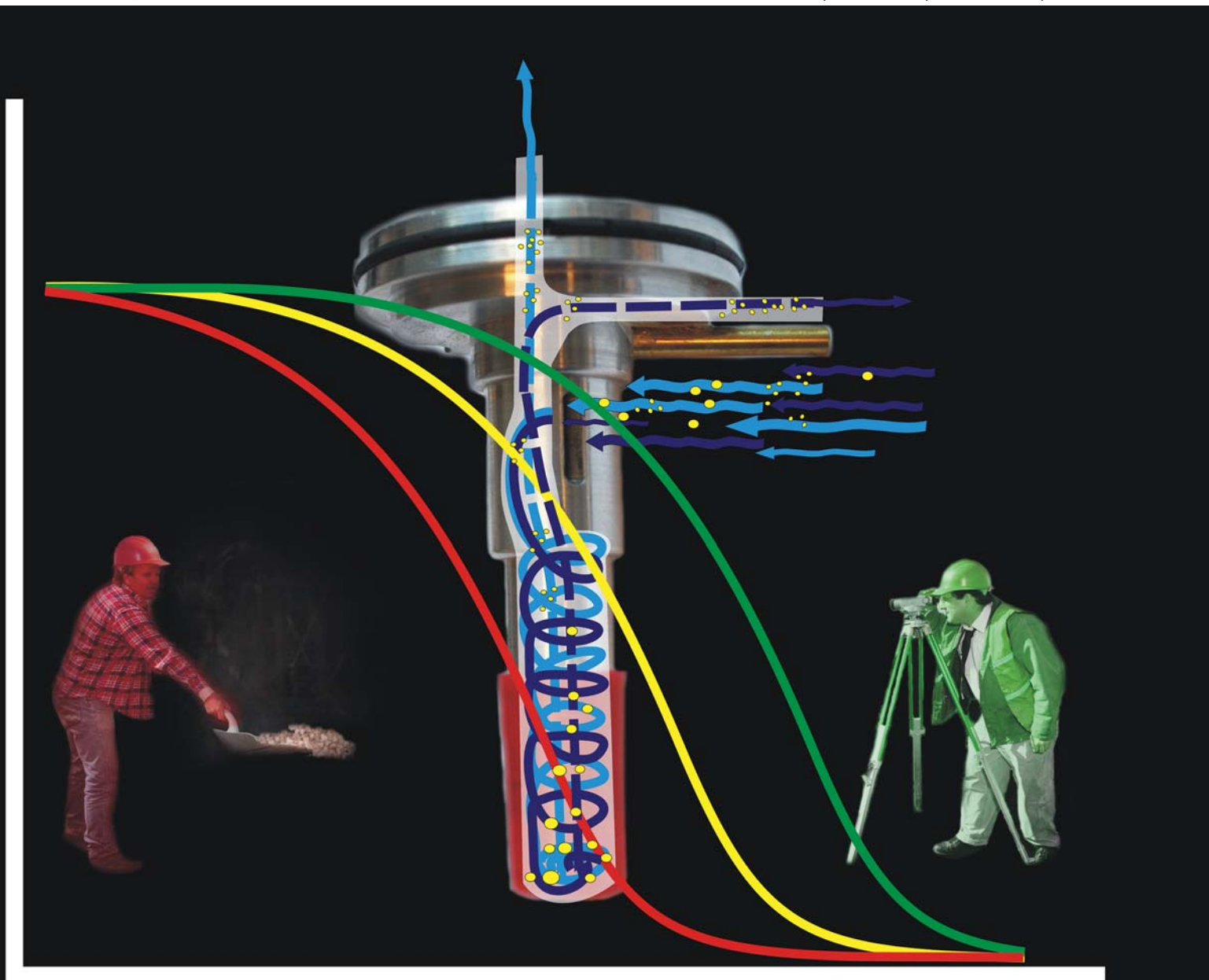


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Lee *et al.*

Size-selective sampling of particulates using a physiologic sampling pump

## Size-selective sampling of particulates using a physiologic sampling pump†‡

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Recent laboratory research indicates physiologic sampling of gas and vapor may provide more representative estimates of personal exposures than traditional methods. Modifications to the physiologic sampling pump (PSP) used in that research are described which extend its usefulness to size-selective sampling of particulates. PSPs used in previous research varied motor speed to keep sampling proportional to the subject's inhalation. This caused airflow and particle velocities through the collection device to continually change making those pumps unsuitable for sampling particulates. The modified implementation of the PSP pulls a constant airflow into and through a cyclone, then uses valves to either direct the airflow through, or divert the airflow around, the sampling filter. By using physiologic inputs to regulate the fraction of each second that air flows through the sampling filter, samples may be collected in proportion to inhalation rate. To evaluate the performance of a functional prototype 5 different sizes of monodisperse aerosols of ammonium fluorescein were generated by a vibrating orifice aerosol generator and introduced into a calm air chamber. To simulate different inhalation rates the valves of the PSP were energized using 9 different duty cycles. Efficiency curves are presented and compared to a standard respirable convention by bias mapping. The performance of the modified cyclone used in the PSP sampling head compared favorably with a commercially available cyclone of the same model, operating at a constant airflow ( $\pm 10\%$  over almost all the size distributions of concern). The new method makes physiologic sampling of the respirable fraction of particulates feasible.

