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HISTORICAL RESEARCH REPORT

Research Report TM/87/06
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Johnston AM, Hughson GW, Vincent JH, Jones AD

WORLD HEALTH ORGANISATION
COLLABORATING CENTRE
FOR OCCUPATIONAL HEALTH

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FINAL REPORT ON PROJECT 534

A PRELIMINARY INVESTIGATION OF AEROSOL EXPOSURE ASSOCIATED WITH OPERATION OF A HAND-HELD ELECTRODYNAMIC CROP SPRAYER

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Final Report on Project No.534
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CONTENTS

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SUMMARY

The principal aim of this investigation was to relate the amount of pesticide material inhaled by an operator to that delivered from his hand-held electrodynamic crop spraying device. The 'operator' in this case was a full size tailor's mannequin fitted with a nasal filtration unit which was connected to a 'breathing machine'. Determination of the amount of material inspired during a given spraying period was by fluorimetric analysis of the amount of the Uvitex-B tracer dye contained in the formulation collected on the nasal filter. Both calm and moving air situations have been investigated, the latter in a large wind tunnel. In calm air the inspired material amounted to about 10^{-4}% of the total volume sprayed. In moving air (wind speed, 3 ms^{-1}), the amount of material inspired during downwind spraying (the recommended orientation) was too low to be assessed by the methods used, but was established as being less than 2.5×10^{-6}% of the total volume sprayed in any period.

Assessment of the correlation between the results obtained for several different types of personal sampling devices and the inspired volume was also undertaken. The results in terms of the mean [and standard deviations] of the sampling ratio (amount collected on sampler/amount inspired) for the three sampler types tested in calm air were 0.74[0.25], 0.60[0.26] and 0.30[0.17] respectively. In the case of downwind spraying, the amounts collected were again too low for analysis.

Data relating to the distribution of dermal exposure was also collected during spraying by clothing the dummy operative in a two-piece overall. These overalls were despatched for analysis elsewhere and the assessment of exposure by this route does not form part of the work reported here.

A PRELIMINARY INVESTIGATION OF AEROSOL EXPOSURE ASSOCIATED WITH OPERATION OF A HAND-HELD ELECTRODYNAMIC CROP SPRAYER

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1. INTRODUCTION

Successful early trials using a recently-developed hand-held electrodynamic crop sprayer (ECS) suggest that the device may achieve widespread usage with a range of pesticide materials. Inevitably, however, some of these chemicals will have an associated operator exposure risk through a combination of dermal contamination and inhalation of sprayed droplets. The objective of the studies reported here was, therefore, to obtain first estimates of the inhalation exposure levels relating to a range of simulated spraying conditions. The necessity for work of this nature arises mainly from the fact that the sprayed droplets carry very high electric charge levels. Whilst this gives clear advantages in terms of plant coverage, it also makes the airborne behaviour of such a cloud in terms, for example, of the ability of particles to be inspired by human operators during breathing, very difficult to predict from previous studies using 'normally' charged aerosols.

The experience of the Institute of Occupational Medicine in the definition and measurement of inspirability of mineral dust (VINCENT and ARMBRUSTER, 1981; MARK et al., 1985; MARK et al., 1986) and in the measurement of the levels (JOHNSTON et al., 1985) and effects (VINCENT et al., 1981; JONES et al., 1982) of electric charge carried by airborne dust particles placed us in an excellent position in terms of both experience and facilities to undertake some preliminary studies relating to quantification of operator exposure and means of assessment. The work was carried out between July and December 1986.

2. AIMS OF THE STUDY

There are two aspects to the determination of the danger to health associated with exposure to an aerosol; firstly, the amount received by a particular part of the body and secondly the associated toxicity. In the case of the sprayed pesticides, the organs of main interest are the lungs and the skin. In this short, preliminary study we concentrate mainly on the former, the principal objective being to determine the amount of formulation received by the lungs as a fraction of the total amount dispersed by the ECS during a given spraying period.

By clothing the operator in disposable overalls, however, data have also been collected which may be analysed in terms of the total dermal exposure per unit sprayed and its regional variation. Previous experience with other types of sprayer has shown that, in general, it is this dermal route which contributes most to overall systemic uptake. Furthermore, the establishment of a relationship between dermal and inhalation exposure may have further consequences for the assessment of the latter in the field situation. However, analysis of the dermal exposure data which was collected does not come within the scope of this report.

Also included in the study is an examination of the efficiency of some personal sampling devices in terms of the relationship between the amounts of material collected by them and the amount inspired. Both calm and moving air conditions have been investigated, the former in a small laboratory at our Bush House outstation and the latter in the large wind tunnel at our central laboratory.

It should be stressed that a complete characterisation of these various aspects would require a much more extensive study than that carried out here. We have restricted our objectives, therefore to the development of suitable experimental systems, crystallisation of the problems, and obtaining some preliminary answers for a restricted range of conditions.

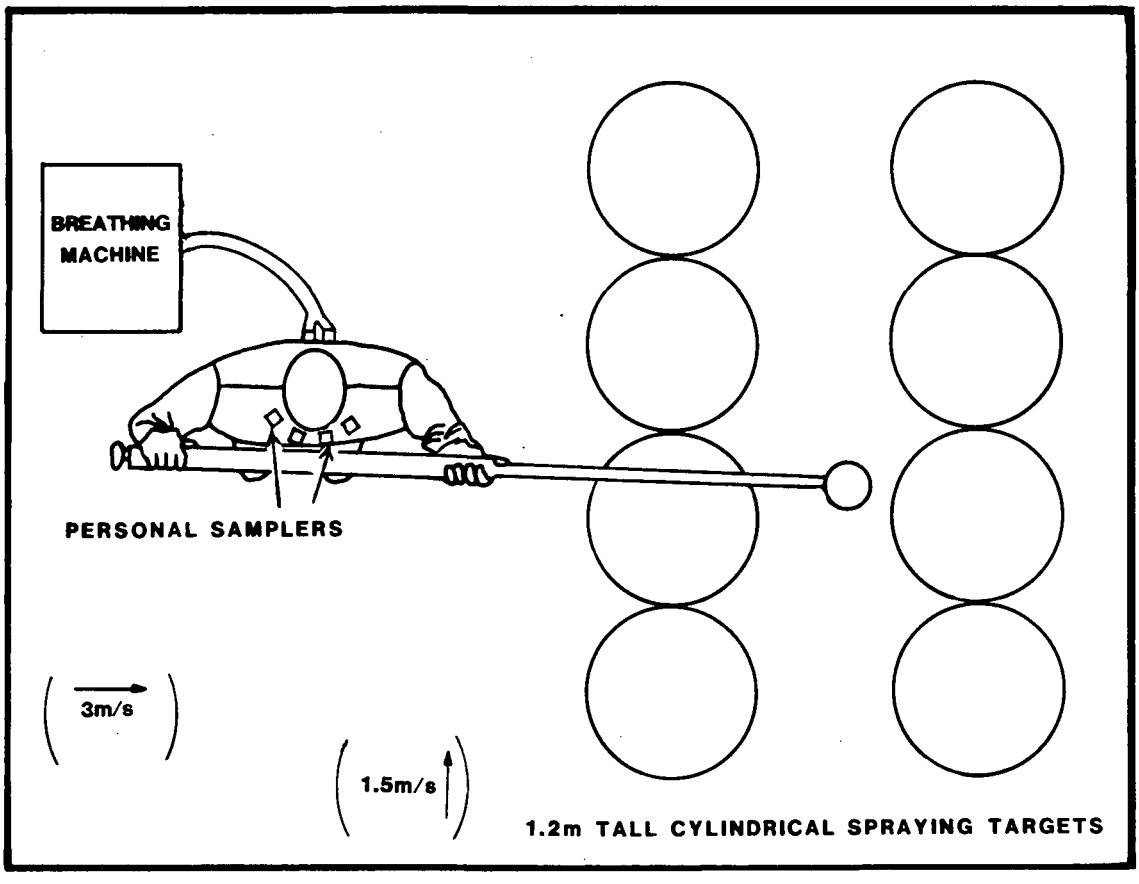

Figure 1 The basic experimental set-up seen from above. The height of the targets was 120 cm and the distance between rows, 85 cm. The wind directions shown refer to later work in the wind tunnel.

3. EXPERIMENTAL FACILITIES AND SET-UP

3.1 Calm-air experiments:

Experiments were carried out at two quite separate locations. The first series of 'calm air' runs were based in a small (25 m^2 floor area) laboratory at our Bush House outstation. The basic experimental set-up is shown in Figure 1 and in Photograph 1. The ECS was attached to the chest of a tailor's mannequin using plastic cable ties. The spraying targets (to simulate actual crops) were made up with corrugated cardboard cylinders covered in polythene with an outside covering consisting of a tube of stockinette material earthed at the top by a wire running down through the cardboard tube. The floor of the whole spraying area was covered by a heavy paper material (normally used as carpet backing) beneath which was a layer of aluminium foil which was connected directly to earth.

In accordance with normal spraying practice the nozzle was positioned between the two rows at a height of about 40 cm above the 'plants' and spraying carried out in accordance with the manufacturer's recommendations. The relevant distances were, therefore

Nose to nozzle	1.80 m
Nozzle to ground	1.55 m
Distance between rows	0.85 m

Several trials in which the liquid drops (with the sprayer switched off) were collected in a measuring cylinder showed that the flowrate from the ECS was about 1.6 ml min^{-1}.

