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PARTICLE SIZE-SELECTIVE SAMPLING IN THE WORKPLACE: RATIONALE AND RECOMMENDED TECHNIQUES

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Abstract—Because many aerosol hazards are dependent upon particle size, the American Conference of Governmental Industrial Hygienists established an Air Sampling Procedures Committee to recommend size-selective aerosol sampling procedures which will permit reliable collection of aerosol fractions which can be expected to be available for deposition in the various major subregions of the human respiratory tract, e.g., the head, tracheobronchial region, and the alveolar (pulmonary) region.

After reviewing available data on regional deposition of inhaled particles and on the collection efficiencies of sampling instruments, the committee recommends use of three particulate mass fractions for workplace sampling. Inspirable Particulate Mass applies to material which is hazardous anywhere in the respiratory tract; Thoracic Particulate Mass applies to materials which are hazardous anywhere within the lung airways and the gas exchange region; Respirable Particulate Mass applies to material which is hazardous only in the gas-exchange region of the lung. Each of these 3 mass fractions are defined and a procedure to provide guidance in establishing corresponding Particle Size-Selective Threshold Limit Values (PSS-TLVs) has been suggested.

INTRODUCTION

DESPITE THE EVIDENCE that aerosol particle size can greatly modify responses to inhaled materials, the use of particle-size selective sampling for worker or public protection has only been applied in a few circumstances. Included are the British Medical Research Council's and the U.S. Atomic-Energy Commission's definitions of 'respirable dust', the American Conference of Governmental Industrial Hygienists' (ACGIH) Threshold Limit Values (TLVs) for quartz, cristobalite and tridymite; and the U.S. EPA's and the International Standards Organization's (ISO) definitions of aerosol fractions related to particle deposition within regions of the human respiratory tract (Air Sampling Procedures Committee, 1985). What follows is a brief summary of the report of the ACGIH technical committee on Air Sampling Procedures which was formed to recommend size-selective aerosol sampling procedures in workplaces (AIR SAMPLING PROCEDURES COMMITTEE, 1985).

Respiratory tract regions

Table 1 shows the major anatomical regions (and their included structures) considered in recommending aerosol sampling techniques. Also shown are essentially...
equivalent regions used by the Task Group on Lung Dynamics of the International Commission on Radiological Protection (TASK GROUP ON LUNG DYNAMICS, 1966) and the Air Quality Committee of the ISO (ISO, 1983).

### Table 1. Respiratory Tract Regions

| Region         | Anatomical Structures | Task Group Region | ISO Region |
|----------------|-----------------------|-------------------|------------|
| 1. HAR (Head Airways Region) | Nose, Mouth, Nasopharynx, Oropharynx, Laryngopharynx, Larynx | Nasopharynx (NP) | Extrathoracic (E) |
| 2. TBR (Tracheobronchial Region) | Trachea, Bronchi, Bronchioles (to terminal bronchioles) | Tracheobronchial (TB) | Tracheobronchial (B) |
| 3. GER (Gas Exchange Region) | Respiratory bronchioles, Alveolar ducts, Alveolar sacs, Alveoli | Pulmonary (P) | Alveolar (A) |

### Size-Selective Sampling Criteria for Inspirable Mass Fraction

The inspirable particulate mass fraction (IPM) of an aerosol is that fraction which can enter the uppermost respiratory system compartment, the head airways region (HAR). Deposited material may be retained, absorbed, swallowed, or expelled by mechanisms such as sneezing, spitting, or nose blowing. IPM may deposit anywhere within the respiratory tract, and there are at least three general classes of airborne toxic materials for which a valid hazard evaluation must consider all inspirable particles: 1) highly soluble materials which can quickly enter the blood; 2) materials which exhibit toxicity after entering the gastrointestinal tract; and 3) materials which exhibit toxicity at deposition sites within the respiratory tract. In the past, sampling has often relied on 'total dust samplers' whose ability to sample large particles has had an unknown relationship to the ability of the head airways to inspire material. It is only relatively recently that data on the inspirability (ratio of inspirable to total particulate mass concentration) as a function of particle size have been evaluated (VINCENT and ARMBRUSTER, 1981).