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‡ Electronic supplementary information (ESI) available: Efficiency curves for the PSP sampling head associated with each PSP duty cycle are superimposed upon those for the reference cyclone and upon the respirable convention. The functions and coefficients used to generate these efficiency curves are also presented. See DOI: 10.1039/c0em00445f

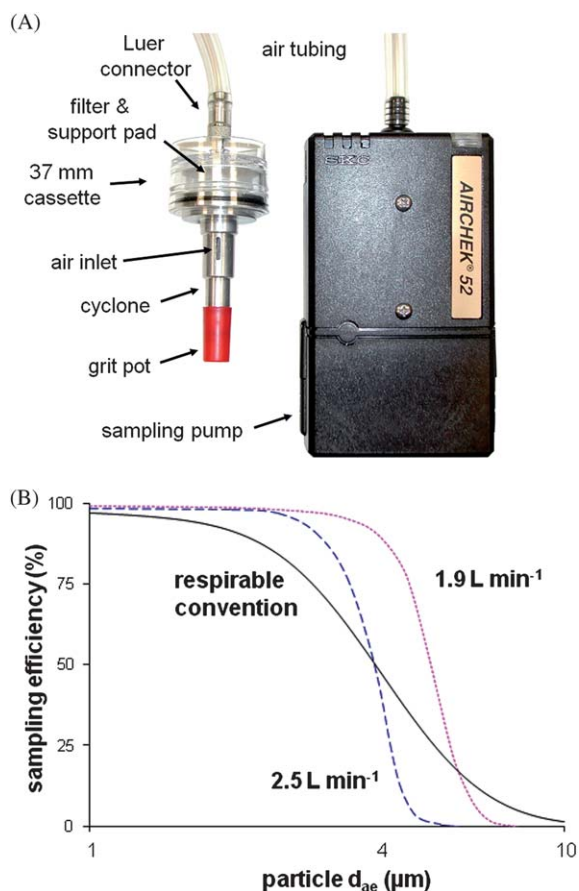
### Introduction

Disease and death may be prevented by knowing and controlling workers' exposures to airborne hazards. Occupational epidemiologists determine dose–response relationships and regulatory agencies set occupational exposure limits (OELs) accordingly. Personal monitoring documents compliance with these limits. Collecting air samples representative of actual exposure is central to personal monitoring, and therefore to safeguarding the workforce.

Inhalation is the primary mode of exposure to airborne hazards. For particulates, the respirable fraction is that portion of the total airborne particulates that may enter the alveolar region of the lungs. Depth of penetration is largely a function of

### Environmental impact

It is likely that situations of increased worker exposure to chemicals occur when the workload is highest and when the rate of breathing is also highest. Traditional worker sampling for airborne contaminants using a constant flow-rate may not accurately assess dose if this correlation exists. A physiologic sampling pump (PSP) has been demonstrated to more accurately assess exposure to gases or vapors under conditions of variable breathing rate positively correlated with exposure. Situations where exposure and breathing-rate are similarly correlated for aerosols could include shoveling sand or pouring bags of powder. In this paper, it is shown that the same technique of the PSP can be applied to sampling the respirable fraction of aerosols to also give a more accurate estimation of dose.



**Fig. 1** (A) The SKC, Inc. aluminium cyclone, part number 225-01-02, as an integral part of a typical sampling train. (B) Two efficiency curves for this cyclone plotted with the ISO/CEN/ACGIH respirable convention curve. 1.9 and 2.5 L min<sup>-1</sup> plots reconstructed from Trakumas and Hall.<sup>3</sup>

the aerodynamic equivalent diameters ( $d_{ae}$ ) of the particles. The respirable fraction is considered to include particles with  $d_{ae}$  up to 10  $\mu\text{m}$ . Sampling a specific size-selective fraction of the aerosol is required to demonstrate compliance with many occupational exposure guidelines.