A 47 mm diameter brass filter holder (Figure 2) was positioned on a moulded base within the head of the mannequin, the skull having been sawn through and hinged to allow access. Two copper tubes projecting from the filter holder opened at the nostrils. The filter holder was attached to a 'breathing machine' through two tubes, one of which opened behind the glass

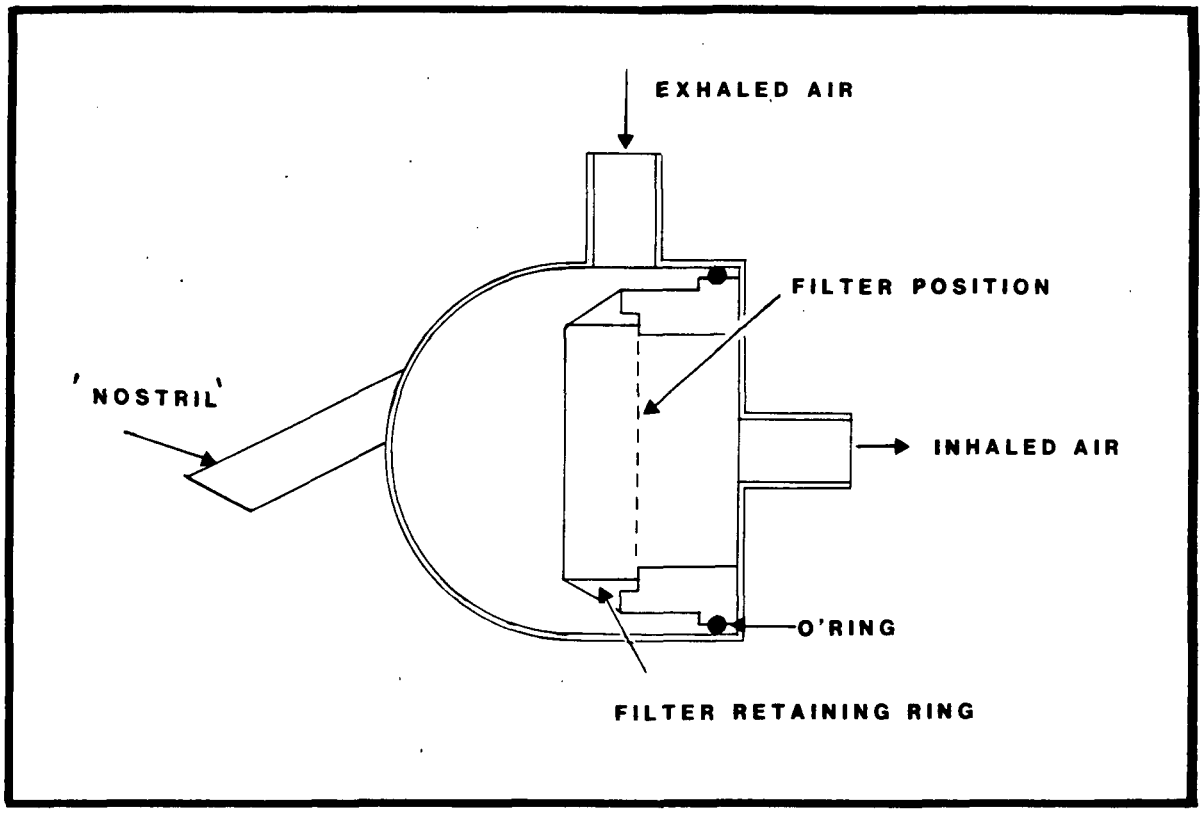

Figure 2 A vertical section of the nasal sampler note that the nostril tubes point towards the top of the sampler so that it is likely that any large particles will be inertially deposited here.

fibre (Whatman GF/A) filter and provided the inward breathing stroke and one which opened in front of the filter to allow exhalation. The purpose of the breathing machine was to provide a realistic simulation of inhalation and exhalation through the nose of the mannequin. The breathing pattern was set at 16 breaths per minute and 25 l/min, which corresponds (ASTRAND and RODAHL, 1977) to an energy dissipation of ~500W (corresponding, in terms of activity, to walking at 4 mph).

Four personal sampling devices, typical of those used routinely in occupational hygiene in the United Kingdom for sampling 'total' aerosol, were also mounted on the chest of the mannequin during each run. These were, an open-faced plastic Gelman sampler, two carbon-fibre cowled samplers (Millipore), and an aluminium-bodied '7-hole' sampler (HSE 1983). Each was operated at a sampling flowrate of 2 l min^{-1} through 25 mm diameter Whatman GF/A filters. Their positions on the chest were as shown in Figure 3 and in Photograph 2 for all the runs described in this report.

In order to assess the distribution of potential dermal deposition the mannequin was clothed in a two-piece Tyvek overall which was changed between runs. After photographing, (see for example Photograph 3) the suits were turned inside out (to prevent cross-contamination), rolled up and packed individually in sealed polythene bags. These were despatched elsewhere for further analysis, the results of which will not be given or discussed here.

3.2 Wind tunnel experiments

A further series of experiments has been carried out in the large open-cycle, open-jet wind tunnel at the Institute of Occupational Medicine. This facility has a working region of cross-section 1.5 m x 2.5 m and length about 4 m. The basic experimental set-up was similar to that described previously except that, in this case, the height of all components in the system had to be reduced by 0.4 m to bring the sprayer and head of the mannequin into the working region of the tunnel. Two

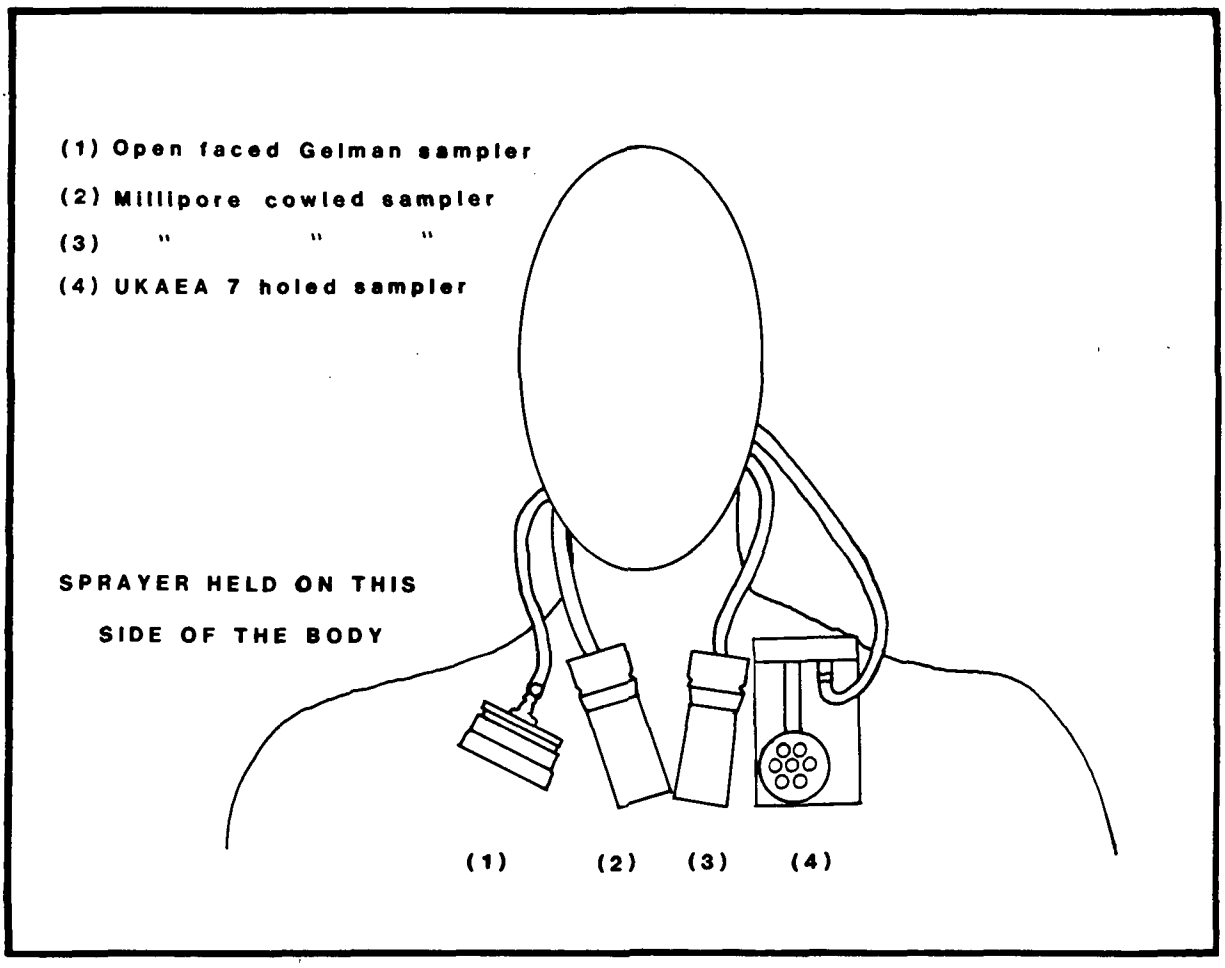

Figure 3 The positioning of the personal samplers on the mannequin. This was the same in all runs. In the wind tunnel experiments, however, the cowled sampler adjacent to the open face Gelman sampler was replaced by the IOM sampler.

basic orientations were investigated: firstly, the 'downwind' spraying orientation recommended by the manufacturer (see Figure 1); and secondly, the situation in 'calm air' where the operator is walking steadily (simulated by placing the mannequin facing into a wind of 1.5 ms^{-1}). All other details were as previously described. Two runs with no air movement were also performed for comparison with the results obtained at Bush House. In addition to the four personal samplers described above, some experiments were also conducted using the new IOM personal inspirable dust sampler (MARK et al., 1986)

Photograph 4 shows the effect of a 1.5 ms^{-1} wind on the spray close to the nozzle. It is clear that even at this low velocity, moving air is likely to have a strong influence on the distribution of the airborne droplets.

