LUNG DEPOSITION OF AEROSOLS FROM INDUSTRIAL ENVIRONMENTS

H J Annegarn, University of the Witwatersrand
S Bauman, A C D Leslie, J W Winchester
Florida State University U.S.A.

OPSOMMING:

Verslag word gedoen oor toetsresultate van 'n aerosol monsternemingstelsel vir menslike uitgeasemde lug om die respiratoriese neerslagkurwe van die swaarmetaalkomponente van die aerosol te bepaal. Gelyktydige monsterneming van ingeasemde omgewingsaerosol en uitgeasemde aerosol deur kaskade stofsuiers en element-analise deur deeltjiegeïndusseerde x-straal emissie (PIXE) laat die bepaling van deeltjieneerslag in die aerodinamiese deursnee bereik <0,25 tot > 4 μm toe. Resultate waar die apparaat in 'n sweiswinkel gebruik word, word gegee en daar word aangetoon hoe die neerslag verander met deeltjiegrootte en hoe die akkuraatheid in die bepaling van die neerslag afhang van monsternemingstoestande. Toepassing in die myne sal meebring dat metings op mynwerkers gemaak kan word terwyl hulle werk.

SYNOPSIS:

Test results are reported on an aerosol sampling system for exhaled human breath, to determine the respiratory deposition curve of the heavy metal components of the aerosol. Simultaneous sampling of inhaled ambient aerosol and exhaled breath aerosol by cascade impactors and elemental analysis by particle-induced X-ray emission (PIXE) permits determination of particle deposition in the range <0,25 to > 4 μm aerodynamic diameter. Results are reported using the apparatus in a welding shop, showing how the deposition varies with the particle size and how the precision determining percent deposition depends on sampling conditions. The application to mining environments would permit measurements to be made on miners at work.

INTRODUCTION

The likelihood for deposition of inhaled particles depends on the aerodynamic diameter of the particles as well as position along the airway, so that upper airways tend to intercept larger particles while permitting finer sizes to enter and be deposited in the lower regions. Overall deposition efficiency is expected to be a minimum at $\sim$ 0,5 μm aerodynamic diameter. This is due to the contrary trends with decreasing particle size for (i) decreasing impaction efficiency and (ii) increasing Brownian diffusion to the lung walls. The atmosphere of industrial work environments may frequently be polluted with respirable size range i.e. < 2 μmad, aerosol particles which can bypass the defences of the upper respiratory tract and be deposited in the lung.

A prediction of aerosol deposition efficiency in the human respiratory tract may be based on simultaneous measurements, as a function of particle size, of concentrations i) in the exhaled breath, and ii) in the ambient atmosphere. These measurements may be made for the elemental constituents of aerosols, by means of size selective sampling using cascade impactors and analysis using particle-induced X-ray emission (PIXE) on particle elastic scattering analysis (PESA) in the proton beam of a Van de Graaff accelerator. Preliminary results have been reported earlier[1-7]. This paper reviews conditions of sampling which must be met for a quantitative determination of lung deposition of aerosols from an industrial environment.

EXPERIMENTAL

To quantify the measurement of lung deposition of aerosols several conditions must be met which in general are more stringent than is customary for ambient aerosol concentration measurements. The experimental arrangement used for sampling exhaled human breath is shown in Fig. 1. The subject exhales into a chamber maintained at body temperature and thereby body relative humidity is also maintained in the aerosol. While sampling from the chamber at a constant rate, pulsating breathing patterns are compensated by a balloon arrangement which maintains a constant atmospheric pressure in the chamber. Sampling is carried out by four cascade impactors operating simultaneously or in pairs as desired. A spirometer records breathing patterns during sampling. Two modifications have been made on an earlier design for greater sampling precision: the human subject breathes vertically downward into the chamber, thus avoiding particle losses due to settling in a horizontal entry tube for exhaled air, as used previously. We also position the cascade impactors below rather than above the chamber volume, thus allowing the largest particles to settle into rather than away from the impactor; this also shortens the length of the exhaled air inlet. The interior of the chamber is composed of electrically conducting materials including a metal foil cover for the balloon, so as to avoid electrostatic effects which may interfere with proper aerosol sampling.