Several generalisations regarding inspirability can be drawn. It is clear that aerodynamic diameter is an important parameter in determining whether a particle enters the head. Inspirability is affected by wind speed, especially when facing the wind and it seems relatively insensitive to minute volume except in the case of facing a wind. Also, there seems to be little difference between nose and mouth breathing. For the purposes of presenting sampling criteria for IPM, the manikin results for orientation-averaged inspirability as a function of aerodynamic particle size are appropriate (MARK et al., 1985). These data cover a reasonable minute volume of 20L, include the
full torso, are averaged over orientation and include realistic workplace wind speeds.

The following function (see Fig. 1) can be used to define the IPM sampling efficiency $E(%)$ vs. aerodynamic diameter $d_a (\mu m)$: $E = 50 (1 + \exp[-0.06 d_a]) + 10$; for $0 < d_a \leq 100 \mu m$.

**Fig. 1.** The three aerosol mass fractions recommended for use in particle size-selective aerosol sampling by the ACGIH Air Sampling Procedures Committee.

**SIZE-SELECTIVE SAMPLING CRITERIA FOR THORACIC AND RESPIRABLE MASS FRACTIONS**

Thoracic particulate mass fraction (TPM) refers to the portion of the airborne particles that may penetrate the head airways and enter the lung airways when inhaling through the mouth. Respirable particulate mass fraction (RPM) refers to the ACGIH respirable dust sample (but with elaborated criteria) describing that portion of airborne particles that penetrate the head and tracheobronchial airways and enter the gas exchange region (GER) of the lung.

The most widely used models of regional deposition versus particle size were developed by the International Commission on Radiological Protection Task Group on Lung Dynamics (**TASK GROUP ON LUNG DYNAMICS, 1966**). Human deposition studies are numerous (**CHAN and LIPPMANN, 1980**; and **STAHLHOFEN et al., 1980**).

In light of the above, the TPM sampling criteria are recommended to be a tolerance band consisting of those particles that penetrate a separator whose size collection efficiency is described by a cumulative lognormal function with media ($d_{50}$) of $10 \pm 1 \mu m$ aerodynamic diameter and with geometric standard deviation ($\sigma_g$) of $1.5 \pm 0.1$ (Fig. 1).
Using the ACGIH respirable dust sample as the basis, the RPM sampling criteria are recommended to be a tolerance band consisting of those particles that penetrate a separator whose size collection efficiency is described by a cumulative lognormal function with median \( d_{50} \) of 3.5 \pm 0.3 \( \mu \)m aerodynamic diameter and with geometric standard deviation \( \sigma_g \) of 1.5 \pm 0.1 (Fig. 1).

**Sampler Efficiencies: Inspirable Mass Fraction**

Sampling in calm air has been evaluated by several investigators (DAVIES, 1968; OGDEN, 1983; AGARWAL and LIU, 1980). OGDEN (1983) concludes that blunt samplers in calm air at a 90% sampling efficiency yield particle aerodynamic diameters that are roughly one-half of those seen using thin-wall tube samplers. When settling velocities are small compared with air velocity, accurate thin-wall probe samples of large particles can be obtained by isokinetic sampling. Blunt samplers operating in a wind present a complicated situation and there is no unique probe velocity that permits sampling with 100% efficiency for all particle sizes in a given wind.

The VC 25G sampler approximately follows the inspirable mass sampling criteria for a narrow range of wind velocities higher than typical indoor wind velocities (ARMBRUSTER, et al., 1983). The sampling efficiency curve averaged for wind velocity for the GS 050/3 sampler is farther from the inspirable mass sampling criteria than the VC 25G (ARMBRUSTER, et al., 1983). HAMEED, et al. (1983) used a vertical axis, rotating-arm isokinetic sampler. A sampler described by OGDEN and BIRKETT (1978) known as the Orb sampler has also been used for inspirable mass sampling.