3.3 Analytical method

The spray liquid used in the test was supplied by the manufacturer of the ECS, and consisted of a pesticide formulation with the fluorescent tracer UVITEX-OB (CIBA-GEIGY) substituted for the active ingredient. The aerosol produced by the ECS was collected onto Whatman GF/A glass microfibre filters, both after inspiration through the nose of the mannequin and after sampling by the various personal samplers mounted on the torso of the mannequin. The formulation was extracted from the filters by soaking in a solvent (HPLC Grade Acetone/n-hexane mixture). The content of the UVITEX-OB tag was determined by fluorescence spectrophotometry (Perkin Elmer Model 1000M fluorimeter, with emission filters R.UV 376 12 and excitation filter B.40 4357).

To prepare a standard solution, a sample of the UVITEX-tagged formulation (25 µl), from the bottle in use at the time, was dissolved in the acetone n-hexane (1:4 v/v) mixture and the volume adjusted to 25 ml in a standard flask. This stock solution was dissolved by a further factor of 1000 to give a working dilution of 1 µl of formulation per litre of solvent.

The filters taken from the samplers were placed into filter tins until they were ready to be analysed. The filters were then taken from the tins and dropped into individual glass beakers into which was added approximately 10 mls of the solvent mixture. The samples were left for 30 minutes with occasional agitation using an ultrasonic bath. Each of these solutions was then transferred into individual 25 ml volumetric flasks by filtering through a pipette tip with a glass wool plug inserted into the neck. A further 10 mls of solvent was used to wash further deposits off the filter and out of the beaker. The volume of each sample was then adjusted to 25 mls. A blank solution of solvent was kept in a 1 ltr glass bottle and dispensed using a 10 ml pipette and bulb.

The fluorimeter was zeroed by inserting a quartz cuvette filled with blank solvent into the sample holder and adjusting the zero control to give a reading of 0.000. The instrument was calibrated by inserting a standard of 1 µl formulation/litre of solvent and adjusting the variable gain control to obtain a reading of 1.000. Each sample was then analysed, the concentration of formulation being read directly from the fluorimeter. This was repeated three times for each sample, alternating with blanks to correct any zero drift or contamination of cuvettes. From the above procedure, the calibration coefficient for the fluorimeter (β) was determined to be 0.025 µl/unit of fluorimeter readout.

It was found that the use of polythene bottles to dispense the solvent caused major contamination to the solvent mixture. Therefore glassware was used for all storage vessels and dispensing equipment. Furthermore, since the quantities of formulation being analysed were so small, cleanliness was especially important, each flask and beaker required thorough rinsing with freshly made up solvent.

It is well known that fluorescent material held in weak solution will deteriorate over time and also if exposed to strong sunlight. Therefore all samples and standard solutions were kept in darkened cupboards and fresh standards made up daily. In addition all samples were left for no more than 24 hours before being analysed.

To keep a check on the repeatability of the analysis a number of check samples were prepared by pressing a spiked pad into a cardboard strip soaked in formulation and then pressing the pad onto the filter. The reproducibility of samples produced in this way was found to be very good, the variability being normally less than 15% of a mean fluorimeter reading of about 0.4. These filters were analysed at various times along with the sample filters in the normal manner.

4. RESULTS

4.1 Calm air measurements

The results from a series of measurements carried out in calm air at our Bush House laboratory are presented in Table 1. The numbers given are the actual readouts from the fluorimeter. A value of 1.00 therefore would indicate that the amount of formulation present in the 25 ml wash-off was equivalent to 1 µl in 1000 ml (i.e., a value of 1.00 means 0.025 µl of formulation in the wash-off). The results presented in the final column are obtained by washing both the inside of the nasal sampler and the brass ring which retained the filter in 25 ml of the blank solvent to remove any material deposited there.

Looking firstly at the results for the 20-minute samples the most obvious features are that there exists a high degree of run-to-run variability and that results obtained for the blank run [28.10.4] where no sampling was carried out were, in some cases, not noticeably different from the results obtained in the spraying runs. It was also evident from this run that greater care needed to be taken to avoid contamination in the handling of the nasal sampler although 'wash-out' results in the approximate range 0.1 to 0.5 (as seen from other blank runs) seem to be inevitable due to chemical contamination from the brass and solder of the 'nasal passages'. It was decided, therefore, that the run times needed to be substantially increased in order that the material collected would give a fluorescence signal well above the variability range due to machine calibration drift and contamination. An increase to 180 minutes (run [30.10.1]) proved more than adequate.

1A

Run number	Run time (mins)	Sampled aerosol (f)				Inspired aerosol (I_f) (I_w)	
		Gelman	Cowled 1	Cowled 2	UKAEA	Nasal filter	Nasal wash-out
27/10/3	20	0.019	0.019	0.014	0.028	0.059	1.48
27/10/4	20	0.040	0.033	0.037	0.042	0.070	1.02
28/10/1	20	0.049	0.033	0.040	0.029	0.165	1.35
28/10/3	20	0.044	0.037	0.022	0.021	0.059	1.27
28/10/5	20	0.086	0.094	0.060	0.079	0.534	0.810
28/10/4	blank	0.030	0.012	0.013	0.015	0.12	1.128
30/10/1*	180	0.49	0.31	0.40	0.61	1.64	8.39
30/10/2*	180	0.43	0.17	0.22	0.53	1.70	5.95
3/11/1	110	0.34	0.17	0.14	0.29	1.27	9.36
3/11/2*	110	0.48	0.22	0.18	0.47	1.71	6.35
3/11/3	110	0.13	0.17	0.14	0.28	1.14	3.64
4/11/2*	150	0.17	0.11	0.15	0.26	1.22	3.09
4/11/3*	150	0.13	0.13	0.11	0.16	0.98	9.24
10/11/1*	120	0.27	0.05	0.08	0.34	0.81	2.9
10/11/2*	120	0.40	0.04	0.06	0.68	1.68	7.68
10/11/3*	120	0.07	0.04	0.04	0.15	0.65	2.15
11/11/1*	120	0.33	0.21	0.19	0.28	2.41	1.12
11/11/2	120	0.46	0.17	0.21	0.45	2.87	5.82
11/11/3*	120	0.25	0.16	0.06	0.30	1.68	2.88

1B

Mean and (Std. dev.) of results from runs marked *, normalised to two hours spraying	0.27 (0.14)	0.125 (0.075)	0.125 (0.073)	0.31 (0.18)	1.30 (0.57)	4.30 (2.40)

Table 1a Actual experimental results from calm air runs carried out at Bush House. A value of 1.00 corresponds to 0.025 µl of sprayed oil so, for example, the fluorimeter reading for the Gelman sampler in run [30.10.1] indicates that 0.012 µl of oil were collected on this filter during the three hour sampling period.

(b) Means and standard deviations of the results in Table 1a for the longer period runs (from 30.10.1 onwards). Three runs have been specifically excluded from these calculations because the experimental conditions were different.

Concentrating, then, on the longer period runs, it is obvious that the amount of material collected on the internal walls of the nasal sampler (the 'nasal wash-out') was greater than that actually collected on the nasal filter. The reason for this finding became apparent when the filters were viewed under strong U.V. illumination. The fluorescent oil was seen (Photograph 5) to be concentrated near the edge of the filter which had been uppermost in the sampling head. This indicates (see Figure 2) that the dynamic behaviour of the aerosol particles, once they entered the body of the sampler through the nostrils, is inertially rather than electrostatically or gravitationally controlled. Also just apparent from Photograph 5 are the seven spots on the filter corresponding to the seven holes on the UKAEA sampler, supporting the inertially-controlled hypothesis. In Runs [3.11.1], [3.11.2] and [3.11.3] the height of the ECS nozzle above the plants was varied from 48 to 38 to 28 cm respectively to see whether any noticeable trend occurred. There is some evidence of a progressive reduction in the amount inspired as the sprayer is lowered but a definite conclusion would require more extensive testing. In all other runs the spraying height was 38 cm above the 'plants'. The only other run in which there was some departure from the normal was Run [11.11.2] for which a small radioactive source of α-particles (in turn producing positive and negative ions) was placed just below the nose to discharge the inspired aerosol. No obvious differences in the results were apparent in this case as compared to all the other runs.

