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PARTICLE SIZE DISTRIBUTION AND HUMAN RESPIRATORY DEPOSITION OF TRACE METALS IN INDOOR WORK ENVIRONMENTS

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Abstract—Respiratory response to inhalation of fine particles has been investigated for the aerosol generated by welding. Particles were sampled using a pair of 5-stage cascade impactors operating at 1 l./min flow rate. The subject exhaled into one impactor through an air ballast arrangement, and the other impactor simultaneously sampled the surrounding air. Particle size fractions were analyzed for principal elemental constituents from sulphur to lead using proton induced X-ray emission, PIXE. The results indicated a complex respiratory response, including both increase in particle size due to exposure to high humidity in the respiratory tract and deposition of particles during inhalation. The response was found to be different for the element group Mn, Cr, Fe, Ni compared to the group K, Ca, Ti by observing the associations among the elements as a function of particle size in the inhaled and exhaled aerosol. However, for respiratory deposition efficiency alone in all runs averaged together, no systematic differences between the different elements are demonstrated at the 99% confidence level.

INTRODUCTION

HEALTH effects of airborne particulate matter depend on physiological and anatomical factors as well as on the physical properties of aerosols (DAVIES, 1961; 1967; WALTON, 1971; TASK GROUP ON LUNG DYNAMICS, 1966; NATIONAL AIR POLLUTION CONTROL ASSOCIATION, 1969). There has been little possibility for direct measurement of the effect of these properties for indoor or outdoor ambient aerosols on humans. Laboratory experiments using synthetic aerosol particles cannot readily reproduce all properties of a natural aerosol, and predictions of pulmonary deposition or response to particle diameter, composition, solubility, and shape in respiratory cavities can be made only approximately. If a technique were available which could utilize a natural aerosol in human breathing experiments, some of the uncertainties of such predictions might be reduced. It is the purpose of the present investigation to test the feasibility of using a new technique: elemental analysis by proton-induced X-ray emission, PIXE (JOHANSSON *et al.*, 1970), of particle size fractions $>0.1 \mu\text{m}$, collected by cascade impactors sampling ambient air and exhaled breath. The technique is widely employed in studying urban air pollution on a regional scale (WINCHESTER *et al.*, 1974), and extensions of the technique to a continental scale are in progress on a routine basis.

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Application of PIXE to analysis of cascade impactor samples of ambient welding aerosol has already been reported (AKSELSSON *et al.*, 1974).

PIXE is a method of elemental analysis which enjoys certain advantages over other high sensitivity X-ray methods, and it may be applied to measuring human respiratory response to aerosol inhalation from ambient atmospheres. In our procedure (JOHANSSON *et al.*, 1975), small samples of millimeter dimensions, supported by thin filter or polystyrene backings, are exposed to a beam of 3.75 MeV protons from a Van de Graaff accelerator, for periods of 2–10 min. Characteristic X-rays are detected by a Si(Li) detector and are recorded in a computer data acquisition system during the course of the proton irradiation. Afterwards the resulting multi-element spectrum is resolved by computer (KAUFMANN and AKSELSSON, 1975), such that a quantitative analysis for each element present in nanogram amounts or more is reported. Elements sulphur and heavier can be determined by this technique. The high intensity of proton radiation reaching the target sample, which assures high analytical sensitivity, permits analysis of particle size fractions collected by cascade impactors of the Battelle design (MITCHELL and PILCHER, 1959) having a single orifice per stage. The high sensitivity of PIXE and its capability for analysing samples of small dimensions are of decided advantage in meeting this sampling condition.

For elemental concentrations of micrograms per cubic meter, only a few litres of air sample may be required, and a flow rate of 1 l./min is suitable for a few minutes sampling time. Our impactors with this flow rate (Delcron Inc, Powell, Ohio, U.S.A.) have a 50% cutoff diameter for the final impaction stage of 0.25 μm , and it is therefore possible to investigate particle size dependence of concentrations in a range where respiratory deposition and other response may be a significant function of aerodynamic diameter. Table 1 presents the aerodynamic cutoff diameters for equivalent unit density spheres for five impaction stages and 0.4 μm Nuclepore backup filter. Retention of smallest particles by Nuclepore filters is theoretically predicted (SPURNY, 1972) and measured (LIU and LEE, 1976) to be high because of impaction of larger

TABLE 1. PARTICLE COLLECTION CHARACTERISTICS FOR 5-STAGE CASCADE IMPACTOR*

Stage	1	2	3	4	5	6†
Size range (μm)‡	>4	4–2	2–1	1–0.5	0.5–0.25	<0.25

* When operating at 1.05 l./min air flow regulated by stage 5 limiting flow orifice.

† Backup Nuclepore filter with 0.4 μm pore diameter.