There are at present no commercially available samplers that fall within the IPM criteria envelope over the entire range of 0 to 100 \( \mu \)m aerodynamic diameter, but researchers at the Institute of Occupational Medicine, Edinburgh, UK have recently developed an inspirable sampler, the IOM/STD1, that comes close to matching the ACGIH criteria (MARK, et al., 1985).

**Sampler Efficiencies: Thoracic Mass Fraction**

The dichotomous sampler is a virtual impactor having a flow rate of 16.7 L/min. The thoracic particulate mass fraction is selectively passed through the inlet; the virtual impaction stage further fractionates the aerosol into coarse and fine fractions (LOO, et al., 1976).

Medium volume (20 to 150 L/min) samplers have been developed by MCFARLAND and ORTIZ (1982) and by WEDDING, et al. (1983). MCFARLAND and ORTIZ employ a sampler geometry which fractionates particles by a combination of impaction and sedimentation. A high-volume sampler based on a similar geometry, called the Size-Selective Inlet (SSI), converts a standard hi-vol into a thoracic mass sampler (MCFARLAND and ORTIZ, 1982). A small, portable sampler has been developed by BRIGHT and FLETCHER (1983). The thoracic cut is provided by the inlet, which contains a single stage impactor with an oil-soaked porous plate to suppress particle bounce. There are several samplers that fall within the TPM criteria envelope.
SAMPLER EFFICIENCIES: RESPIRABLE MASS FRACTION

Cyclones are probably the most commonly used RPM samplers. They are available having a wide range of flow rates including miniature cyclones for personal sampling. The sampling efficiency can be closely matched to that of a respirable curve. Cyclones have advantages including minimal particle bounce, large capacity for loading and insensitivity to orientation. A disadvantage of the cyclone is a lack of a fundamental theory which can predict performance. However, empirical theories are available to assist the designer (Chan and Lippmann, 1977; John and Reischl, 1980; Saltzman and Hochstrasser, 1983). Considerable data are available on the performance of the widely-used 10 mm nylon cyclone.

Horizontal elutriators have been widely used, particularly in the United Kingdom. Their main advantage is the predictable performance based on gravitational settling of particles. Disadvantages include the restriction to a fixed orientation, possible reentrainment of particle deposits and the difficulty of miniaturization.

The collection efficiency of an impactor can be accurately predicted by theory (Marple and Willeke, 1976). These instruments can be designed over a wide range of flow rates and operated in any orientation. A further advantage of the impactor is the possibility of cascading stages to provide size-segregated samples which can be analysed to produce the particle size distributions of specific chemical species. On the other hand, important problems such as particle bounce and wall losses cannot be reliably predicted.

USE OF SIZE-SELECTION IN ESTABLISHING TLVS

As shown in the decision flow diagram in Figure 2, the first step in deriving a Particle Size-Selective Sampling TLV (PSS-TLV) is the identification of the chemical substance that constitutes a potential air pollutant, including examination of physicochemical properties related to its airborne and biological behaviour, epidemiology, industrial hygiene, and toxicology. If no potential diseases related to the chemical substance are found, or if no airborne particles can be produced, then the evaluation can be terminated. However, if the physicochemical properties of the substance suggest that it may become airborne as an aerosol, the analysis proceeds and the physical and chemical properties are evaluated under conditions likely to be encountered by workers. Particle size-selective sampling is then necessary to estimate the quantity of the substance that will enter the three principal regions of the respiratory tract, the HAR, TBR and GER.

Once the substance is deposited in a particular region or regions of the respiratory tract, a critical consideration in selecting the appropriate particle mass fraction (IPM, TPM or RPM) is the extent of dissolution and systemic transport of the substance. Concurrent examination of the clinical diseases that may affect any systemic organ will identify extrapulmonary sites of action. Subsequently, it will be determined whether the incorporated dose of the substance is a critical dose that is likely to cause acute or chronic injury. Once the particle size, particulate mass fraction, and the hazard analyses are completed, a critical mass concentration will be determined for an appropriate size fraction. This assessment will result in a recommendation for a particle size-selective threshold limit value (PSS-TLV).
FLOW DIAGRAM OF THE INFORMATION TO BE CONSIDERED IN THE DEVELOPMENT OF PARTICLE SIZE SELECTIVE THRESHOLD LIMIT VALUES (PSS-TLVs)

Fig. 2. Flow diagram of the information to be considered in the development of Particle Size-Selective Threshold Limit Values.