The results in Table 1 can be expressed and interpreted in a number of different useful ways. If the fluorimeter reading for a given sampler is f, and those for the nasal filter and nasal wash-out are I_f and I_w respectively (so that the equivalent reading for the overall inspired aerosol is $I = I_f + I_w$), then we have

(a) Total amount of oil inspired (in μl) = $I\beta$

(b) Total amount of oil sampled (in μl) = $f\beta$

Run number	Run time	Calculated sampling ratios (R)			
		Gelman	Cowled	Cowled	UKAEA
30/10/1	180	0.61	0.39	0.50	0.76
30/10/2	180	0.70	0.28	0.36	0.87
3/11/1	110	0.40	0.20	0.16	0.34
3/11/2	110	0.74	0.34	0.28	0.73
3/11/3	110	0.34	0.44	0.37	0.73
4/11/2	150	0.49	0.32	0.43	0.75
4/11/3	150	0.16	0.16	0.13	0.20
10/11/1	120	0.91	0.17	0.27	1.14
10/11/2	120	0.53	0.05	0.08	0.91
10/11/3	120	0.37	0.18	0.18	0.67
11/11/1	120	1.16	0.74	0.67	0.99
11/11/2	120	0.66	0.24	0.30	0.65
11/11/3	120	0.68	0.44	0.16	0.82
Mean (standard dev)		0.60 (0.26)	0.30 (0.18)	0.30 (0.17)	0.74 (0.25)

Table 2

Here the results presented in Table 1, for personal samplers in the longer period runs, are expressed as a fraction of the total amount inspired (nasal filter + nasal wash-out) after normalising for the difference in flowrates (i.e., all personal sampler results multiplied by $^{25}/_{2}$). The positioning of the personal samplers on the body was the same for all runs.

(c) Percentage of the total amount sprayed which is inspired = $100\ (I\beta/10^3\ Qt)$

(d) Ratio of sampled to inspired concentration, $R = (f/I)\ (25/2)$

(e) Mean airborne concentration in breathing zone as determined by the nasal sampler (in µl of formulation/litre of air) = $I\beta/25t$

where Q is the rate of liquid flow from the ECS (1.6 ml/min) and t the duration of sampling. (Recall; β is the fluorimeter calibration coefficient (0.025 µl of formulation per unit of fluorimeter reading) and the numbers 2 and 25 refer to the sampling and inspiration flow rates in l min^{-1}).

In Table 2, the results of the longer period runs shown in Table 1 for the personal samplers have been recalculated according to (d) above. The means and standard deviations of the results are also presented. These results indicate that the '7-hole' sampler is perhaps the most appropriate of those tested for the assessment of the inspired fraction. Further testing in a range of conditions would, however, be required before any definite statements could be made.

In Table 3 the results given in Table 1 for the longer period inspired samples ($I_f + I_w$) are expressed in terms of (c) above, (i.e., as a percentage of the total amount of oil sprayed). Also given are the mean and standard deviation. These results suggest that a figure of 1 µl inspired per litre sprayed is a fair assessment for this calm air situation.

Comparison of equations given in (c) and (e) above indicates that a result quoted in terms of (c) may be converted to an estimate of the mean concentration of aerosol in the breathing zone by multiplying by (Q/2.5). Based on the mean value quoted in Table 3, therefore, the estimated mean concentration in the breathing zone is 5×10^{-5} µl l^{-1} of air. Assuming a droplet diameter of 50 µm (i.e., a droplet volume of 6.25×10^{-14} m^3)

Run number	Percentage of total amount sprayed which was inspired
30/10/1	8.7×10^{-5}
30/10/2	6.6 "
3/11/1 *	15.1 "
3/11/2	11.4 "
3/11/3 *	6.8 "
4/11/2	4.5 "
4/11/3	10.6 "
10/11/1	4.8 "
10/11/2	12.1 "
10/11/3	3.6 "
11/11/1	4.6 "
11/11/2 *	11.3 "
11/11/3	5.9 "
Mean (std.dev)	$7.28\ (3.17) \times 10^{-5}$

* These results excluded from the calculation of the mean since the experimental conditions were changed in these runs.

Table 3 In this case the results for the nasal sampler (filter + wash-out) in Table 1, are presented as percentages of the total amount of formulation sprayed during the run, based on a flow rate at the ECS of 1.6 ml min^{-1}. For example, in run [30.10.1] the total amount sprayed was 288 ml and the total amount collected in the nasal sampler was 0.25 µl. This means that the ratio between amount inspired and amount sprayed is 8.7×10^{-7} (8.7×10^{-5}%).

yields an approximate concentration of only one droplet per litre of inspired air.

In addition to the results given in Table 1, tests have also been carried out with the sprayer switched off to assess the background fluorescence levels. The result obtained was that the fluorescence emission due to material not arising from the sprayer was equivalent to about 0.002 µl of UVITEX dyed oil per cubic metre of air (i.e., less than 4% of the value with the sprayer on).

4.2 Moving air results

When the equipment was transferred from our countryside Bush House laboratory to our Edinburgh wind tunnel, the difference in background air quality was immediately obvious from the first filter samples collected. Whereas the samples collected in the former location had always looked clean, those obtained in the wind tunnel, at similar flow rates, had a greyish appearance. Further samples taken outside the tunnel and of the air outside the building confirmed that this pollution was a feature of the city air in general.

The extent to which this pollution would cause problems in the analysis of results became apparent when samples collected in the wind tunnel, without operation of the sprayer, were analysed fluorimetrically. Table 4 shows the relationship between the results for two sets of filters, the first of which was simply loaded into the sampling heads then removed with no sampling having been carried out and the second of which was run for two hours with the sprayer off. Using the result for the nasal filter to estimate the contamination level in terms of the amount of UVITEX-dyed formulation required to give an equivalent fluorimetric emission signal, a figure of 0.011 µl m^{-3} of air is obtained, a factor of 5 greater than the levels measured in the calm air laboratory. In the expectation that downwind spraying will reduce the amount of oil inspired, it is certain that these

Run number	Run time	Personal sampler results (f)				Inspired aerosol (I_f) (I_w)	
		Gelman	Cowled 1	Cowled 2	UKAEA	Nasal filter	Nasal wash-out
9/12/1	0	0.024	0.012	0.011	0.093	0.029	0.337
9/12/1	120	0.135	0.112	0.129	0.284	1.180	0.130

Table 4 Showing a comparison between two calm air, wind tunnel runs. In the first of these, filters were simply taken from the box, loaded into the samplers and then unloaded without any sampling being carried out. In the second run sampling was carried out for two hours with the sprayer switched off to record the background levels. A comparison of these background levels with the calm air results from Bush House gives clear indication that interference from background pollution is likely to cause severe problems in data analysis.

background levels would cause severe problems with the data analysis. A further noteworthy feature of the results from the second run (Run [9.12.2] of Table 4) is that in contrast to the results in Table 1 the result for the nasal filter is very much higher than that for the wash-out. This suggests that the aerosol or fume responsible for the background fluorescence either has a particle size very much smaller than the sprayed oil droplets and so is not subject to inertial impaction onto the body of the sampler, or is subject to particle bounce.

Attempts to remove the contaminant from the filter wash-off solution by filtration through several types of filter proved unsuccessful.

It was decided, at this point, that any further progress in alleviating this problem would require identification of the nature of the pollutant. A sample of the material washed from a filter was, therefore, analysed on a U.V. spectrofluorimeter. The results, showing a comparison of the emission peaks (for a 375 mm excitation wavelength) for the wash-off and the 'standard' 1 µl per litre UVITEX dyed formulation are shown in Figure 4. Unfortunately, the emission peaks for both materials are seen to occur at approximately the same wavelengths. The only real difference between the two solutions is that the line widths for the ambient contaminant is broader. This difference was not, however, considered sufficient to assist in the analysis. (Examination of the excitation spectra again indicated coicidence between the peak wavelengths).

It is well known that a common component of urban contamination is a group of chemicals called polyaromatic hydrocarbons (PAHs) which arise from traffic exhausts. It is also known that, dependent upon their aromatic nature, certain PAHs give rise to fluorescence. A sample of the contaminant was, therefore, washed from a filter in the normal way and analysed for PAH content using gas chromatography/mass spectrometry with selected ion monitoring. Low concentrations of anthracene, a known fluorescer, were identified.