‡ Range in micrometers between 50% cutoff diameters between successive impaction stages for equivalent unit density spheres. Stage 1 upper limit to particle size is determined by sampler entrance design.

particles and Brownian diffusion and sticking of smaller particles on the filter surface and pore walls. For 0.4 μm pores and aerodynamic diameters <0.02 μm the filter efficiency is >99%, but in the range 0.02–0.4 μm the efficiency is less owing to lower diffusion rates for larger particles. At reduced pressure, however, uptake of particles by impaction, and to a lesser extent by diffusion, is enhanced in comparison with filter operation at atmospheric pressure. When using 0.4 μm filters at 0.15 atm in the reduced pressure zone following the fifth impactor stage, we have calculated the minimum efficiency to be >85% for particles of 0.05–0.06 μm diameter.

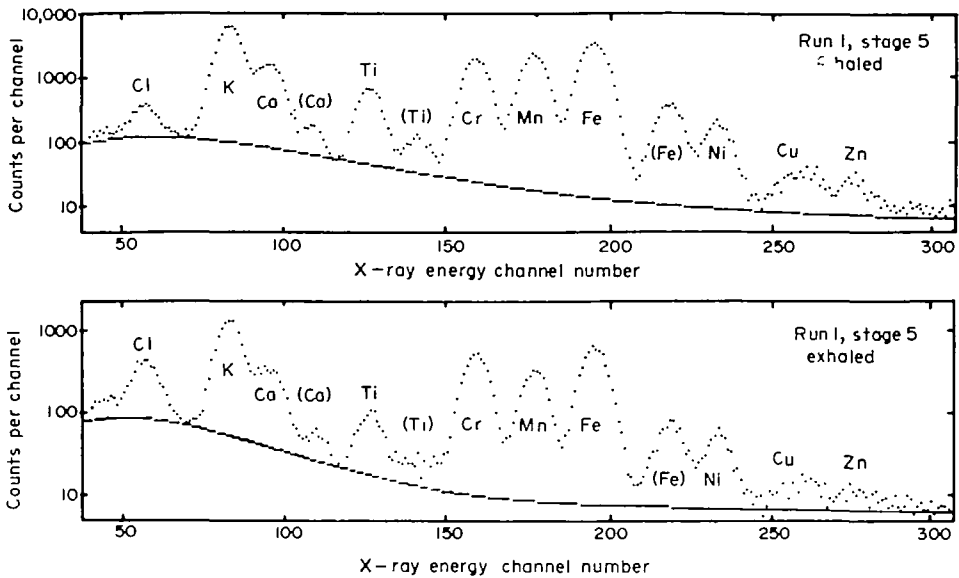


FIG. 1. X-ray spectrum of PIXE from 0.25–0.5 μm fraction of inhaled (top) and exhaled (bottom) aerosol particles from Run 1. A relative depletion of Mn relative to Cr and Fe in the exhaled aerosol is apparent from the X-ray counts recorded. Exhaled sample proton bombardment intensity was twice that for inhaled. Relative values of corresponding counts per channel exhaled/inhaled represent twice the relative elemental abundances in the two samples.

A visual impression of the information contained in PIXE analysis of aerosol particle size fractions is given by Fig. 1. Here are shown analyses of 0.25–0.5 μm diameter particle fractions, each collected from 6 l. of air by impaction stage 5 for inhaled and exhaled aerosol of Run 1 described further below. Ten elements are well resolved by computer methods from these spectra, such, for example, that 0.17 μg of Fe and 0.024 μg of Ni can be reliably calculated to be in the exhaled aerosol sample.

Metals contained in the aerosol generated by welding operations are concentrated in a very small particle size range, and they may be taken up deep in the respiratory tract. In Fig. 2 we present a comparison of the distribution of Fe with particle size for welding Run 3 and for natural aerosol sampled in two locations: in the city of St Louis, Missouri (AKSELSSON *et al.*, 1975) and in rural north Florida (JOHANSSON *et al.*, 1976). Atmospheric Fe occurs mostly on large particles because of its origin in low temperature mechanical dispersion of soil and urban or industrial dust. Welding fume aerosol, on the other hand, is generated at high temperatures, and vapour condensation as well as high energy dispersion processes are operative, so that Fe occurs mostly on fine particles.