If the inhaled chemical material is likely to dissolve only slowly, or is essentially insoluble after deposition in any of the three principal regions of the respiratory tract, selection of the appropriate particle size-selective sample should be based on the specific site of action within the respiratory tract that is associated with the most restrictive PSS-TLV based on each potential disease.

RECOMMENDATIONS

Having considered many aspects of size-selective sampling in evaluating inhalation hazards in the workplace including: 1) effects of particle size on deposition site within the respiratory tract, 2) the tendency for many occupational diseases to be associated with chemical substances deposited in particular regions of the respiratory tract, 3) the availability of suitable samplers, and 4) the relative inappropriateness of poorly defined ‘total dust’ samples presently collected, the Air Sampling Procedures Committee recommends that the ACGIH Chemical Substance TLV Committee develop Particle Size-Selective TLVs (PSS-TLVs). PSS-TLV is a general term for: Inspirable Particulate Mass TLVs (IPM-TLVs) for those materials which are hazardous when deposited anywhere in the respiratory tract; Thoracic Particulate Mass TLVs (TPM-TLVs) for those materials which are hazardous when deposited anywhere within the lung airways and the gas-exchange region; Respirable Particulate Mass (RPM-TLVs) for those materials which are hazardous when deposited in the gas-exchange region.

These recommendations differ from those of the ISO primarily with respect to the definition of IPM. Although our criteria curves cannot be used to reliably estimate
particle deposition in any airway region, they are more readily applicable to sampling in the occupational setting.

REFERENCES

AGARWAL, J.K. and LIU, B.Y.H. (1980) A criterion for accurate aerosol sampling in calm air. Am. Ind. Hyg. Assoc. J. 41, 191–197.

AIR SAMPLING PROCEDURES COMMITTEE (1985) Particle Size-Selective Sampling in the Workplace, American Conference of Governmental Industrial Hygienists, Cincinnati, OH.

ARMBRUSTER, L., BREUER, H., VINCENT, J.H. and MARK, D. (1983) Definition and measurement of inhalable dust. Aerosols in the Mining and Industrial Work Environment. (V.A. MARPLE and B.Y.H. LIU, Eds.) Ann Arbor Science Publishers, Ann Arbor, MI.

BRIGHT, D.S. and FLETCHER, R.A. (1983) New portable ambient aerosol sampler. Am. Ind. Hyg. Assoc. J. 44, 528–536.

CAPLAN, K.J., DOEMENY, L.J. and SORENSON, S.D. (1977) Performance characteristics of the 10 mm cyclone respirable mass sampler. Part 1 — Monodisperse studies. Am. Ind. Hyg. Assoc. J. 38, 83–95.

CHAN, T. and LIPPMANN, M. (1977) Particle collection efficiencies of air sampling cyclones: an empirical theory. Environ. Sci. Technol. 11, 377–382.

CHAN, T. and LIPPMANN, M. (1980) Experimental measurements and empirical modelling of the regional deposition of inhaled particles in humans. Am. Ind. Hyg. Assoc. J. 41, 399–409.

DAVIES, C.N. (1968) The entry of aerosols into sampling tubes and heads. Br. J. Appl. Phys. D. 2s(1), 921–932.

HAMEED, R., McMURRY, P.H. and WHITBY, K.T. (1983) A new rotating coarse particle sampler. Aerosol Sci. Tech. 2, 69–78.

ISO (1983) Air Quality-Particle Size Fraction Definitions for Health Related Sampling. ISO/TR 7708–1983 (E). International Standards Organization.

JOHN, W. and REISCHL, G. (1980) A cyclone for size-selective sampling of ambient air. J. Air Pollut. Control Assoc. 30, 872–876.

LOO, B.W., JAKLEVIC, J.M. and GULDING, F.S. (1976) Dichotomous virtual impactors for large scale monitoring of airborne particulate matter. Fine Particles. (B.Y.H. LIU, Ed.) Academic Press, Inc., New York, NY, pp. 312–350.