Equipment typical of that used for size-selective sampling is shown in Fig. 1A. A precollector, such as a cyclone, is placed at the front of the sampling train so that only the respirable fraction of the airborne particles may penetrate (exit) the cyclone and deposit on the sampling filter. While the pump is running, the air is pulled into, and whirls inside of, the cyclone, then passes through the sampling filter, the air tube, and the pump, to exhaust into the ambient. Larger particles, *i.e.*, those with greater inertia, tend to impact the inside surface of the cyclone and either remain there, or drop into the grit pot. Smaller particles tend to exit the cyclone in the airstream and deposit on the sampling filter. During use the cyclone is clipped to the subject's clothing in the breathing zone, the air tube is draped over the shoulder, and the sampling pump is secured by a clip (on the back of the pump) to the belt. Fig. 1B plots sampling efficiency *vs.* particle  $d_{ae}$  for a cyclone of the same model as pictured. Sampling efficiency is not only a function of the particle  $d_{ae}$ , but also of the magnitude of the airflow through the cyclone.

Standard methods for sampling have been developed to promote objective measurements of personal exposure and

facilitate uniform enforcement of OELs. NIOSH methods 0600 and 7500 offer two examples,<sup>1</sup> 0600 for respirable particulates not otherwise regulated and 7500 for silica. Both of these methods allow the use of an SKC, Inc. (Eighty Four, PA, USA) aluminium cyclone with a 5  $\mu\text{m}$  pore size polyvinyl chloride (PVC) filter housed in a 37 mm sampling cassette (amongst other devices). Use of a particular make or model of pump is not required, but for method 0600, pulsations in the pump flow must be within  $\pm 20\%$  of the mean flow (European Standard EN1232,<sup>2</sup> however, requires pulsations to be within  $\pm 10\%$  of the mean flow). Both methods specify sampling with a mean airflow of 2.5 L min<sup>-1</sup> when using the aluminium cyclone. The total volume of air sampled may therefore be calculated by multiplying the specified flow by the duration of the sampling session. A corresponding time-weighted average (TWA) concentration may be obtained by dividing the mass collected (total mass or mass of silica) on the filter by this volume.

Size-separation efficiency within the cyclone is affected by the magnitude of the airflow. The International Standards Organization/Comité Européen de Normalisation/American Conference of Governmental Industrial Hygienists (ISO/CEN/ACGIH) respirable convention curve describes the probability of penetration for different sizes of respirable particles within workers' airways when working under a typical workload. This probability curve is often described in terms of the point of 50% probability of penetration, which corresponds in this case to particles having a  $d_{ae}$  of about 4  $\mu\text{m}$ . Samplers are designed to and operated at a flow that minimizes the overall bias relative to the convention for a range of typical aerosol size distributions.<sup>4</sup> According to the manufacturer<sup>5</sup> and Trakumas and Hall,<sup>3</sup> this 50% cut-flow is 2.5 L min<sup>-1</sup> for their aluminium cyclone, but other researchers<sup>6,7</sup> (including the authors of this paper) have found it to be 2.2 L min<sup>-1</sup>, which is a value more consistent with the original design specifications. In 1998 Gudmundsson and Lidén<sup>8</sup> published a new method to generate a polydisperse polystyrene aerosol by which they observed the 50% cut-off at  $d_{ae} = 5.3 \mu\text{m}$  for a flow of 2.1 L min<sup>-1</sup>. Measurement of the 50% cut-off is confounded so much by various factors, such as the use of different experimental methods and test aerosols that Görner *et al.*<sup>7</sup> consider agreement within 1  $\mu\text{m}$  a fair achievement.

Since standard methods require sampling at a constant mean flow, total mass collected may not correlate well with the inhaled dose.<sup>9-15</sup> Satoh *et al.*<sup>15</sup> note that pulmonary ventilation during strenuous work may be as much as ten times the resting rate. Lin *et al.*<sup>9</sup> used six workload scenarios to estimate the inhaled volume and multiplied each of these by the TWA concentration obtained in their study. They found that estimated dose varied from -31% to 115% relative to a common baseline. To compound the uncertainty, different subjects performing the same task will likely have different pulmonary ventilation rates due to various factors such as differences in gender or age.<sup>14</sup>

Sampling at a constant flow also ignores any correlation between a worker's inhalation and the concentration of contaminants.<sup>9,12-14</sup> For example, Lin *et al.*<sup>9</sup> suggested a worker shoveling dirt at a faster rate might increase the amount of dust generated. The converse might also apply: the onset or increase of an airborne contaminant might cause a worker to move and breathe more cautiously and perhaps take longer to complete the

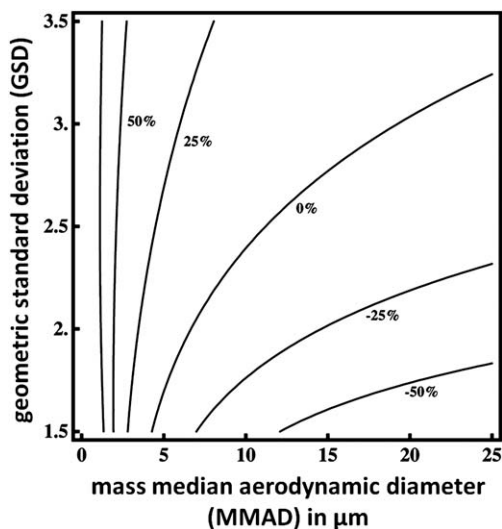
task. This might happen at outdoor worksites when the wind changes direction. There are no field studies that document the strength of this correlation. The necessary personalized equipment to conduct the research has not been available.