EXPERIMENTAL

Aerosol particles were sampled in an experimental arrangement similar to that employed in an earlier study (DESAEDELER and WINCHESTER, 1975), shown schematically in Fig. 3. The present experiments were conducted in an indoor atmosphere of

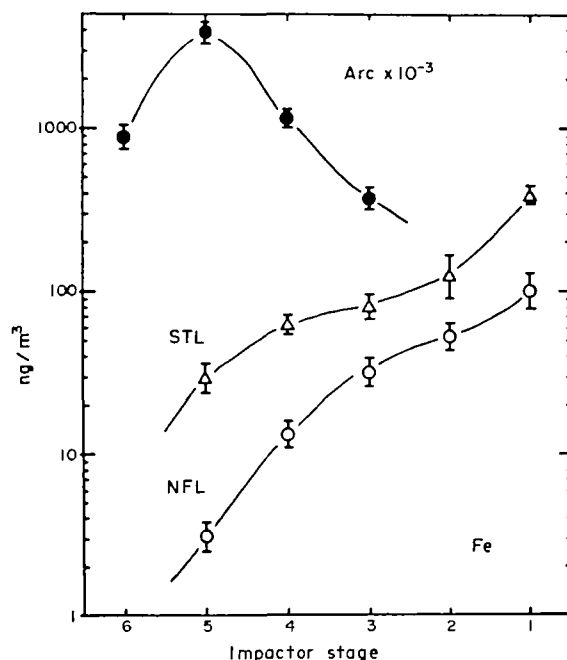


FIG. 2. Comparison of Fe concentrations in arc welding aerosol, sampled for 30 s close to the welder during welding, with average ambient air samples from St Louis, August 1973 (STL), and north Florida, July 1973 (NFL) at rooftop locations. Welding aerosol Fe is found predominantly in the 0.25–0.5 μm dia range at a concentration 10^5 times greater than St Louis concentration in the same size range.

a machine shop contaminated by fumes from arc welding operations. The sampling devices, placed on a table about 1 m from where welding was carried out in the physics shop, Florida State University, were positioned so as to sample both the air which the subject inhaled through his nose and that which he exhaled through his mouth. The exhaled air flow entered a box heated to just above body temperature, through a plastic bag which served as a ballast permitting pulsating breathing but constant flow sampling, and into a preheated cascade impactor. In addition to the impactors operating at 1 l./min., shown in Fig. 3, we also included duplicate larger impactors operating at 12 l./min in parallel for another chemical test, and the total air flow rate of 13 l./min was easily maintained by the subject breathing normally. Alternatively, an air leak arrangement might be provided to permit comfortable breathing conditions and a 1 l./min sampling rate.

It is important to provide for a means to warm the sampling apparatus for exhaled air to above body temperature so as to avoid condensation of water vapour on wall surfaces and particles. As will be shown below, our procedures, although still preliminary and imperfect, indicate that accurate prediction of respiratory response to particles as a function of their size in the atmosphere requires measurements carried out with temperature and humidity control in the sampling apparatus, especially for particles with pronounced hygroscopic character.

Each cascade impactor was equipped with paraffin-coated polystyrene film impaction surfaces, supported by 25 mm dia. glass slides, and a 0.4 μm Nuclepore backup

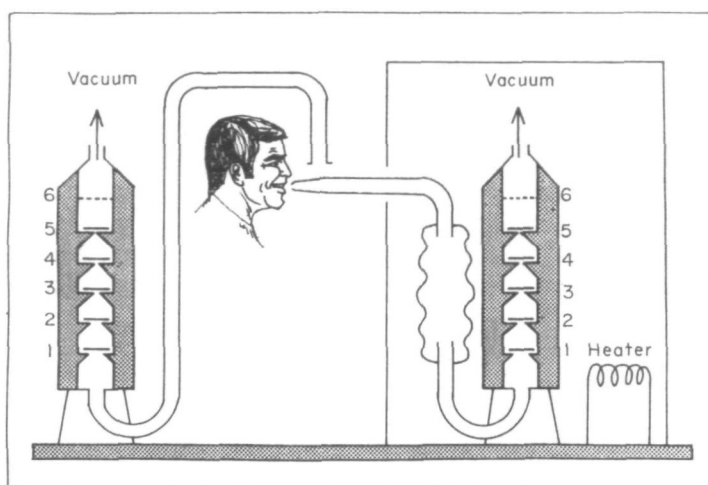


FIG. 3. Schematic diagram of sampling arrangement for aerosol inhalation experiments.

fine particle filter. After sampling, these samples were removed from the impactor, separated from the glass support surfaces, and mounted on frames for proton irradiation. They were then placed in turn in a vacuum chamber connected to the Van de Graaff accelerator, and X-radiation was measured by a Si(Li) detector during short exposures to the proton beam. Spectra similar to those shown in Fig. 1 were stored on magnetic tape and later resolved by computer programs so that a quantitative analysis was made for each element in the sample.

Elements lighter than sulphur cannot ordinarily be determined by this technique because their soft X-rays are absorbed appreciably in the sample, and absorption corrections are difficult to make accurately. However, heavier elements over a broad range can be determined by K X-ray measurement, in practice from S to about Mo, or by L X-ray measurement, in practice from about Cd to Pb. Complex mixtures of elements can be resolved, providing the relative abundances of elements with similar X-ray energies are not so different that X-ray counting statistics becomes a limiting consideration. By inspection of Fig. 1, it is seen that the major elemental constituents produce distinctive X-ray components, but some, e.g. Cu and Zn, may be limited by the statistical variation of counts recorded in the region of their characteristic X-rays. More precise data for these elements could be obtained by longer proton irradiations.