MARK, D., VINCENT, J.H., GIBSON, H. and LYNCH, G. (1985) A new static sampler for airborne total dust in workplaces. Am. Ind. Hyg. Assoc. J. 46, 127–133.

MARPLE, V.A. and WILLEKE, K. (1976) Impactor design. Atmos. Environ. 10, 891–896.

MCFARLAND, A.R. and ORTIZ, C.A. (1982) A 10 µm cutpoint ambient aerosol sampling inlet. Atmos. Environ. 16, 2959–2965.

OGDEN, T.L. and BIRKETT, J.L. (1978) An inhalable-dust sampler, for measuring the hazard from total airborne particulate. Ann. Occup. Hyg. 21, 41–50.

OGDEN, T.L. (1983) Inhalable, inspirable and total dust. Aerosols in the Mining and Industrial Work Environment. (V.A. MARPLE and B.Y.H. LIU, Eds.) Ann Arbor Science Publishers, Ann Arbor, MI, pp. 185–204.

SALTZMAN, B. and HOCHESTRASSER, J.M. (1983) Design and performance of miniature cyclones for respirable aerosol sampling. Environ. Sci. Technol. 17, 418–424.

STAHLHOFEN, W., GERHART, J. and HEYDER, J. (1980) Experimental determination of the regional deposition of aerosol particles in the human respiratory tract. Am. Ind. Hyg. Assoc. J. 41, 385–398.

 Task Group on Lung Dynamics (1966) Deposition and retention models for internal dosimetry of the human respiratory tract. Health Phys. 12, 173–207.

VINCENT, J.H. and ARMBRUSTER, L. (1981) On the quantitative definition of the inhalability of airborne dust. Ann. Occup. Hyg. 24, 245–248.

WEEDING, J.B., WEIGAND, M.S., LIGOTKE, M.W. and BAUMGARDNER, R. (1983) The Wedding ambient aerosol sampling inlet for an intermediate flow rate (4 cfm) sampler. Environ. Sci. Technol. 17, 379–383.
DISCUSSION

D. C. F. Muir: Does the term ‘thoracic fraction’ mean that dust can first be inhaled, then pass through the head, and then enter the thorax? I did not see the first of these steps on your slide.

What sampling curve would you suggest for the case of a dust which causes carcinoma of the nose?

O. Raabe: As a member of the ACGIH Committee, I would like to respond to Dr. Muir’s question.

It was the intention of the ACGIH to recommend that the three types of samples be independently and separately collected. This simplifies the job of the occupational hygienist, since he can collect whatever sample is most appropriate for his application. The thoracic particulate matter fraction can be collected for dusts that affect the tracheobronchial and gas exchange region, without collecting an inspirable sample first. There is no need for taking samples in tandem. There is only a small error in this procedure, which we believe to be acceptable, considering the sampling ease given to the hygienist.

M. Lippmann: If the hazard is nasal cancer, then IPM is clearly the appropriate sampling criterion. On the other hand, if wood dust causes diseases in the lungs also, then other size-selective TLVs, with appropriate size cuts, may also be needed to protect against excessive exposures.

M. T. R. Reisner: I wonder why present recommendations for size-selective sampling in the workplace are made which do not consider the effect-related, biologically-relevant dust fractions. To prevent pneumoconiosis, for example, it is necessary that the responsible men in the industry get a correct target on which they can focus their activities in dust suppression, and deployment of workers.

By over-estimating particles less than 2 µm, and under-estimating particles greater than 4 µm, they will be misled, as we were in our epidemiological studies in West German coal-mines. * On the basis of wrongly-estimated dust exposure data, we got misleading exposure-response relationships, which generally resulted in the false conclusions, namely that dusts rich in minerals and quartz are less harmful than those with lesser contents of minerals, including quartz.

Also, for biological tests to find out the specific harmfulness of dusts, it is very important to use the human ‘alveolar dust fraction’, because the in vitro and in vivo effects are related to both the site of the dust, and its mineral composition which, in turn, depends on size. This will be demonstrated by 3 papers in a later session.