The cascade impactor collects samples in particle size fractions <0,25, 0,25–0,5, 0,5–1, 1–2, 2–4 and >4 μm aerodynamic diameter on specially treated Mylar surfaces which achieve efficient particle retention. Additional low pressure stages may also permit separation of the smallest size fraction into <0,06, 0,06–0,12, and 0,12–0,25 μmad ranges. Therefore the impactors permit separation of the aerosol into fractions on both sides of the expected respiratory deposition minimum near 0,25–0,5 μmad.
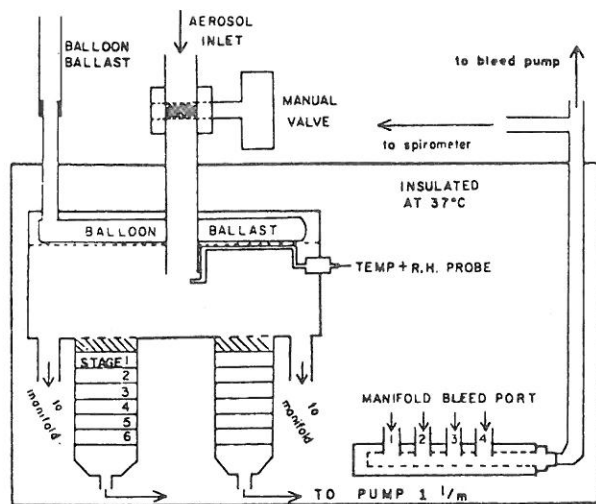
FIG 1   A schematic diagram of the modified exhaled human breath sampling chamber. Only 2 of the 4 impactors are shown. Manual valve at aerosol inlet is operated by subject in accordance with his or her breathing pattern.

FIG 2   Particle size distributions for iron and dry ambient welding aerosol, humidified ambient aerosol, and duplicate samples of exhaled aerosol.

Simultaneously with the sampling of exhaled air, the ambient atmosphere is sampled by four or more impactors. All samples are analyzed by particle X-ray emission, PIXE, for the concentration of elemental constitutents ranging from sulphur to lead in atomic number.[8]

Due to absorption by hygroscopic particles the particle distribution in the lung may shift to larger aerodynamic diameters compared to the exhaled air.

Hence it is important to compare inhaled and exhaled elemental concentrations in particle size ranges for the same relative humidity conditions. For relatively dry atmosphere the ambient impactors may be fitted with a vertical channel lined with moistened filter papers to allow the relative humidity to rise above 90% before the particles are size fractionated. In this way, the relative concentrations of chamber and ambient aerosol particle size fractions may be interpreted as indicating directly the relative amount of particle loss in the respiratory tract.

RESULTS

1.   Human subject deposition experiments

Welding aerosol has been selected as representative of some indoor working atmospheres of industrial health interest. Fig. 2 shows the concentrations of iron found by Ahlberg et al.[6] in the exhaled breath of a human subject by duplicate chamber impactors and the corresponding ambient Fe concetntrations as measured by cascade impactors with and without pre-humidification of the indoor welding 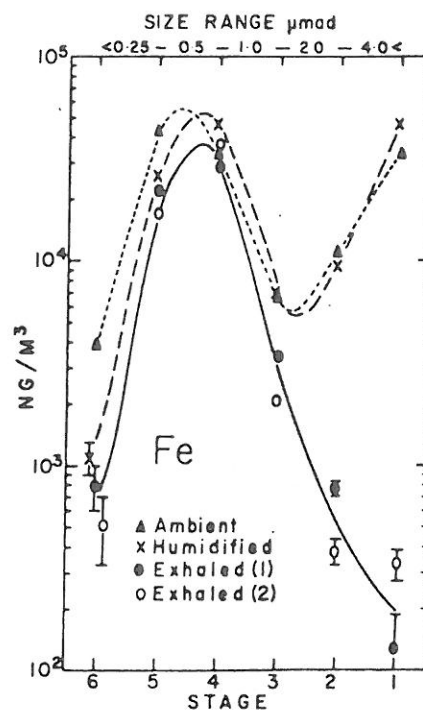shop atmosphere. Clearly exhaled concentrations are lower than ambient, and the ambient atmosphere particle size distribution shows a shift to larger particle sizes following humidification.

Fig 3 shows the same Fe data as well as corresponding data for Ni, Cr and Mn in the same samples, expressed as percent deposition obtained from the ratios of the exhaled aerosol to either humidified or dry ambient samples. The apparent minimum in the deposition curve is lower by one impactor stage, i.e. half the particle diameter, for the humidified case compared with the dry ambient aerosol. Therefore, when sampling in a dry ambient atmosphere, a correct estimation of the lung deposition curve requires knowledge of particle size shifts due to aerosol humidification as well as overall deposition in the respiratory tract.