A typical PIXE analysis report is shown in Table 2, for the exhaled sample portrayed in Fig. 1. The report indicates the weight found for each element, in nanograms, with an indication of two standard percentage deviations. The first of these, the ratio error, is due to the fit of the pulse height spectrum, based on uncertainties due to counting statistics and interferences. This error is important in computing uncertainties in element ratios within each sample and comparing these ratios in different size fractions or experimental runs. The second, the error in concentration of the element in the air which was sampled, includes uncertainties in system parameters and calibrations which approximately cancel in element abundance ratio calculations but which are significant in reporting absolute concentrations of elements contained in the aerosol samples. Sampling errors, such as due to contamination or imperfect impactor

TABLE 2. PIXE ANALYSIS REPORT: RUN 1, STAGE 5, EXHALED SAMPLE

	Amount (ng)	Standard deviation (%)		Detection limit (ng)
		Ratios	Concentrations	
S	5.9	14	31	1.5
Cl	29.8	2	19	1.2
K	105.0	1	18	0.7
Ca	20.1	2	18	1.3
Ti	12.2	3	16	0.6
V	0.7*	32	34	0.9
Cr	96.6	1	13	0.2
Mn	63.0	2	15	1.7
Fe	166.0	1	13	1.1
Ni	23.8	3	15	1.0
Cu	3.8	12	17	1.3
Zn	4.4	12	18	1.4
Pb	6.6*	29	37	9.8
Br	—†	—	—	4.0
Sr	—†	—	—	8.7

* These elements are near the detection limit.

† These elements are well below the detection limit.

performance, are additional but in the present experiments are considered to be small compared to the absolute analytical error listed. The final column of Table 2 indicates the detection limit, in nanograms, caused by counting statistics and other features of the X-ray spectrum. Most elements are reported at levels well above the detection limit, but two, V and Pb, are slightly below and not significant.

The experimental conditions for the breathing experiments in this investigation are indicated in Table 3. Four preliminary experiments were carried out during a three day interval, each lasting a few minutes, and a series of five experiments were then performed over a 6-h interval. Three different persons served as subjects to provide the exhaled aerosol samples, and the concentration of aerosol particles in the unventilated machine shop atmosphere ($6 \times 10 \times 3 \text{ m}^3$) was carried by employing from one to four welding rods per experiment. Normally each experiment began with a welder striking an arc and burning the rods in contact with a piece of steel, a wait of several minutes to allow the haze to diffuse to a uniformly low intensity in the shop, then a sampling of several minutes of both the inhaled air, by means of a tube held near the subject's nose and leading to one impactor, and the exhaled air, expelled through the mouth into the heated arrangement shown in Fig. 3.

Temperature and humidity conditions in the machine shop atmosphere and the sampling apparatus were only approximately known and are useful for qualitative rather than quantitative interpretation of some of the data. Humidity in the shop was lower than outdoor air because of several °C space heating. The impactor for exhaled air sampling was preheated to about 50°C before Runs 1, 2, 3, 4, 5, and 9, but cooling during sampling or possible temperature increases in the enclosure due to its electric heater, especially in the series of Runs 5, 6, 7, 8, were not monitored. The results obtained in this study suggest that monitoring of both temperature and humidity at several strategic points in the experimental arrangement are desirable in continuation studies.

TABLE 3. EXPERIMENTAL CONDITIONS OF THE BREATHING EXPERIMENTS*

Run	Date/time	Air volume sampled (l.)†	Subject	Number of welding rods used
1	28/1400	6	RA	4
2	29/1415	5	RA	4
3	29/1600	5	RA	4
4	30/1455	5	RA	3
5	31/0900	4	GD	4
6	31/1100	4	GD	2
7	31/1040	7	GD	1
8	31/1140	5	JW	1
9	31/1440	5	RA	1

* All experiments were performed 28–31 January 1975 in a machine shop after a 1–5 min operation of welding a piece of steel, using Hobart stainless steel arc welding rods, type 308, of rated composition: C 0.07%, Si 0.7%, P 0.03%, S 0.02%, Cr 20%, Mn 2%, Ni 10%, Fe remainder.

† The data also represent breathing time in minutes because of constant sampling rate of 1 l./min. A steady rate of about 10 breaths per minute was maintained, and breathing commenced approximately 2–10 min after completion of burning the arc welding rods, to ensure that the aerosol was visibly well-mixed in the machine shop.