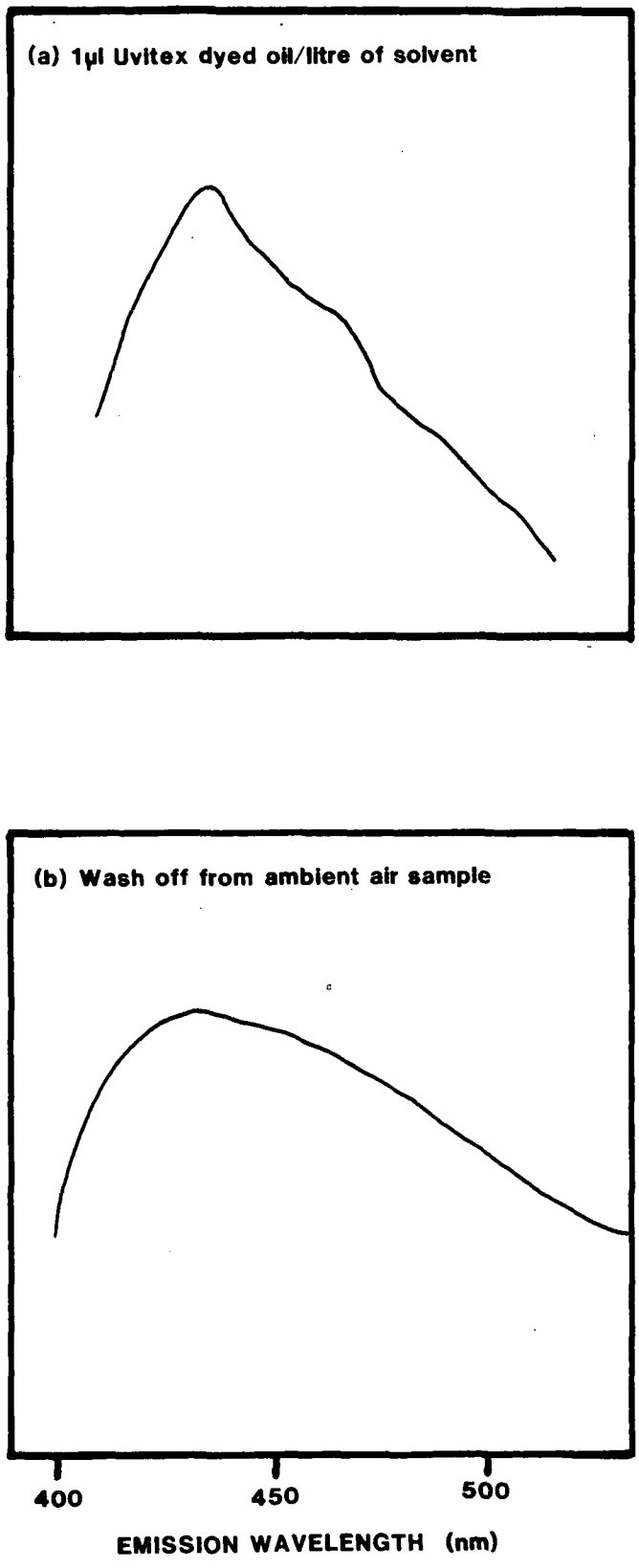

Figure 4 The fluorescence emission spectra from samples of (a) UVITEX in acetone : hexane and (b) the background pollutant in the same solvent. The emission peaks occur at roughly the same wavelength but the line width for the UVITEX is narrower.

From these tests we concluded that action to reduce the relative concentration of contaminant was not practicable in the time available. The best solution, therefore, was considered to be to run two high volume open-faced samplers, one inside and one outside the wind tunnel simultaneously with the spraying. The sampler within the tunnel was placed about 4 m upwind of the spraying position runs. By assessing the background level in this way the results from the personal and nasal samplers could then be appropriately adjusted (albeit roughly) by subtraction of the background.

The experimental results, for runs in which the wind speed was either 0 or 3 ms^{-1} and the sprayer was held in the recommended downwind orientation, are shown in Table 5 in the same form as the calm air results from Table 1. Also presented are the results from three runs where the wind speed was 1.5 ms^{-1} and the sprayer was held at right angles to the wind direction with the mannequin facing upwind. This was an attempt to simulate aerodynamic conditions equivalent to those experienced by an operator walking at a speed of 1.5 ms^{-1} in calm air. The sampling rates of the high volume, background samplers were 56 and 31 l min^{-1} for that outside (sampler HV1) and inside the tunnel (sampler HV2) respectively.

Comparing, firstly, the results for the high volume samplers at the 0 and 3 ms^{-1} windspeeds, we find that the mean value of the flow adjusted ratios between the two high-volume samples (HV1/HV2 x 31/56) changes from 1.00 (standard deviation 0.03) to 0.60 (0.07), a clear indication that, in the calm air situation, sprayed oil is being collected by the high volume sampler inside the tunnel. (Making the assumption that no oil is collected on the upwind sampler HV2 during the 3 ms^{-1} runs, the above result indicates that in calm air the airborne oil concentration near this sampler was approximately 0.004 $\mu l\ m^{-3}$ of air).

Moving on now to consider the results for the inspired sample we see that the nasal wash-out results for the 3 ms^{-1} wind speed are fairly constant at around the low value of 0.3 and are

Run number	Wind speed (ms^{-1})	Sampled aerosol (f)				Inspired aerosol (I_f) (I_w)		High volume sampler HV1	High volume sampler HV2
		Gelman	IOM	Cowled	UKAEA	Nasal filter	Nasal wash-out		
12/12/2	3	0.090	0.079	0.067	0.084	1.304	0.410	2.80	
15/12/1	3	0.092	0.080	0.069	0.084	1.077	0.303	2.43	1.31
15/12/2	3	0.103	0.077		0.079	1.014	0.209	2.29	1.28
15/12/3	3	0.089		0.070	0.073	0.956	0.295	2.20	1.18
16/12/1	3	0.036	0.030	0.025		0.410	0.244	1.02	0.56
16/12/2	3	0.045	0.037	0.026	0.037	0.458	0.204	1.06	0.61
16/12/3	3	0.071	0.055	0.051	0.053	0.744		1.72	0.99
17/12/1	0	0.059	0.117	0.060	0.169	0.863	1.168	1.45	1.24
17/12/2	0	0.050	0.087	0.028	0.172	0.855	1.665	1.17	1.17
17/12/3	1.5	0.033	0.016	0.012	0.013	0.238	0.349	0.47	0.25
18/12/1	1.5	0.061	0.072	0.030	0.044	0.607		1.43	0.79
18/12/2	1.5	0.069	0.052	0.026	0.055	0.630	0.404	1.51	0.86

Table 5 Results, of experiments carried out in the wind tunnel presented in the same form as in Table 1. The run time was two hours in all cases. The high volume samplers HV1 (56 l min^{-1}) and HV2 (31 l min^{-1}) were used to assess the background levels, one of them (HV1) being placed outside the tunnel and the other inside but well upstream of the spraying area. In the 3 ms^{-1} runs the sprayer was held downwind of the operator whereas in the 1.5 ms^{-1} runs the sprayer was held perpendicular to the wind direction.

always less than their corresponding nasal filter result. In the calm air runs the opposite was true and the high values for the wash-outs clearly indicated the presence of oil.

In order to interpret these results, the background contamination levels need to be subtracted. This has been done in Table 6 using the high volume sampler outside the tunnel (HV1) as a means of assessing the background levels. In performing this subtraction, we make the assumption that the efficiencies with which the personal and nasal samplers collect the background contaminent are the same as that for the high volume sampler. Clearly, from the existence of the small negative values this is not quite true. (The choice of sampler HV1 rather than HV2 to assess the background level is based on the aforementioned close correlation between the flow adjusted values for each and the desire to be positive that this 'background' filter was totally free of sprayed material).

Allowing for the expected inter-run variations, all results in the 3 ms^{-1} and 1.5 ms^{-1} runs are internally consistent. For the two calm air runs, however, the the amounts of oil collected on the open-faced Gelman and the cowled samplers are almost zero. This is not too surprising for the cowled sampler where the efficiency is expected, from the previous calm air results, to be low. We can offer no explanation, however, for the anomalously low result for the Gelman sampler.

The results presented in Table 6 are summarised in Table 7 in terms of their means and standard deviations.

With the results obtained so far it is difficult to comment further on the accuracy of the technique used in subtracting background levels. It may be, for example, that the sampler positioned outside the tunnel has a much higher sampling efficiency than either the personal or nasal samplers. In this case the values subtracted from the original data will be too high and the small negative results in, for example, the 3 ms^{-1} case could mask the fact that significant quantities of sprayed

Run number	Wind speed (ms^{-1})	Sampled aerosol (f) (corrected)				Inspired aerosol (corrected)		Subt'd from nasal filter	Subt'd from personal samplers
		Gelman	ICM	Cowled	UKAEA	Nasal filter	Nasal wash-out		
12/12/2	3	-0.010	-0.021	-0.033	-0.016	0.054	0.410	1.250	0.100
15/12/1	3	0.006	-0.006	-0.017	-0.002	-0.007	0.303	1.084	0.086
15/12/2	3	0.021	-0.005		-0.003	-0.008	0.209	1.022	0.082
15/12/3	3	0.010		-0.009	-0.006	-0.026	0.295	0.982	0.079
16/12/1	3	0.000	-0.006	-0.011		-0.045	0.244	0.455	0.036
16/12/2	3	0.007	-0.001	-0.012	-0.001	-0.015	0.204	0.473	0.038
16/12/3	3	0.010	-0.006	-0.010	-0.008	-0.023		0.767	0.061
17/12/1	0	0.007	0.065	0.008	0.117	0.216	1.168	0.647	0.052
17/12/2	0	0.008	0.045	-0.014	0.130	0.341	1.665	0.522	0.042
17/12/3	1.5	0.016	-0.001	-0.005	-0.004	0.029	0.349	0.209	0.017
18/12/1	1.5	0.013	0.013	-0.018	-0.004	0.037		0.640	0.048
18/12/2	1.5	0.015	-0.002	-0.028	0.001	-0.034	0.404	0.674	0.054

Table 6 The values given in the final two columns are the background levels as assessed by sampler HV1 and normalised to flowrates of 25 l min^{-1} (second last column) and 2 l min^{-1} (last column) respectively. The results for the filters are those presented in the previous table with the appropriate background level subtracted. The nasal wash-off results have not been altered since it appears that, in contrast to the oil droplets, the vast majority of particles of background pollutant are collected on the nasal filter as opposed to the body of the sampler. (Compare for example the results in Table 4 (second set) with the calm air results in Table 1).