Fig. 4 shows the Fe deposition curve, calculated from the humidified ambient aerosol sample, compared with (i) the Task Group on Lung Dynamics[9] curve and (ii) early experimental measurement of automotive Pb aerosol carried out in a naturally humid atmosphere.[1] In all three cases the region of the deposition minimum is in the range 0,25 − 0,5 μmad. However, only one measurement point defines the greater deposition in the smallest particle size range. Two points are needed to determine the minimum more accurately, and the apparent minimum in Fig. 4 should be regarded as an upper size limit. The addition of low pressure impactor stages, as mentioned previously, may provide this information.
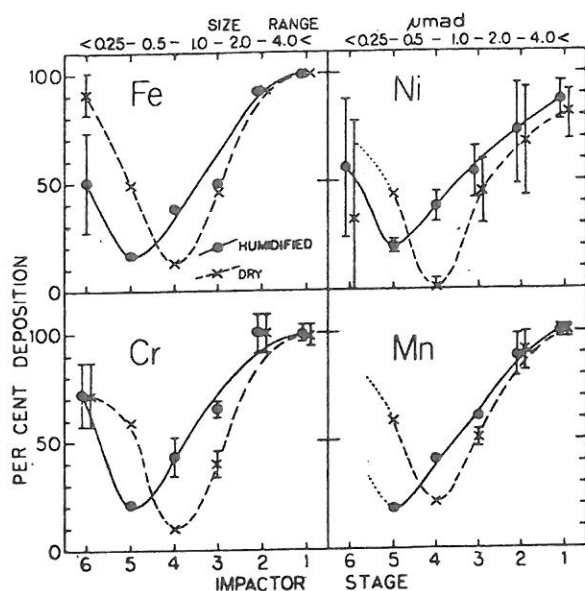
FIG 3   Percent deposition of Fe, Ni, Cr and Mn in the respiratory tract referenced to the humidified welding aerosol (80% R.H.) and ambient welding aerosol (=50% R.H.).
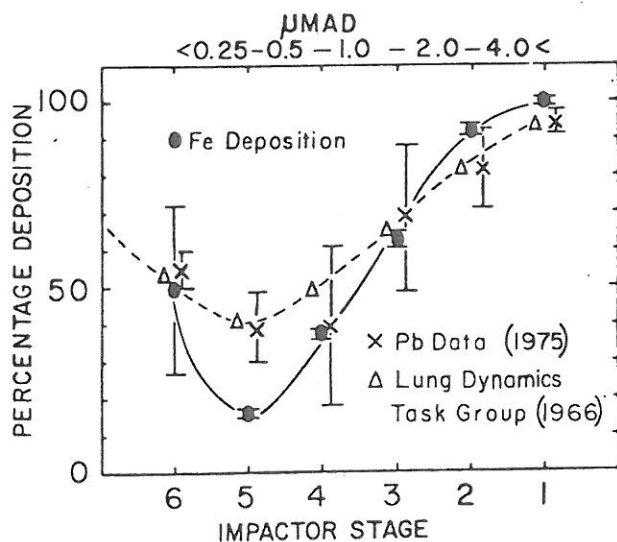

FIG 4   Percent deposition of Fe in the respiratory tract compared to the deposition curve of the Task Group on Lung Dynamics.[9] The Pb deposition data obtained in earlier experiments are also shown.[1] The minimum deposition point in all cases appears to the 0,25—0,5 μm aerodynamic diameter.

2.   Tests of sources of sampling error

The precise determination of lung deposition of aerosol particles by the difference method reported here requires careful controls of sampling conditions far more stringent than often needed in general ambient atmosphere investigations.

The need for accurate humidity control has already been discussed. A transfer of material from a smaller to a larger particle size range on humidification could deceptively indicate an apparent respiratory deposition of the smaller particles unless it is realized that they may be merely shifted to a larger particle size fraction. Since welding aerosol has a maximum in its particle size distribution, shown in Fig. 2 near the point of minimum respiratory deposition, the transfer from the smallest range to larger sizes by humidification causes a greater relative decrease in the < 0,25 μmad fraction than the corresponding increase in the 0,25—0,5 μmad fraction. Together with humidity control temperature must be controlled since at high humidities a 1°C temperature change corresponds to about 6% change in relative humidity.

TABLE 1   ELEMENTAL WEIGHT RATIOS OF SUMS < 4 μmad PARTICLE SIZE FRACTIONS IN CHAMBER AND AMBIENT SAMPLES OF WELDING AEROSOL.