RESULTS

Figure 4 reveals that the ratio of two elements, Fe/Cr, is quite constant in the inhaled aerosol over the series of nine runs carried out and is generally invariant with particle size over the four size fractions analysed. The ratios in the exhaled samples show no significant differences from the inhaled. We have found that other element ratios also generally show consistency compatible with experimental error from run to run and indicate reproducibility in the experimental conditions. Similarity of the interelement ratios between inhaled and exhaled aerosol and in different particle size ranges suggests that the elements are components of the same particles.

Figures 5(a) and 5(b) present for the nine runs the ratio of concentrations in exhaled to inhaled air for Fe and for Cl, two elements showing contrasting behaviour in our experiments. For Fe, the smallest stage 6 particles are markedly depleted in exhaled air compared to inhaled, the intermediate stage 5 particles are usually only moderately depleted, and the larger stage 4 particles are found at higher concentrations in exhaled air than in inhaled. Because of the largest amounts of Fe being initially in stage 5 particles, as shown in Fig. 2, the results suggest a shift in particle size by exposure to the lung environment, such as by water vapour condensation, leading to collection on stage 4 of exhaled particles which were inhaled in the next smaller size range. For Cl, the data show considerable irregularity from run to run, even though exhaled concentrations are generally lower than inhaled, but in most cases the progressive change with increasing particle size is not seen. These results indicate that changes in particle size, e.g. by water vapour condensation, may occur in addition to particle loss through deposition during breathing.

Figures 6(a) and 6(b) show the distributions with particle size for eight elements in inhaled and exhaled air found in Run 5 and Run 8, two runs in a single time series, indicated in Table 3. For Run 5, typical in many respects to most other runs of this investigation, a shift in particle size distribution from smaller to larger particles is

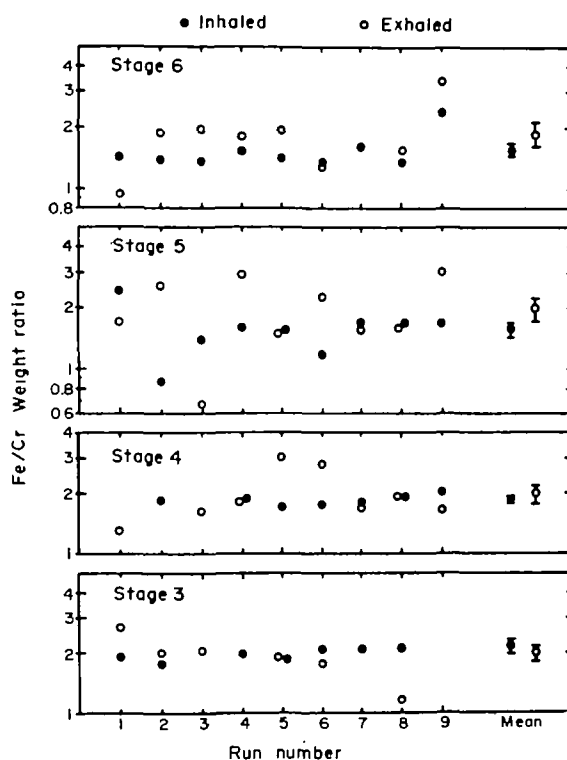


FIG. 4. Weight ratio Fe/Cr for each particle size fraction found in the series of nine experimental runs. Variations are within random uncertainties of the measurement.

found for all elements other than Cl. Runs 1, 3, and 9 show similar size shifts, and Runs 2, 4, 6, and 7 show smaller shifts; Run 8, using a different breathing subject, shows no apparent shift in particle size distribution for the metallic elements. We regard these shifts as an important indication of the complexity of the system being investigated, and the variability in the magnitude of the shifts suggests dependence on details of the experimental conditions which were allowed to vary. It is premature to ascribe any variability to differences in human subjects or breathing patterns, although these should be examined further.

Table 4 presents evidence that two distinct populations of aerosol particles are present in the welding experiments. These two populations are defined by the coherence of the interelement ratios, viz. one group of particles with fixed proportions of Cr, Mn, Fe, and Ni and the other group with fixed proportions of K, Ca, and Ti. Within each group there is no evident variation of element ratio with particle size for the inhaled aerosol, nor is there a resolvable difference between inhaled and exhaled ratios within a particular size fraction. When representative element members of each group are compared with each other, however, there is a clear variation of element ratio with particle size and the exhaled ratio is different from the inhaled ratio for the two largest particle sizes, impactor stages 3 and 4. These results suggest that the two aerosol populations, with different initial size distributions, have different respiratory response even for particles initially in the same size fraction.