Run numbers	Wind speed (ms^{-1})	Sampled aerosol (f) (corrected)				Inspired aerosol (corrected)	
		Gelman	IOM	Cowled	UKAEA	Nasal filter	Nasal wash-out
12/12/2 to 16/12/3	3	0.006 (0.010)	-0.007 (0.007)	-0.015 (0.009)	-0.006 (0.005)	-0.01 (0.03)	0.278 (0.077)
17/12/1 to 17/12/2	0	0.008 (0.0007)	0.055 (0.014)	-0.003 (0.015)	0.123 (0.009)	0.279 (0.088)	1.416 (0.351)
17/12/3 18/12/1 18/12/2	1.5	0.015 (0.002)	0.003 (0.008)	-0.017 (0.011)	-0.002 (0.003)	-0.014 (0.037)	0.376 (0.039)

Table 7 The means and (standard deviations) of the results presented in Table 6.

8a

Run number	Sampled background (f)				Inspired Background		Background
	Gelman	IOM	Cowled	UKAEA	Nasal filter	Nasal wash-out	Collected on HV1
5/1/1	0.053	0.085	0.143	0.049	0.413	0.102	0.952
6/1/1	0.029	0.043	0.048	0.032	0.267	0.215	0.652
6/1/2	0.041	0.040	0.025	0.033	0.362	0.240	0.869
6/1/3	0.040	0.047	0.047	0.045	0.630	0.279	1.480

8b

Run number	Sampled background (f)				Inspired Background		Subt'd from per.sam. results	Subt'd from nas.fil. results
	Gelman	IOM	Cowled	UKAEA	Nasal filter	Nasal wash-out		
5/1/1	0.019	0.051	0.109	0.015	-0.012	0.102	0.034	0.425
6/1/1	0.006	0.020	0.025	0.011	-0.024	0.215	0.023	0.291
6/1/2	0.010	0.009	-0.006	0.002	-0.026	0.240	0.031	0.388
6/1/3	-0.012	-0.005	-0.005	-0.007	-0.031	0.279	0.052	0.661
Mean (Std. dev)	0.006 (0.013)	0.019 (0.024)	0.031 (0.054)	0.005 (0.010)	-0.023 (0.008)	0.209 (0.076)		

Table 8a The results of some 3 ms^{-1}, two hour runs presented in the same form as the results in Table 5. In these runs, however, the sprayer was switched off so that the relative sampling efficiencies of the various samplers for background pollutant could be tested and the results compared with those with the sprayer on. In this way we hoped to test whether the results presented in Table 6 indicated whether the amounts of oil sampled, under these conditions, were significantly different from zero and to establish upper confidence limits.

(b) The results from Table 8a treated in the same way as were those in Table 6. Also shown are the means and (standard deviations) for comparison with the corresponding results in Table 7.

oil were, in fact, being collected. To check this, a series of runs was carried out under the same 3 ms^{-1} wind conditions using the same set-up but with the sprayer switched off. The results are presented in Table 8.

The principal results from the previous tables are summarised in Table 9.

Sampler type	How the results are presented	Location (wind speed)			
		Bush House	Central wind tunnel		
		0 ms^{-1}	0	1.5	3 ms^{-1}
'7-hole'	As the mean of the ratios between the amounts of material collected on these filters to that collected on the nasal sampler after correction for the differences in flow rate	0.74	0.91	*	*
Gelman		0.60	0.06	*	*
Cowled		0.30	0	*	*
ICN		-	0.41	*	*
Nasal filter holder	As a percentage of the total amount of material sprayed during the sampling period	7.28×10^{-5}%	2.21×10^{-5}%	$<2.5 \times 10^{-6}$%	$<4.5 \times 10^{-6}$%

Table 9 A summary of the principal results presented in the foregoing tables. The ratios between personal sampler results and the amounts inspired were indeterminable for the 1.5 and 3 ms^{-1} wind speeds because of the very small amounts of material sampled.

5. DISCUSSION

Referring firstly to the calm air measurements carried out at Bush House it is interesting to consider in more detail the behaviour of the highly charged aerosol droplets in the breathing zone. The relationship between gravitational and electrostatic forces in this region based on some broad assumptions, is considered in Appendix 1. The conclusion is that the behaviour of the aerosol droplets in this region is more likely to be dominated by gravitational rather than either image or local space charge forces. It is possible, however, that the controlling influence may, even at this distance, be the electric field which exists between the earthed mannequin and the combination of the charged spraying head and its local high density cloud. The complexity of the field distribution involved, however, makes this difficult to calculate. What is certain is that once inside the nostrils or the body of one of the personal sampling devices, this latter field will be effectively nullified and the motion of the particle will be determined by a combination of gravitational, inertial and aerodynamic forces. The visual observation of high density of oil droplets near the top of the nasal sampling filter, together with the fact (see Table 1) that a much higher proportion of the droplets is deposited on the inside walls of the sampler than on the filter itself, indicate that the dynamic behaviour is inertially controlled. The clear definition of seven spots on the filter of the '7-hole' sampler corresponding to the seven holes indicated that the same is true for this device.

For the cowled sampler, the cowl is a 6 cm long tube which hangs vertically downwards from the filter and hence acts as a vertical elutriator. At a sampling rate of 2 l min^{-1} the average upward air velocity through this tube is approximately 0.07 ms^{-1}. This is comparable with the falling speed in air of 50 μm diameter droplets (about 0.08 ms^{-1}). Therefore the low mean value of R (about 0.3) for this device (see Table 2) seems reasonable.

The best of the personal samplers tried in this part of the study in terms of both the consistency of results and sampling efficiency representative of inspirability was the '7-hole' sampler, although the open-faced Gelman sampler also performed reasonably well. It is suspected, however, for the reason given above that the performance of the cowled sampler would be highly sensitive to changes in particle size and hence this device is not considered suitable. A further point in favour of the '7-hole' sampler is the difficulty of accidental contamination. It must be stressed again, however, that these are preliminary findings and require further confirmation before being acted upon. Other personal samplers intended specifically for inspirable aerosol in general are at present being developed to meet new sampling criteria (e.g., ACGIH 1985), including the new IOM instrument (MARK et al 1986). These need to be considered in any future enquiry.

For the few runs in which the height of the spraying head above the targets was changed there was some evidence that, as may be expected, raising the sprayer causes an increase in the inhalation hazard. Whether this result is due to the actual height of the spraying head in relation to the face of the operator or to the spraying head to target separation is, however, not clear.

The apparent lack of influence of the radioactive discharging device (Run [11.11.2]) may be due to the aforementioned conclusion that the behaviour of the inspired aerosol is inertially rather than electrostatically controlled. In the absence of particle charge measurements, however, its effectiveness in neutralising the charge carried by the droplet is uncertain. We would be reluctant, therefore, to draw any conclusions from this single measurement.

Moving on now to consider the results obtained in the wind tunnel, and using Student' t-test (see Appendix 2) to compare the results presented in Table 7 for calm air (zero wind speed) to the corresponding means and standard deviations of the

results of the longer period runs in Table 1 (normalised to 120 mins spraying time) we find that the differences, between locations, for the total amounts of material sampled by the nasal sampler is not significant at the 5% level. The same conclusion applies to the results for the '7-hole' sampler. For the open-faced Gelman and the cowled samplers the level of significance is approaching 5%. We find, therefore, that while the changes in experimental set-up (most notably the reduction required in the overall height in the wind tunnel) may have reduced the amount of sprayed oil entering the breathing zone, the difference is not conclusively established.

The '7-hole' sampler again proved itself useful in these calm air runs, the mean sampling efficiency in comparison to the total amount of material inspired being 92% (Std.devn.19%).

The most important factor here, however, is the very obvious difference between the calm air results and those wherein the sprayer was held either downwind or perpendicular to the wind direction. The small sample t-test (Appendix 2) has again been used here to determine whether the 3 and 1.5 ms^{-1} wind speed results are significantly different from the values obtained in the 3 ms^{-1} runs (Table 8) where no spraying was carried out. The findings of this test are that in the cases of both the inspired and personal samplers, the differences between the means for the runs wherein the sprayer was operating and the corresponding means for runs where it was not operating are not statistically significant. A further test (Appendix 2, part (b)) to establish upper confidence limits for the amount of material inspired showed that, in the case of 3 ms^{-1} downwind spraying, we can be 95% confident that the amount of material inspired will not exceed 2.5×10^{-6}% of the total amount sprayed. The corresponding 95% confidence limit for the 1.5 ms^{-1} runs spraying perpendicular to the wind direction is 4.5×10^{-6}%.

The very high protection factor associated with spraying in moving as opposed to calm air poses the question of what might be the effect on total daily exposure of a very brief change in

wind direction. For example, if the difference between the exposure levels between upwind and calm air spraying were of the same order as that measured between calm air and downwind spraying (a factor of at least 50), then a reversal of wind direction for only $^{1}/_{2500}$th of the spraying period need occur to double operator exposure. An investigation of spraying under variable wind conditions is therefore required.

Finally, it is worth considering how the results obtained here relate to previous studies using more conventional hand-held spraying devices. A review of the potential hazards, by both the dermal and respiratory routes, of these and other mechanical spraying devices is given by TURNBULL et al. (1985). The range of inhalation exposure measurements from studies based on the outdoor use of hand-held sprayers delivering volumes of greater than 1000 litres per hectare is quoted as being between 0.002 and 1.4 ml of spray per person per hour. Adopting a figure of 2000 litres per hectare and assuming that a single operator may cover an area of 0.1 hectares per hour implies that a spraying rate of 200 l/hour engenders a respiratory uptake rate of between 2×10^{-6} and 1.4×10^{-3} l/hour. In terms of the units previously used here, this implies that the volume inspired amounts to between 10^{-6}% and 7×10^{-4}% of the total volume sprayed. The 'calm air' figure for the study reported here (10^{-4}%) falls within this range.