The ideal personal sampler would both sample the same volume of air and capture the same number and mix of particles as the subject inhales during the time of interest. This implies that sampling would follow the subject's inhalation. A more compact implementation of this idea would sample air proportional to, but at a preset fraction of, inhalation and would collect the same particles at that same preset ratio. Collection within the limits of detection for the analytical method being used is assumed. Pumps designed with a view towards this ideal are called physiologic sampling pumps (PSPs), a name coined by Hart.<sup>12</sup>

When considering a PSP for sampling aerosols, it may also be necessary to account for changes in the penetration convention for particles with changes in breathing rate. The current fixed flow methodology considers neither changes in aerosol mass penetration by changes in air volume breathed nor changes in rate of breathing. However, the interaction between a changing sampling rate and a changing respirable convention would be difficult to account for in practice. Calculations based on the most recent International Commission on Radiological Protection model<sup>16</sup> for alveolar deposition over the standard range of respirable aerosol size-distributions indicate a range of bias in collected mass (*e.g.*, sitting compared to moderate exercise) from approximately  $-50\%$  to  $+50\%$ , the magnitude depending mainly on the mass median aerodynamic diameter (MMAD) of the aerosol.<sup>17</sup> See Fig. 2.

## Prior art

Development of a PSP presents several design challenges. The subject's inhalation must be determined in real time. The pump must then continually control sampling in proportion to the inhalation. Since inhalation rate may vary greatly from task to task, the sampling head must have a wide dynamic range, yet maintain a constant sampling efficiency over that range.



**Fig. 2** Bias map from Bartley<sup>17</sup> of mass deposition in the alveolar region of the human lung for moderate exercise compared to sitting for comparable aerosol exposures.

All prior PSPs varied pump speed to drive airflow to follow inhalation. This conventional approach used by Kucharski,<sup>10</sup> Levine,<sup>11</sup> Hart,<sup>12</sup> and Delhaye and Kielczewska<sup>18</sup> is detrimental to the performance in two ways: (1) the sluggishness and distortion in response and (2) the indeterminate collection efficiency. As discussed in detail by Lee *et al.*,<sup>13</sup> when a PSP changes speed, the inertia of the pump motor limits its response time and the fidelity with which the airflow tracks inhalation. The magnitude of this error is a function of the rate of change in respiration, the rate of change in airborne contaminant concentration, how frequently the pump speed is updated, *e.g.*, once a second, and the mechanical characteristics of the pump itself.

More detrimental to performance, however, is the effect of changing pump speed, which in turn changes airflow and particle velocity.<sup>19,20</sup> As can be seen from the efficiency curves shown in Fig. 1, a particular cyclone operating at  $2.5 \text{ L min}^{-1}$  flow that captures on its filter particles having a  $d_{ae} = 4 \mu\text{m}$  at about 50% efficiency might collect the same size particles at over 90% efficiency when operating at  $1.9 \text{ L min}^{-1}$ . Without knowing the collection efficiency the samples are quantitatively worthless.

The gas and vapor PSP<sup>13</sup> obviated the problem of sluggish, distorted response by keeping the pump flow constant while a 3-way valve directed the airflow through a charcoal tube for the fraction of each second necessary to keep the volume of air sampled proportional to inhalation. The valve then diverted the flow through another charcoal tube for the remainder of each second. The equivalent of a variable flow was thereby produced through the PSP sampling medium that Lee *et al.*<sup>13</sup> referred to as the effective flow. By using a valve instead of varying pump speed, the response time was about 100 times faster than prior art. Moreover, any residual timing errors were easily offset by using a simple calibration routine since the settling time for discontinuities in the airflow due to valve transitions was independent of the magnitude of change in inhalation.

Lee *et al.*<sup>13</sup> evaluated this version of the PSP using both human subjects and a simulator. The human subjects wore a portable respiratory inductive plethysmography system that supports optional sensors, all embedded within a vest, and known as a LifeShirt® (Vivometrics® Inc., Ventura, CA, USA). Cumulative air volume through the sampling medium during three 80 min acquisitions was within an average of 2.5% of that commanded (as derived from heart rate and motion index sensors within a LifeShirt®, together with the subject's age, gender, height, weight, and resting heart rate processed each second by a regression equation in the PSP). This error was reduced to 0.2% when the same data were rerun on a simulator connected to the PSP after the PSP was updated with software that corrected a math error caused by truncation in the 24-bit floating point calculations. 60 s averages of effective flow through the sampling medium were tracked within 0.3%, 0.7%, and 0.2% below that commanded by the simulator. The total system error for cumulative volume was examined for only one subject, since evaluation of the LifeShirt® used for calculating inhalation was the focus of other studies.<sup>14,21,22</sup> Total system error averaged 6% ( $n = 3$ ) compared to the cumulative volume measured by a pneumotachometer.