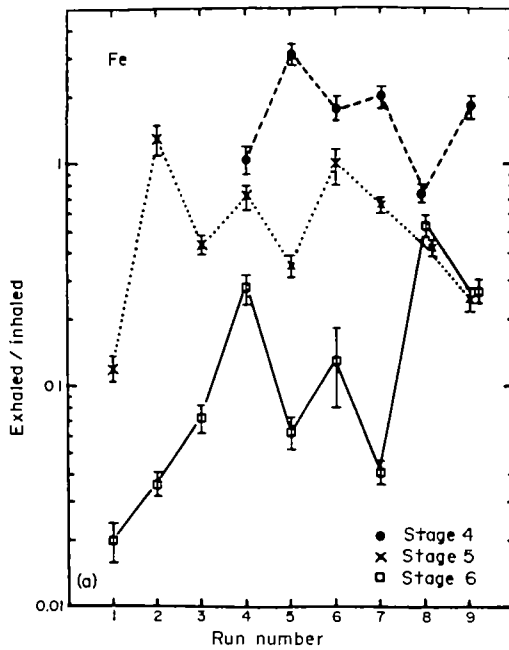


FIG. 5(a). Concentration ratio exhaled/inhaled for Fe in three particle size fractions found in the series of nine runs. Lower concentrations are found in exhaled air compared with inhaled for stages 6 and 5, but greater concentrations are found for stage 4.

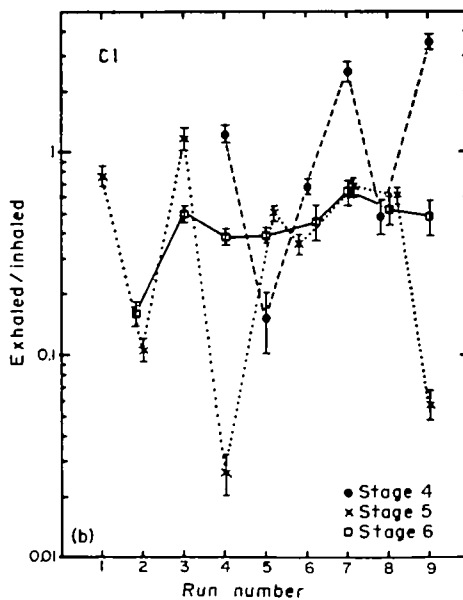


FIG. 5(b). Concentration ratio exhaled/inhaled for Cl in three particle size fractions found in the series of nine runs. Lower concentrations are generally found in exhaled air compared with inhaled, without systematic relationship to particle size.

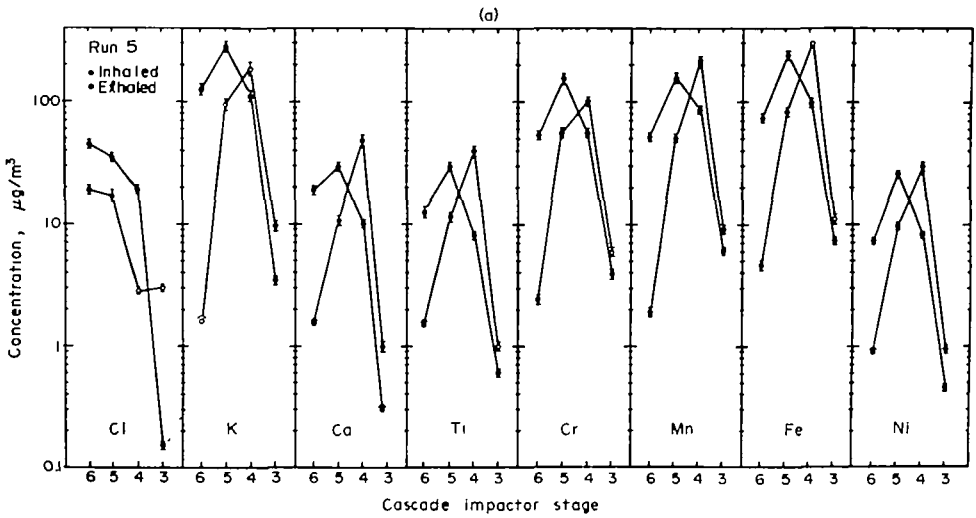


FIG. 6(a). Particle size distributions of trace elements in inhaled and exhaled air for Run 5. Metallic elements generally show a shift toward larger particle sizes in exhaled air.