	Weight Ratio			
Sampler	Mn/Fe	Ca/K	Ca/Fe	K/Fe
Ch. 1	0,146	0,570	0,319	0,559
Ch. 2	0,144	0,550	0,265	0,481
Amb. 1	0,155	0,592	0,315	0,533
Amb. 2	0,154	0,592	0,318	0,537
Amb. 3	0,146	0,589	0,316	0,536
Amb. 4	0,146	0,590	0,302	0,513
Av. Ch. / Av. Amb.	0,97	0,95	0,93	0,98

Table 1 shows weight ratios measured in a chamber calibration experiment using welding aerosol. When concentrations are summed over all particle sizes up to 4 μmad, the overall elemental ratios are found to be in close agreement to within a few percent, for the two chamber and four ambient aerosol samples collected. The precision of PIXE analysis is apparently very great and suitable for the precise determination of lung deposition of aerosols from industrial environments.

Table II compares the particle size distributions of FE, Mn, Ca, and K in the two chamber and four ambient impactor samples just discussed. The distributions are expressed as percent of the <4 μmad summation occuring in each of the five size ranges. Significant variability in these size distribu-

Impactor Stage and Particle Size Range*

Element	Sampler		6 <0,25	5 0,25—0,5	4 0,5—1	3 1—2	2 2 4
Fe	Ch.	1	0,6	25,2	70,5	2,5	1,2
	Ch.	2	5,7	15,2	74,8	3,1	1,1
	Amb.	1	22,5	6,6	65,7	3,3	1,9
	Amb.	2	5,4	14,4	73,2	5,0	2,1
	Amb.	3	22,9	31,7	41,0	3,2	1,2
	Amb.	4	32,9	13,4	49,9	2,2	1,5
	Av. Ch. / Av. Amb.		0,15	1,22	1,26	0,82	0,69
Mn	Ch.	1	–	19,4	76,3	3,2	1,1
	Ch.	2	5,4	9,4	80,1	4,2	1,0
	Amb.	1	19,5	3,6	71,2	4,2	1,5
	Amb.	2	4,9	9,0	77,9	6,4	1,7
	Amb.	3	23,1	25,2	46,6	4,1	1,1
	Amb.	4	32,9	13,2	49,4	3,1	1,4
	Av. Ch. / Av. Amb.		0,13	1,13	1,28	0,83	0,74
Ca	Ch.	1	1,0	22,5	73,4	2,1	1,0
	Ch.	2	7,1	8,3	80,5	3,1	1,1
	Amb.	1	21,4	3,7	70,6	2,8	1,6
	Amb.	2	5,6	8,4	79,9	4,4	1,7
	Amb.	3	25,7	25,2	45,4	2,8	1,0
	Amb.	4	35,7	14,0	46,6	2,2	1,5
	Av. Ch. / Av. Amb.		0,18	1,20	1,27	1,02	0,72
K	Ch.	1	–	21,5	76,6	1,5	0,5
	Ch.	2	5,2	9,1	83,1	2,1	0,6
	Amb.	1	18,8	3,8	75,0	1,8	0,7
	Amb.	2	3,8	9,4	83,7	2,6	0,5
	Amb.	3	23,5	27,7	46,4	2,1	0,4
	Amb.	4	29,5	15,3	53,3	1,1	0,9
	Av. Ch. / Av. Amb.		0,14	1,09	1,24	0,95	0,88

*Size fractions designated by cascade impactor stage number and approximate range of aerodynamic particle diameters in micrometers, μmad.

tions is observed, although the chamber samples and ambient aerosol samples appear to be part of the same distribution, and no systematic difference between chamber and ambient samples can be discerned. The overall ratio of chamber and ambient samples on the average is within 20—30% of unity, except for stage 6, indicating that the chamber aerosol, when operated without a human subject

and at room temperature and humidity conditions, is sampling the aerosol with the same efficiency within this uncertainty, as the unhumidified ambient samplers.

Table III shows the actual concentrations measured by the two chamber and four ambient aerosol impactors as sums of the five particle size fractions less than 4 μmad. The four ambient impactors were arranged symmetrically around the inlet to the chamber, and the two chamber impactors were a few centimeters apart. The welding aerosol had been generated at a distance of two meters away about 15 minutes before a 10-minute sampling period commenced, and there was no fan operating in the room or in the chamber to assure homogeneity of the air during sampling. It is clear from these results that ambient impactors 1 and 2 recorded about half the concentration of impactors 3 and 4, apparently reflecting non-uniformity of the air in the room. The two chamber samples record intermediate concentrations, reflecting the intermediate position of the chamber inlet compared to the arrangement of four ambient impactors. However, the two chamber samplers do not record identical concentrations, suggesting some non-uniformity of the air in the chamber volume. Consequently, precise differences between ambient and chamber concentrations should be determined by multiple sampling and good mixing conditions in the air both in the room and in the chamber. This may be achieved by suitable fans or long sampling times.