Lin *et al.*<sup>9</sup> evaluated this version of the PSP with 15 healthy human subjects who wore the LifeShirt® vest and PSP while performing step exercises at gradually increasing rates inside of

a specially designed chamber. Each subject was exposed to *m*-xylene while exercising for 80 min on 4 different days. During 2 sessions *m*-xylene was released into the chamber in concentrations that were highly correlated ( $\rho = 0.93$ ) with the subject's exertion level (breathing rate). During the other 2 sessions concentrations were uncorrelated ( $\rho = 0.02$ ). When correlated, concentrations of *m*-xylene collected by the PSP averaged 19% higher than the TWA concentrations of *m*-xylene collected by the constant flow pump. No statistically significant difference in concentrations of *m*-xylene was detected for the uncorrelated sessions.

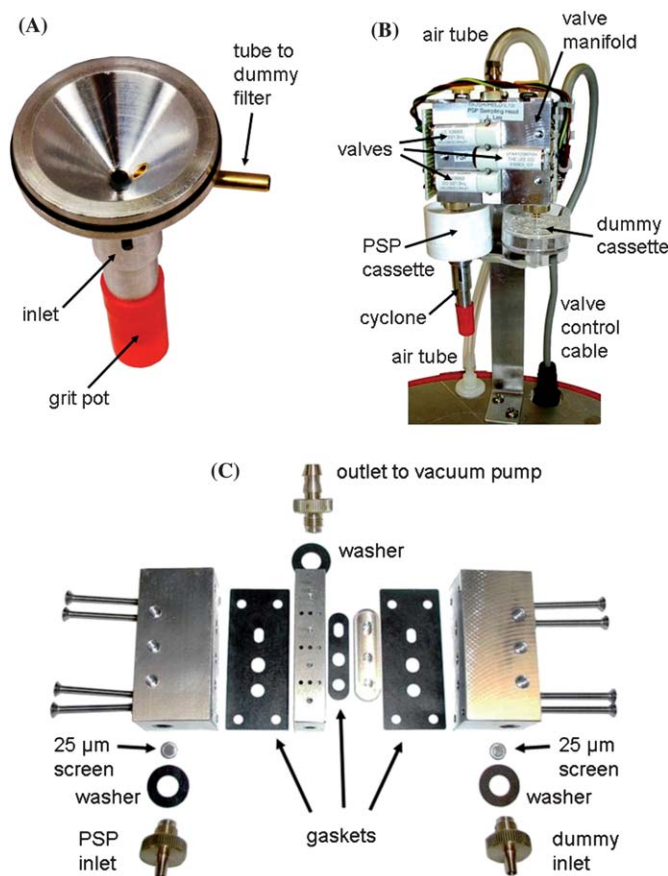
### A functional prototype

A sampling head was designed and built to evaluate the feasibility of conducting size-selective physiologic sampling of particulates. A complete aerosol PSP was not required for this evaluation, since experiments using the gas and vapor PSP tested all other system components, *e.g.*, the valve control electronics and the LifeShirt®. Moreover, only minor programming changes would need to be made to the gas and vapor PSP. For example, an inhalation rate of  $100 \text{ L min}^{-1}$  might be programmed to correspond to a 100% valve duty cycle, making an inhalation rate of  $10 \text{ L min}^{-1}$  correspond to a 10% valve duty cycle, *etc.* For a PSP that maintains a  $2.2 \text{ L min}^{-1}$  airflow, this scenario sets the

scaling factor at approximately 45.5 (the 100% duty cycle inhalation rate of  $100 \text{ L min}^{-1}$  divided by the  $2.2 \text{ L min}^{-1}$  pump flow). The amount collected on the sampling filter would then be multiplied by this scaling factor to determine actual exposure.

An aluminium cyclone was slightly modified to become an integral part of the functional prototype (see Fig. 3A). A 3.25 mm diameter hole was drilled through the side of the funnel and perpendicular to the exit hole near the bottom of the cyclone funnel. The bottom edge of this hole was about 1 mm above the exit hole. A 25 mm long, 3.2 mm outer diameter brass tube was machined with one end tapered to match the contour of the funnel. The tube was inserted into the hole and glued to the funnel to form an airtight seal. A short length of flexible silicon tubing connected the brass tube to a right angle barbed elbow adapter, which in turn was glued into the threaded inlet of the dummy cassette. Particle loss in the elbow was irrelevant since collection on the dummy filter was not measured. This modification provided a path for the airflow to be diverted from the PSP filter in order to keep the volume of air sampled proportional to inhalation while allowing the flow through the cyclone, and therefore the efficiency of separation within the cyclone, to remain constant.