A further point worth noting from the aforementioned summary is that this range (0.002 - 1.4 ml/person/hour) of inhalation exposure is commensurate with a dermal exposure range of 2 - 350 ml/person/hour. Pending further data analysis we are unable to quantify the relationship between exposure by these two routes for the ECS used here. However, in view of the generality of this funding in previous studies, and the qualitative appearance (under UV illumination) of the overalls worn by the dummy operative in this study, we are confident that the relationship between dermal and respiratory exposure is of the same order for the ECS under the conditions investigated.

6. CONCLUSIONS

Our preliminary conclusions may be summarised as follows:-

1. In the calm air situation where the operator is stationary, the percentage of the total amount of oil sprayed which is inspired is of the order of 10^{-4}%.

2. Assuming a mean oil droplet diameter of 50 μm and comparing the breathing and spraying rates, this figure corresponds to approximately one oil droplet per litre of inspired air.

3. The higher the spraying head is held above the target the greater is the concentration of sprayed material which reaches the breathing zone.

4. The dynamic behaviour of inspired droplets in the head and upper airways will be dominated by mechanical as opposed to electrostatic forces.

5. The performance of personal samplers is critically dependent on sampler type as may be expected for droplets of this size. Of the types tried the '7-hole' sampler gave values most representative of the amount of material inspired. However, other personal samplers aimed specifically at sampling inspirable aerosol are now becoming available, and these should be examined in any future enquiry, particularly in the expectation that TLVs for 'total' aerosol will, in due course, be defined in terms of Inspirable Particulate Mass (IPM).

6. For the moving air experiments, the accuracy of our experimental measurements was not sufficient to permit the very small amounts of oil collected by either the nasal or personal samplers to be distinguished from zero with any statistical certainty.

7. Whilst the low results obtained for moving air preclude definite measurements of the amounts of material inspired in these runs, we can be 95% confident that, in the case of the 3 m s^{-1} downwind results, the inspired fraction does not exceed 2.5 x 10^{-6}% of the total amount sprayed. The corresponding result for the 1.5 m s^{-1} wind speed is 4.5 x 10^{-6}%. (These compare (see Appendix 2) with an upper 95% confidence limit of 9.6 x 10^{-5} for calm air spraying in this location).

8. The photographs of the suits from the calm air runs under U.V. illumination show a strong regional variation in dermal contamination, the most heavily exposed regions being the lower parts of the body on the same side as the spraying device.

9. The results obtained under calm air conditions are an encouraging indication that a suitable sampling technique can be identified. However, it is highly desirable that the effectiveness of the sampling technique should be validated for a wider range of operating conditions (especially moving air).

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APPENDIX 1

The relative effects of the various forces experienced by the droplet on its dynamics in the breathing zone.

The lack of detailed measurements of particle size and charge and of the air movement around the mannequin make accurate assessment of the effect of charge on the dynamics of the droplet impossible. It is worthwhile, however, to base some rough calculations and some of the assumptions already made. Consider, for example, the ratio between the electrostatic and gravitational forces acting on a 'typical' droplet of diameter 50 μm. The gravitational force (F_g) (assuming that the formulation has the same density (1000 kg m^{-3}) as water is given by

$$F_g = mg \rightarrow 6.5 \times 10^{-10} \text{ N for a 50 } \mu\text{m particle}$$

where g is the gravitational acceleration and m is the particle mass.

The electrostatic force (F_e) which exists between two charged particles (the Coulomb force) is

$$F_e = \frac{q_1 q_2}{4 \pi \varepsilon_o r^2} \rightarrow 9 \times 10^9 \left(\frac{q}{r}\right)^2 \text{ for equal charges} \qquad (A1.1)$$

where q_1 and q_2 are the charges carried by the respective particles r is the distance between them and ε_o is the permitivity of air (8.85×10^{-12} F m^{-1}).

When a charged object approaches an earthed surface it induces a charge of opposite polarity on that surface and hence is attracted to it by a force (F_i) which is equivalent to that which would result if an oppositely charged 'image' of the object were placed at the same distance behind the earthed plane. In this case,

$$F_i = \frac{q^2}{4 \pi \varepsilon_o (2 d)^2} \rightarrow 9 \times 10^9 \left(\frac{q}{2d}\right)^2 \qquad (A1.2)$$

where d is the separation between the object and the earthed plane.

In the absence of a direct measurement of q, we can make an estimate of its value by considering the relationship between the electrical current flowing to the spraying head and the rate of droplet production as follows:-

The spraying rate is approximately 2 ml min^{-1}, (i.e., 3.3 x 10^{-8} m^3 s^{-1}).

The volume of a 50 μm diameter droplet is approximately 6 x 10^{-14} m^3.

Therefore, the rate of droplet production is approximately 5 x 10^5 s^{-1}.

The current drawn by the sampler (as measured in the earth wire) is 1.6 x 10^{-7} A.

The charge per droplet (q), on this basis, is given by

$$q = \frac{1.6 \times 10^{-7}}{5 \times 10^5} \sim 3 \times 10^{-13} \text{ Coulombs} \qquad (A1.3)$$

or 1.7 x 10^6 electric charges per droplet.

Substituting q from Equation A1.3 into Equation A1.1 gives

$$Fe = \frac{8 \times 10^{-16}}{r^2}$$

so that, for $F_e = F_g$,

$$r = \left(\frac{8 \times 10^{-16}}{6.5 \times 10^{-10}}\right)^{1/2} \sim 10^{-3} \text{ m}$$

This means that for inter-particle electrical forces to dominate dominate over gravitational forces the droplets should be less than 1 mm apart.

A similar treatment for the image force (Equation A1.2) indicates that for the image force to dominate over gravitational effects the particle should be within 0.5 mm of the earthed surface.

APPENDIX 2

(a) Calculation of the significance of the difference in the means of two small samples using Student's t-test

In this case we wish to determine if any apparent difference between the means x_1 and x_2 of two sets of samples is significant or not. This difference is defined as

$$x_d = x_1 - x_2 \qquad \text{(A2.1)}$$

The first step in this calculation is to make a pooled estimate of the variance (σ^2) for the two samples based on the "Null Hypothesis' that the mean and variance of the two groups of samples are identical, i.e.,

$$\sigma^2 = \frac{n_1 s_1^2 + n_2 s_2^2}{n_1 + n_2 - 2} \qquad \text{(A2.2)}$$

where n_1 and n_2, and s_1 and s_2 are numbers of samples and the standard deviations for each group.

The best estimate of the standard error of the difference in means is then

$$\sigma_W = \sigma \left(\frac{1}{n_1} + \frac{1}{n_2}\right)^{1/2} \qquad \text{(A2.3)}$$

The value of Student's t is then given by

$$t = \frac{\left| x_1 - x_2 \right|}{\sigma_W} \qquad \text{(A2.4)}$$

where x_1 and x_2 are the group means.

The level of significance of the difference between the means is then determined by comparing this value of t with tabulated values for the appropriate number of degrees of freedom ($n_1 + n_2 - 2$).

(b) Upper confidence limits

The foregoing test has been used to establish the fact that the mean values given in Table 7 for 3 ms^{-1} sampling were not significantly different from those recorded in Table 8 where a similar sampling regimen was carried out but the sprayer was switched off. We now attempt to use a first estimate of the mean value of the amount of oil inhaled together with the variability of the measured results to define an upper limit above which we can say with a certain degree of confidence that the true mean does not lie. Our first estimate of the mean is, in fact, as defined as x_d in Equation A2.1 where x_1 is the mean value for the runs in which the sprayer was operating and x_2 that in which it was not operating.

For this analysis we return to tabulated values of Student's 't' for the appropriate number of degrees of freedom (again 9). These tables show that we can be 95% confident that the amount of sprayed oil collected, for example, on the nasal filter does not exceed 1.83 standard errors (Equation A2.3) of the estimated mean (Equation A2.1) and that we can be 99% confident that it does not exceed 2.82 standard errors of the same estimated mean. For the 3 ms^{-1} windspeed nasal filter results (Tables 7 and 8b), therefore, where the estimated mean (x_d) is 0.013 and the best estimate of the standard error (σ_w) is 0.011, the upper 95% confidence limit is,

$$C_{95} = 0.013 + (1.83 \times 0.011) = 0.032$$

and the upper 99% confidence limit is

$$C_{99} = 0.013 + (2.82 \times 0.011) = 0.044$$

The corresponding values for the nasal wash-out are

$$C_{95} = 0.069 + (1.83 \times 0.048) = 0.157$$

and

$$C_{99} = 0.069 + (2.82 \times 0.048) = 0.204$$

We can compare these with the corresponding calm air measurements presented in Table 1b. Here the upper 95% confidence limits are