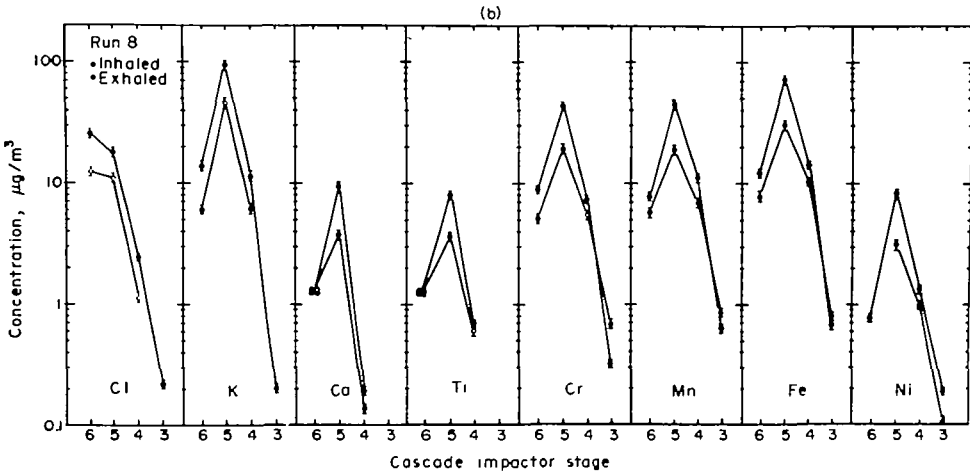


FIG. 6(b). Particle size distributions of trace elements in inhaled and exhaled air for Run 8. No systematic shift toward larger sizes is found, and exhaled concentrations are generally lower than inhaled.

We have also calculated for each element the sum of masses on all stages for each inhaled and exhaled sample and the ratio of corresponding exhaled to inhaled amounts for each run. The results are summarized in Table 5. (Run 2, in which exhaled stage 4 was not analysed, is not included). The ratio averages for all elements range from 0.5 to >0.9 for the different runs, indicating total apparent respiratory deposition of 50% to <10%. Coherence among Fe, Mn, and Ni is exceptionally close within each run, and departures of the other elements may reflect differences in chemical composition which are important in determining deposition efficiency. Statistical analysis of

TABLE 4. ELEMENT RATIOS IN INHALED AND EXHALED AEROSOL*

Elements	Impactor stage							
	6		5		4		3	
	Inhaled	Exhaled	Inhaled	Exhaled	Inhaled	Exhaled	Inhaled	Exhaled
Fe/Mn	1.7 ± 0.5	2.1 ± 0.5	1.62 ± 0.13	2.0 ± 0.7	1.20 ± 0.11	1.45 ± 0.12	1.3 ± 0.4	1.21 ± 0.18
Fe/Cr	1.6 ± 0.3	1.8 ± 0.7	1.6 ± 0.4	2.0 ± 0.8	1.85 ± 0.09	2.0 ± 0.6	2.2 ± 0.5	2.0 ± 0.5
Mn/Cr	0.93 ± 0.06	0.8 ± 0.3	0.96 ± 0.19	1.1 ± 0.4	1.55 ± 0.01	1.4 ± 0.5	1.75 ± 0.2	1.62 ± 0.2
Ni/Fe	0.1 ± 0.02	~0.06	0.11 ± 0.01	0.11 ± 0.01	0.09 ± 0.01	0.10 ± 0.01	0.08 ± 0.02	0.09 ± 0.01
Ni/Mn	0.15 ± 0.03	~0.12	0.18 ± 0.02	0.23 ± 0.08	0.10 ± 0.01	0.15 ± 0.02	0.1 ± 0.02	0.12 ± 0.01
Ni/Cr	0.14 ± 0.02	~0.11	0.17 ± 0.05	0.22 ± 0.08	0.16 ± 0.01	0.19 ± 0.05	0.17 ± 0.04	0.20 ± 0.05
Ca/K	0.12 ± 0.03		0.11 ± 0.03	0.16 ± 0.11	0.10 ± 0.03	0.19 ± 0.1	0.10 ± 0.04	0.4 ± 0.4
Ti/K	0.1 ± 0.02		0.10 ± 0.03	0.15 ± 0.08	0.07 ± 0.02	0.12 ± 0.05	0.15 ± 0.02	0.13 ± 0.05
Ti/Ca	0.9 ± 0.3		0.94 ± 0.07	0.93 ± 0.12	0.82 ± 0.19	0.72 ± 0.2	1.0 ± 0.9	0.6 ± 0.4
Mn/Ti	4.4 ± 0.8		4.9 ± 0.7	4.8 ± 0.5	13.0 ± 4.0	7.0 ± 3.0	13.0 ± 4.0	5.0 ± 3.0
Fe/Ca	6.0 ± 2.0		7.5 ± 1.6	7.6 ± 1.6	12.0 ± 4.0	6.0 ± 1.2	26.0 ± 13.0	6.0 ± 4.0

* Average values over nine experiments for inhaled and exhaled element weight ratios, with approximate 2σ (95% confidence) uncertainties, as a function of particle size (impactor stage).

