the sampler is not size selective. In principle at least, it collects all sizes with equal efficiency, although the range of sizes over which this holds has not been measured. Finally, as already described, the collection rate is dependent on the electrical mobility, and hence composition of the particles. It is therefore necessary to calibrate the collection rate for different types of aerosol by comparing the passive sampler with a conventional pumped sampler in each case. This may lead to the requirement for material specific designs, coupled with appropriate analytical methods to be produced. Further work is necessary if these limitations are to be overcome.

Conclusions

There have been many developments in the sampling of aerosols over the past decade. In this paper we have highlighted the important attempts to standardise the fraction of the aerosol that is sampled so that it roughly corresponds to the fraction penetrating to sections of the respiratory tract. There is now, for the first time, agreement between all those involved in sampling workplace aerosols to use the same definitions so that measurements of respirable aerosol made in the United States, Europe, and other parts of the world are comparable. It would be advantageous for those involved with sampling non-occupational aerosols to ensure that their measurements are obtained to the same standard.

A consequence of moving to biologically relevant size fractions is the desirability of obtaining measurements of multiple size fractions for some aerosols. We have used the example of lead, where measurement of the inhalable and respirable aerosol might be appropriate. Use of this type of approach would certainly allow a more precise estimate of the biologically relevant exposure to be made and would enable more accurate estimates of risk from inhaled aerosol. It is more important to select an appropriate sampling system for personal exposure measurements than for fixed point monitoring because of the likelihood of resuspended dust playing a greater part and this should be carefully considered when sampling aerosols outside the workplace.

Finally, selecting an appropriate averaging time provides a particular challenge for all hazardous aerosols that have a chronic effect on health. Here there is an advantage in having sample averaging times as long as possible, consistent with the biological half life of the pollutant, so as to reduce the variability associated with short term samples. Continuous personal sampling for aerosols over several days or weeks is currently impracticable, but further development of passive samplers offers the possibility of adopting such strategies in the future. The key difficulty will be to develop systems which combine the need to obtain size selective samples with that of measuring over prolonged periods.

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- 2 Soter NA, Wasserman SI, Austen KF. Cold urticaria: release into the circulation of histamine and eosinophil chemotactic factor of anaphylaxis during cold challenge. *N Engl J Med* 1976;**294**:687–90.
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