Fig. 3B shows the assembled sampling head. Electrical signals from the valve control cable determine the state of the valves which switch the airflow between the PSP and dummy filters. Lee



**Fig. 3** The PSP cyclone sampling head: (A) the modified cyclone and (B) the sampling head ready for testing. Three valves are visible; the other 3 are mounted on the back of the valve manifold. The sampling head was easily removed from the mounting bracket by loosening two thumbscrews on top of the valve manifold, then sliding the head forward and out from two slots. The air tube and valve control cable plugged into the bottom plate of the test chamber. (C) Exploded view of the valve manifold.

Company (Essex, CT, USA) LFAA1208010H three-way valves that have a typical response time of about 3 ms were selected to give a fast response. Six of these valves were used in parallel to lower the pressure drop across the manifold. The valves were configured as two 3-valve banks to enhance the balance in the air streams, and thereby, reduce the magnitude of discontinuities. One bank of valves is actuated while the other is deactivated. Except during valve transitions, at any instant air flows through only one of the filters, that flow being the sum of the flow through the bank of actuated valves plus the flow through the bank of deactivated valves.

Fig. 3C shows an exploded view of the valve manifold. O'Keefe Controls, Co. (Monroe, CT, USA) SSB-5 in-line screens placed within the inlet ports protect the miniature valves from potential particle breakthrough in the filters and support pads, or if the airflow is turned on inadvertently without a filter installed inside of a cassette. Slight imbalances in airflow through the manifold are inherent since machining of the center (outlet) airway required entry of the milling tool from one side. As can be seen in the exploded view, an aluminium insert and a small gasket fit into the right side of the center section to fill the unwanted void. The manifold could be greatly simplified if produced as an integral part of a two-piece molded (conductive) plastic sampling head that would fit together to secure the filter elements and support pads. Such a molded plastic version would favor mass production, be smaller and lighter, thereby increasing the user acceptance in the field, and further improve balance in airflow.

The duration and magnitude of airflow disruptions during valve transitions were measured. Even though the valves typically change states in 3 ms, total response time of the sampling head extends beyond valve transition until airflow settles to the steady state. To determine the total response time and magnitude of the disruptions, the laboratory vacuum was connected through an Aalborg, Inc. (Orangeburg, NY, USA) model GFC17 mass flow controller (MFC) to the outlet port of the sampling head; a TSI, Inc. (Shoreview, MN, USA) model 4143 Thermal Mass Flow meter was connected by a calibration adapter at the cyclone inlet; and a Tektronix, Inc. (Beaverton, OR, USA) TDS-754A oscilloscope was connected to the analog output from the flow meter to monitor the airflow. With the sampling head valves set to channel air through the PSP filter, the MFC was adjusted until the flow meter reported a  $2.2 \text{ L min}^{-1}$  of volumetric flow. The valves were then cycled and the oscilloscope

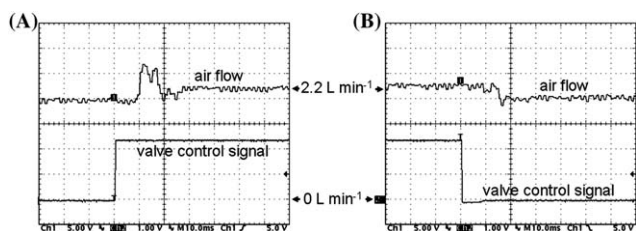
displays saved to disk. As recorded in the oscilloscope plots presented in Fig. 4, when airflow was switched from going through the dummy filter to going through the PSP filter, perturbations in the flow settled to within 10% in about 15 ms. When airflow was returned to the dummy filter, flow settled to within 10% in under 10 ms.

As can also be seen in Fig. 4, the steady state flow through the dummy filter was about  $2 \text{ L min}^{-1}$ , or 9% less than the  $2.2 \text{ L min}^{-1}$  flow through the PSP filter. This difference in steady state flow between the two paths is the result of unavoidable imbalance in machining of the prototype, and underscores the importance of using valves having a fast response, as well as the requirement that calibration be performed while airflow is directed through the collection medium, *i.e.*, the PSP filter, *vice* the dummy filter.