*M. T. R. Reisner, (1984) VIth International Pneumoconiosis Conference 1983, Bochum, Vol.1, Geneva, IL0, pp. 132-155.

M. Lippmann: There are conflicting processes, (upper airway removal, expiration of particles) which combine to produce a relatively constant fraction of alveolar deposition for ‘respirable dust’. Thus, a mass concentration limit which recognizes that deposition is only 20% of available dust, can provide good protection.

In theory, samplers which collect dust which deposits in the alveolar region are preferable. However, we felt that their complexity in practice outweighed their theoretical value.

Oscar Williams: Could you advise if a millipore closed-face cassette, with 4mm orifice and flow of 21/min is suitable for the measurement of inert or nuisance dust? If not, could you suggest an alternative?

S. Soderholm: There are some data available in the literature on the collection efficiency of current samplers, but not enough to compare to the recommendations. In general they may be adequate for a range of windspeeds, and small enough particle sizes, but no definite recommendations can be made now for improved sampling in a particular workplace with the available information, in my opinion.

The explicit inclusion of particles as large as 100 µm in these recommendations, may prompt a re-thinking of the biological and toxicological basis for a nuisance dust standard.

There is serious thought being given to the term ‘nuisance dust’ by the ACGIH TLV Committee. Is it a meaningful term, in view of increasing knowledge of susceptible persons? The next few years may see consideration given to doing away with the term.

W. H. Walton: The abbreviation TPM is used for total particulate matter. Could you use something else for the thoracic mass fraction?

R. Phalen: The committee discussed the issue and felt that TPM would be understood in this context.

T. L. Ogden: The problem of terminology in this field is almost as difficult as the problem of deciding where the aerosol size cuts come.

C. Santos-Bergoa: What assumptions did you include for your recommendation on inspired minute volumes, particle surface characteristics (not all are spheres), and particle density?

I am very glad that again we are paying attention to something else beyond the so-called respirable dust. Bronchitic COPD, and bronchogenic cancer groups, are very likely to be affected by the TPM fraction.
When such measures become available in the future, it will be an advantage for epidemiologists, when studying the variability and inconsistencies in dose-response curves.

The real problem with your recommendations is at the extremes of the curves. In the small-size side of your last slide curves, you have non-independent probabilities of a particle to be included in any of the fractions; however, at the larger sizes, they are independent one to the other (at least they seem to be in your graph), and I think this will give a big problem for epidemiological hazard evaluation. Would you like to comment on this?

R. PHALEN: We used the minute volume recommended by the International Commission on Radiological Protection, that is 21.75 litres. All particle diameters are aerodynamic, except for those below about 0.5 micrometers where a diffusive diameter should be used. (For a suitable reference see Raabe, O.G., Physical properties of aerosols affecting inhalation toxicology, in Pulmonary Toxicology of Respirable Particles, Conf.79(002, Ed. Sanders et al., pp. 1-28, 1980. Technical Information Center, US. Department of Energy).

Epidemiologists have different requirements for their studies from industrial hygienists. I certainly recommend that research studies use more extensive aerosol sampling procedures than those recommended solely for worker protection.

J. N. PRITCHARD: In view of the work of Vincent, that 50% of particles at 100 μm and beyond are inspirable, I would suggest that setting an inspirable curve is adding an unnecessary complication in the design of samplers.

There are a great many assumptions going into the setting of a size-selective TLV, so that adding an extra safety margin of a factor of 2 between sampling total dust and inspirable dust is unnecessary precision.

In your concluding remarks, you suggested that a total dust sample could underestimate a hazard. Can you expand on this?

R. PHALEN: An inspirable curve is necessary, as all samplers have size-selection characteristics which are typically undefined. Samplers should conform to a known characteristic curve.

Total dust samples, as they are often taken, can significantly undersample the inspirable airborne mass. This is clearly shown in a recent paper by Buchan et al., (Aerosol sampling efficiency of 37 mm filter cassettes, Am. Ind. Hyg. Assoc. J., 47 (12), 825-831, 1986.