Elemental Concentration, ng/m³

Sampler		Fe	Mn	Ca	K
Ch.	1	395	58	126	221
Ch.	2	302	44	80	146
Amb.	1	257	40	81	137
Amb.	2	256	39	81	138
Amb.	3	483	71	152	259
Amb.	4	500	73	151	256
Av. Ch. / Av. Amb.		0,93	0,91	0,89	0,93

DISCUSSION

The results presented above indicate that elemental concentrations in exhaled human breath aerosol samples, collected in particle size fractions in the respirable size range, may be precisely determined by PIXE analysis. A human subject breathing normally contaminated air for a few minures or longer can provide a sample large enough for this analysis. Since the cascade impactors used draw only

one liter per minute, several samplers may operate simultaneously and still provide enough exhaled air for monitoring of breathing patterns by a spirometer.

High precision machining or casting techniques should be used in the manufacture of cascade impactors, especially when resolution of very fine particle size fractions is required. The impactors used in this study were optimized in their manufacture for general ambient aerosol studies, not for the stringent needs of exhaled human breath sampling. Considerably greater precision of measurement could be obtained using more precisely built impactors. The use of low pressure stages may simplify the collection of smaller particles size fractions as well as extend the range of particle resolution to well below the expected respiratory deposition minimum.

Uniformity of the air surrounding the human subject being tested is perhaps the most severely limiting condition in determining the relative concentrations of ambient and exhaled aerosol collected in the chamber. This uniformity may be improved considerably over that of the conditions of these tests by suitable air mixing devices as discussed above.

The need for humidification of the ambient aerosol for studies of dry atmospheres is important. The vertical humidifying device we have employed apparently does not lead to substantial particle losses and is in principle suitable for this purpose.

Application of the technique to industrial environments, where dust deposition in the respiratory tracts of workers is of concern, should be straightforward. The sampling devices are compact and portable and require little electrical power for their operation. In mining environments in particular the present technique would permit the testing of workers under their normal conditions of physiological stress and air quality.

REFERENCES

1. G G DESAEDELEER and J W WINCHESTER, Trace Metal Analysis of Atmospheric Aerosol Particle Size Fractions in Exhaled Human Breath, Environmental Science & Technology 9 (1975) 972–972.

2. K AKSELSSON, G G DESAEDELEER, B JOHANNSON and J W WINCHESTER, Particle Size Distribution and Human Respiratory Deposition of Trace Metals in Indoor Work Environments, Ann. Occup. Hyg. 19 (1976) 225–238.

3. A C D LESLIE, J W WINCHESTER, F W LEYSIEFFER and M S AHLBERG, Prediction of Health Effects of Pollution Aerosols, in Trace Substances in Environmental Health – X, D D Hemphill, ed., University of Missouri, Columbia, 1976.

4. G G DESAEDELEER, J W WINCHESTER and K R AKSELSSON, Monitoring Aerosol Elemental Composition in Particle Size Fractions for Predicting Human Respiratory Uptake, Nucl. Instr. and Methods 142 (1977) 97–99.

5. M S AHLBERG, A C D LESLIE and J W WINCHESTER, Environmental and occupational Health Analyses Using Proton-Induced X-ray Emission, in Electron Microscopy and X-ray Applications to Environmental and Occupational Health Analysis, Philip A Russell and Alan E Hutchings, eds., Ann Arbor Science Publishers, Ann Arbor, 1978.

6. M S AHLBERG, H J ANNEGARN, O DAHLBÄCK, A C D LESLIE and J W WINCHESTER, Particle Size Dependent Aerosol Deposition in the Human Respiratory Tract, paper presented at 12th Annual Conference on Trace Substances in Environmental Health, University of Missouri, Columbia, MO, June 5–8, 1978.

7. H J ANNEGARN, S E BAUMAN, G T HALL, M M JOHNSTON, A C R LESLIE, D SCHUYKENS and J W WINCHESTER, Sampling Exhaled Aerosol in Ambient Atmosphere Human Inhalation Studies, paper presented at session of "Particle Analysis and Sampling" at the American Institute of Chemical Engineers 86th National Meeting, Houston, Texas, April 1–5, 1979.

8. T B JOHANNSON, R E VAN GRIEKEN, J NELSON and J W WINCHESTER, Elemental Trace Analysis of Small Samples by Proton Induced X-ray Emission, Analytical Chemistry 47 (1975) 855–860.

9. Task Group on Lung Dynamics Committee II, Health Physics 12 (1966) 173–208.