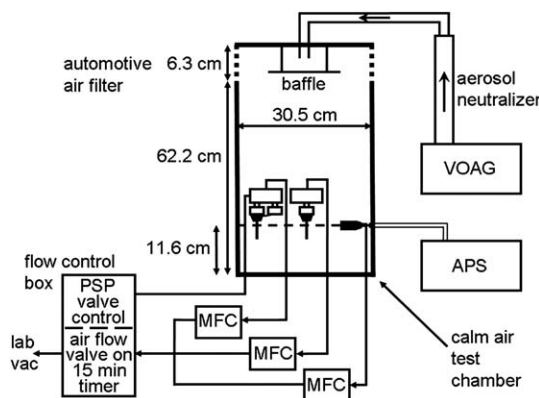
## Experimental

A  $5 \times 9$  full factorial design was used to investigate performance of the prototype (particles having approximate  $d_{ae} = 2, 3, 4, 5,$  and  $6 \mu\text{m}$  and duty cycles of 0, 5, 10, 25, 50, 75, 90, 95, and 100%). Each combination was replicated either 3, 4 or 6 times. To minimize part-to-part variations in performance, 6 new aluminium cyclones were tested. Two of these 6 that had the most similar collection efficiencies were selected for use in this study: one was used in the reference sampler and the other was subsequently modified and used in the PSP sampling head. The collection efficiencies of these 2 cyclones were within 0.2% when exposed to Kaolin [MMAD  $2.21 \mu\text{m}$ ; GSD 1.78, as determined by a TSI, Inc. Aerodynamic Particle Sizer (APS)] in a calm air chamber.<sup>23</sup> Changes in performance caused by the modification made to the cyclone were isolated from changes in performance caused by switching the valves in experiments that did not redirect the airflow (0% and 100% duty cycles). Performance as a function of inhalation rate was simulated by the 9 duty cycles.

The experimental setup is diagrammed in Fig. 5. A TSI, Inc. model 3450 vibrating orifice aerosol generator (VOAG) generated monodisperse ammonium fluorescein particles having a density of  $1.35 \text{ g cm}^{-3}$  (as discussed by Vanderpool and Rubow<sup>24</sup>). These particles passed through a TSI, Inc. model 3054A Kr-85 aerosol neutralizer, and then into an aluminium calm air chamber. This same chamber when used by Feather and Chen<sup>25</sup> and by Lee *et al.*<sup>26</sup> provided a uniform distribution of the



**Fig. 4** Airflow measured at the inlet of the cyclone during valve transitions: (A) disruption settles to within 10% in about 15 ms when flow is switched from the dummy filter to the PSP filter and (B) in under 10 ms when flow is switched from the PSP filter to the dummy filter. Noise on the airflow traces is due to artifacts of the  $1000 \text{ samples s}^{-1}$  digital output from the thermal mass flow meter.



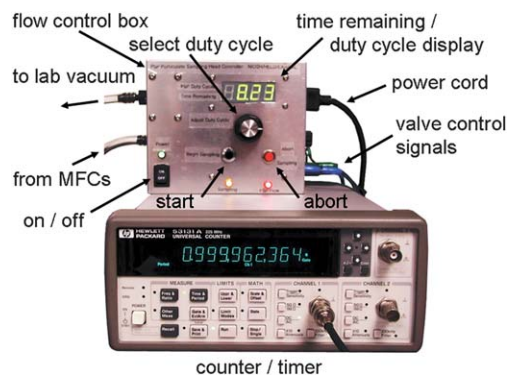
**Fig. 5** The experimental setup.

airborne particles. The original bottom plate was replaced, however, with one that allowed easier and less time consuming setup, and that accommodated mounting the functional prototype (see Fig. 3B), another aluminium cyclone used as a reference cyclone (not modified), and a horizontal reference in a 50 mm extension cowl. To help equalize inlet aspiration efficiency between the cyclones, the reference cyclone was mounted underneath a dummy valve manifold similar in shape to the one used by the PSP sampling head. Since our data showed the variation in particle dispersion throughout the chamber ranged from 1.7% to 4.4% for particles having  $d_{ae} = 4 \mu\text{m}$  and from 1.8% to 4.8% for particles with  $d_{ae} = 7 \mu\text{m}$ , placement of these 3 samplers was not randomized during the experiments. A TSI, Inc. model 3321 APS was used before and after each sampling session to verify the size distribution and concentration of the particles generated. The regulated laboratory vacuum provided a pure source of airflow without the pulsations that typically modulate personal sampling pumps and that might confound the experimental results.<sup>20</sup>

Fig. 5 also shows the setup used to control airflow through the 3 samplers. Before each sampling session the operator rotated the knob on the flow control box (FCB) until the required PSP duty cycle was displayed. Then the operator pressed the start button on the FCB which automatically opened a valve inside the box so that the laboratory vacuum pulled air through the Aalborg, Inc. model GFC17 MFCs and the samplers. During each 15 min sampling session the FCB displayed the time remaining while also exercising the valves in the PSP sampling head each second at the preset duty cycle. When time expired the FCB shut off airflow through the samplers.

Fig. 6 shows the FCB connected to a Hewlett Packard, Inc. (now Agilent Technologies, Inc., Santa Clara, CA, USA) model 53131A Universal Counter setup in timer mode. In this figure, the Universal Counter displays the 1 s update period as 0.999962364 s. Any such absolute timing errors are of no consequence, however, since all timing was derived from a common clock within the FCB, forcing all timing to be ratio-metric.