$$C_{95}\ (\text{filter}) = 1.3 + (1.83 \times 0.19) = 1.65$$

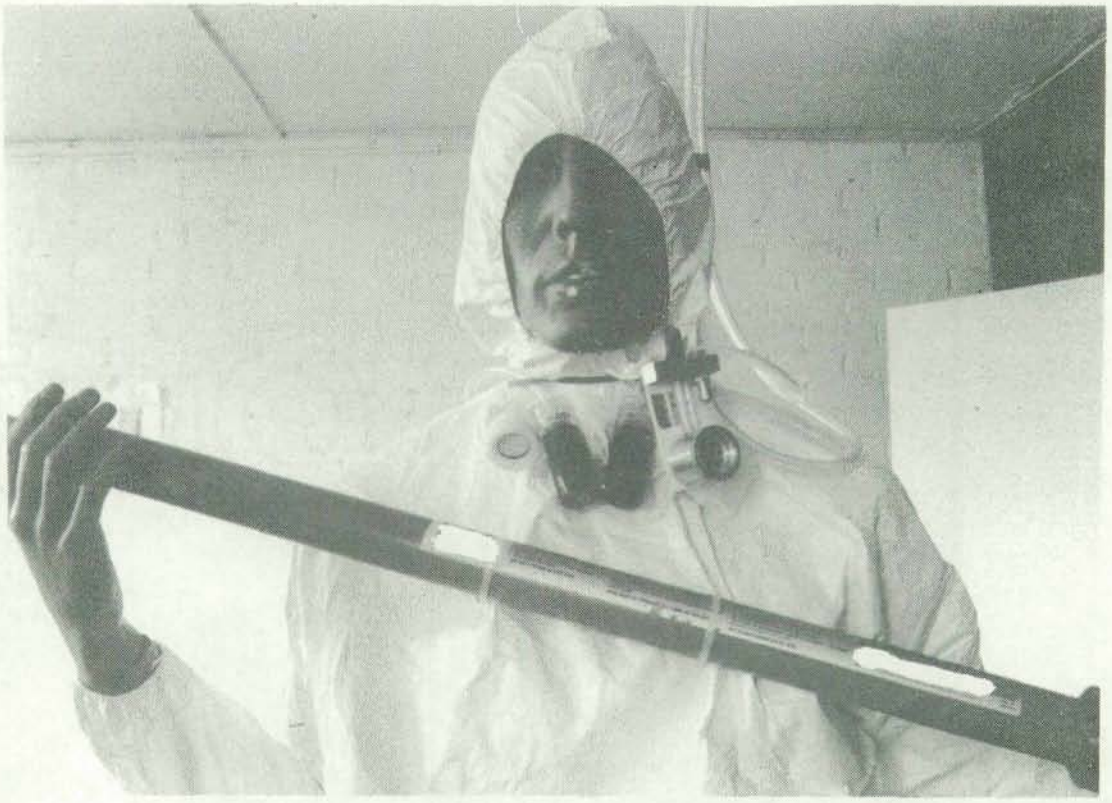
$$C_{95}\ (\text{wash-out}) = 4.3 + (1.83 \times 0.8) = 5.76$$

The ratio between the upper 95% confidence limit for the total amount inspired (filter + wash-off) in calm air spraying to that for downwind spraying is, therefore, of the order of 40 : 1.

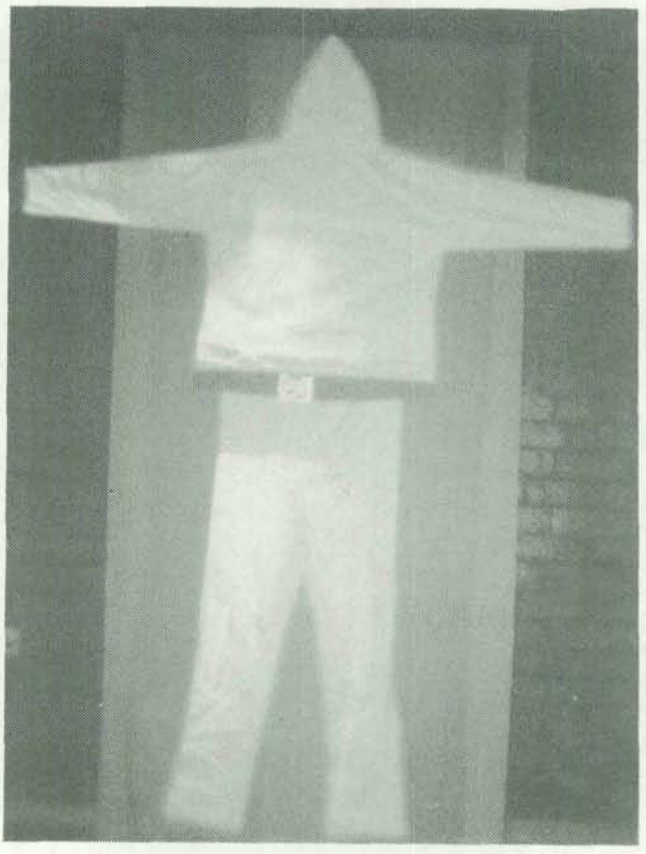
Converting the results obtained here, as in (c) of Section 4.1 of the main text, we find that the upper 95% confidence level expressed as in Table 3, i.e., in terms of the percentage inspired, is 2.5×10^{-6}% for 3 m s^{-1} downwind spraying. (Or, again assuming a droplet diameter of 50 μm, this corresponds to 25 droplets per m^3 of inspired air).

If we assume that the relative sampling efficiencies, for background pollutant, of the nasal and high volume samplers is similar in the 1.5 ms^{-1} perpendicular spraying case to that in the above 3 ms^{-1} downwind case, then the corresponding values of C_{95} for the nasal filter and nasal wash-out are 0.045 and 0.292 respectively.

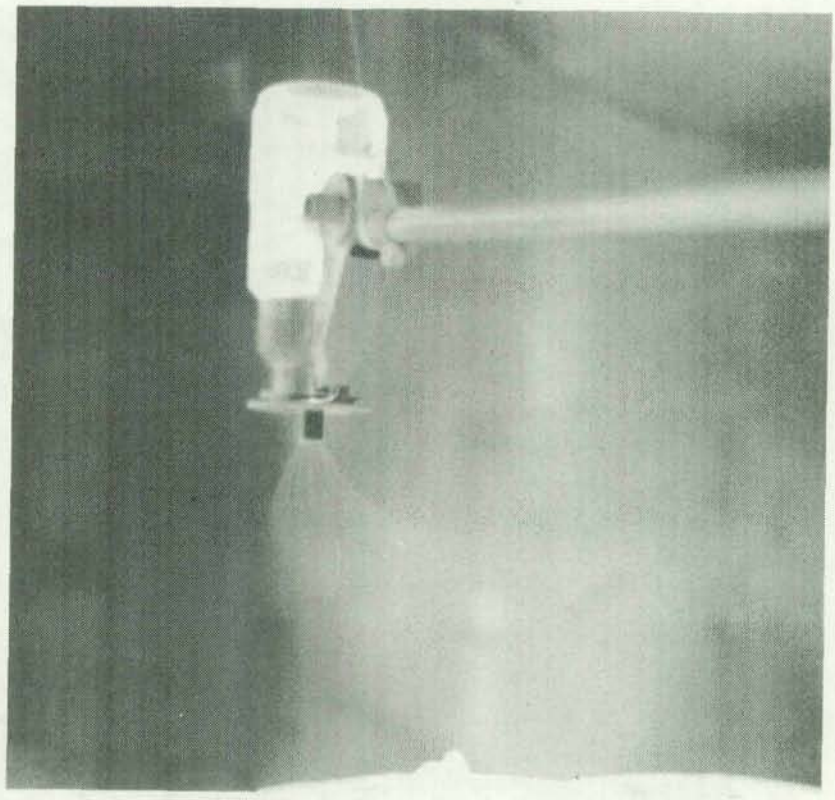
Photograph 1 : The basic experimental set-up in the calm air chamber.



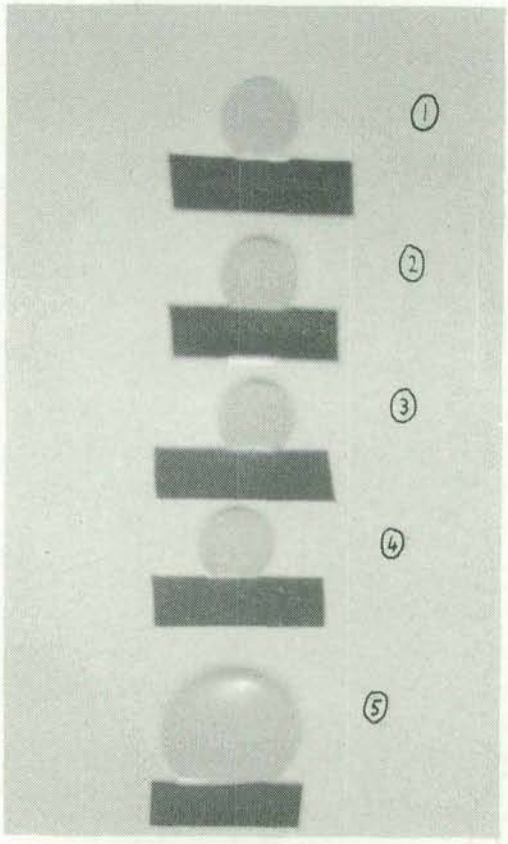
Photograph 2: Showing the positions of the personal samplers. From left to right: the Gelman, two Millipore cowled and the '7-hole' sampler.



Photograph 3 : An example of the distribution of sprayed oil on the front of a suit from the calm air chamber. The photograph was taken under U.V. illumination.



Photograph 4 : Showing the effect of a 1.5 m s^{-1} wind on the spray near the nozzle.



Photograph 5: A photograph under U.V. illumination of a set of exposed filters. Just visible on filter 4 are seven spots corresponding to the holes on the '7-hole' sampler. On filter 5 the high concentration of formulation at the point which was nearest the top of the nasal sampler is clearly visible.

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