TABLE 5. EXHALED/INHALED WEIGHT RATIOS OF SAMPLES SUMMED OVER STAGES 3-6

	1	3	4	Run number		7	8	9	mean	s.d.
				5	6					
Fe	0.51	0.78	0.67	0.96	1.05	0.83	0.49	0.55	0.73	0.21
Mn	0.53	0.84	0.63	0.93	0.99	0.85	0.51	0.50	0.72	0.20
Ni	0.54	0.82	0.68	0.94	0.97	0.93	0.42	0.57	0.73	0.21
Cr	0.52	0.85	0.47	0.62	0.58	0.90	0.51	0.48	0.62	0.17
Ti	0.52	0.75	0.71	1.03	1.06	1.17	0.47	0.67	0.80	0.26
Ca	0.48	0.80	1.08	1.03	1.20	0.93	0.39	0.62	0.82	0.29
K	0.49	0.84	0.34	0.56	0.52	0.87	0.49	0.46	0.57	0.19
Cl	0.42	1.20	0.30	0.41	0.44	0.92	0.55	0.56	0.60	0.30
mean	0.50	0.86	0.61	0.81	0.85	0.92	0.48	0.55		
s.d.	0.04	0.14	0.25	0.24	0.29	0.11	0.05	0.07		

of inter-element relationships shows that the largest differences in apparent deposition efficiency (1 - exhaled/inhaled weight ratio) are between K and the Ca-Ti pair, especially in runs 4, 5 and 6. For all runs averaged together, the deposition efficiencies of the eight elements range from 0.18 for Ca to 0.43 for K and at most are significant at the 95%, though not at the 99%, confidence level.

The exhaled/inhaled deficit on stage 6, which can be considerably greater than on larger particle stages as exemplified by Fig. 5, may be the result of a combination of deposition and shift to larger sizes. However, since the size distribution of welding aerosol shows a maximum around stage 5, a shift from smaller sizes into this range causes a relatively smaller concentration increase than the stage 6 decrease. Since deposition efficiency may be substantial for $<0.25 \mu\text{m}$ particles (TASK GROUP ON LUNG DYNAMICS, 1966; DESAELEER and WINCHESTER, 1975), a major part of the overall observed $<10\%$ to 50% deposition may be preferential deposition of the smallest particles. This could be confirmed by use of an aerosol initially with mainly $<0.25 \mu\text{m}$ particles and preferably not so hygroscopic as we have found the welding aerosol to be.

DISCUSSION

We have attempted in this investigation to test the feasibility of directly measuring exhaled welding fume aerosol composition in order to determine the type and extent of aerosol modification upon exposure to the human respiratory tract. Because of the very great sensitivity of PIXE analysis, samples could be as small as a few litres of normally contaminated air from indoor working conditions. In contrast, the not nearly so sensitive method of instrumental neutron activation analysis (DAMS *et al.*, 1970) which has been applied to aerosol characterization for inhalation studies (HEWITT, 1972; HEWITT and HICKS, 1973) may require several cubic metres of air with similar heavy metal concentrations. Thus, we could carry out our experiments in 4-7 min of breathing time in an actual welding shop atmosphere, and human subjects could be conveniently used.

Metallic constituents in the inhaled aerosol were found to be generally most abundant in the particles collected by cascade impactor stage 5, aerodynamic diameter $0.25-0.5 \mu\text{m}$, with lesser amounts on filter stage 6, $<0.25 \mu\text{m}$. This result is consistent with our earlier observation using a low pressure impactor (AKSELSSON *et al.*, 1974).

It is also consistent with the findings of HEWITT (1972), where he measured the median diameter of welding aerosol by electron microscopy to be $0.15 \mu\text{m}$ (σ_g 1.7), providing we realize that equivalent aerodynamic particle diameter varies directly with the square root of the particle density and that our measurement is of elemental mass per finite particle size interval, not particle size itself.

For welding fume studies, a further advantage of high analytical sensitivity and being able to sample very small air volumes is that a cascade impactor can be employed which sorts very small particle size classes. Thus, at 1 l./min flow rate a size cut at $0.25 \mu\text{m}$ aerodynamic diameter can be obtained with our impactors, and this cut reveals the preponderance of elemental mass above that diameter, not below. If our analytical method required larger air volumes, commercially available cascade impactors would provide minimum size cuts at much greater particle sizes; for example, using a larger impactor of similar design but operating at 12 l./min, the final filter stage collects all particles smaller than $0.5 \mu\text{m}$ aerodynamic diameter, and the important information contained in the metal contents of the greater and less than $0.25 \mu\text{m}$ fractions would be lost.

Our initial results presented in this report indicate the respiratory response to aerosol inhalation to be a complex process, and the combination of PIXE and its compatible particle sampling methods makes the study of some of these complexities quite feasible. Because only 1 l./min flow rate is required, there is the possibility of studying the aerosol as a function of phase of exhalation by air switching in a multiple impactor sampling apparatus. Moreover, because PIXE analysis requires only a few minutes per analysis and the results are obtained immediately, rather large data sets can be acquired to investigate the complexities in detail. The technique can be applied to atmospheres normally contaminated under indoor working conditions with low concentrations of trace metals and makes use of aerosol particles as they are encountered without need for special tracer methods.

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