Airflow was calibrated before sampling sessions using a TSI, Inc. model 4143 Thermal Mass Flow meter configured to display volumetric flow. The FCB was set to 100% duty cycle and the start button was pressed to begin the calibration. The flow set



**Fig. 6** The Flow control box atop a Hewlett Packard, Inc. model 53131A Universal Counter.

screw on each Aalborg, Inc. model GFC17 MFC was then adjusted until the flow stabilized to  $2.2 \text{ L min}^{-1}$  for each corresponding sampler. Flow through the PSP sampling head was calibrated through the PSP filter, not the dummy filter. Both the PSP sampling head and the reference cyclone used  $5 \mu\text{m}$  pore size PVC filters and support pads in 37 mm plastic cassettes. The horizontal reference used the 25 mm version of the same filter and support pad (the inlet diameter was checked to assure it would meet both the inertial and gravitational settling conditions of the Davies criterion<sup>27</sup>). An SKC, Inc. Aero-Chek Cassette Leak Tester, model 225-8531, was connected to the cassettes before each sampling session to assure integrity of the assembled cyclone samplers.

After each sampling session the filters were removed from the cassettes and the samples analyzed. Each filter was placed in a separate container containing 5% ammonium hydroxide which extracted the fluorescein that was collected. A Perkin-Elmer (Waltham, MA, USA) model LS50B luminescence spectrometer was then used to measure the fluorescent intensities and to thereby determine the amounts of particles collected by the PSP sampling head and by the reference cyclone relative to the horizontal reference sampler. The cassettes and cyclones were washed to prevent particle accumulation from biasing subsequent experiments.<sup>28</sup>

Efficiency curves were plotted from the corresponding fluorescein intensity ratios adjusted by the applicable duty cycles, *e.g.*, for a 25% PSP duty cycle, the fractional collection efficiency equals the intensity on the PSP filter divided by the intensity on the horizontal reference filter divided again by 0.25. A sigmoid function with 3 parameters was used to generate each curve. The reference cyclone always sampled at a 100% duty cycle since it was a part of a conventional sampling train instead of being regulated by valves. Bias maps were then plotted by comparing these efficiency curves to the ISO/CEN/ACGIH respirable convention by determining the bias for particle-size distributions with MMADs up to  $25 \mu\text{m}$  and GSDs between 1.5 and 3.5 per Bartley *et al.*,<sup>29</sup> Kenny and Bartley,<sup>30</sup> and Aitken *et al.*<sup>31</sup> Bias maps were also plotted to compare performance of the PSP sampling head directly to the commercially available cyclone.

## Results and discussion

Modification of the cyclone had negligible effect on performance. Ideally, for the 0% duty cycle case, all particles exiting the cyclone would be diverted away from the PSP filter. Instead, an average of 0.6% ( $n = 18$ ) of the amount of the particles collected on the horizontal reference sampler was deposited on the PSP filter. There was no apparent relationship between the amounts collected and particle size (0.5, 1.4, 0.5, 0.5, and 0.4% for particles having  $d_{ae} = 2.12$  (CV = 0.06), 2.98 (CV = 0.02), 3.98 (CV = 0.01), 4.96 (CV = 0.02), and 5.89 (CV = 0.03)  $\mu\text{m}$ , respectively). These negligible deposits on the PSP filter suggest that for other duty cycles, the efficiencies reported were not biased by particle leakage during the fraction of each second that the airflow was being diverted through the dummy filter. Moreover, for the 100% duty cycle case, the collection efficiency of the PSP sampling head would ideally be equal to the reference cyclone, but was found to be an average of 3.9% higher. There was no apparent relationship between the difference in collection efficiencies for the 100% duty

cycle experiments and particle size (3.6, 2.6, 7.8, 4.6, and 0.6% for particles having  $d_{ae} = 2.12, 2.98, 3.98, 4.96,$  and  $5.89 \mu\text{m}$ , respectively). This result is inconsistent with a negative bias that might be attributed to drag that might be created from the edge of the hole machined in the funnel, and likely represents experimental error. Any significant deviation in performance between the 2 cyclones should, therefore, be attributed to transient perturbations in airflow induced by valve switching.

Fig. 7A shows two efficiency curves plotted from samples collected during 157 experimental runs. See eqn (1) and (2) for the corresponding equations. These curves are almost identical, but present steeper slopes than the respirable convention curve. The 50% cut point for the modified cyclone was at  $d_{ae} = 4.2 \mu\text{m}$ , and for the reference cyclone, at  $d_{ae} = 4.1 \mu\text{m}$ . PSP data from 18 experiments that used the 0% duty cycle was excluded from the PSP curve since, as discussed, collection was negligible for all particle sizes. The 0% duty cycle data were also excluded because of the indeterminate resulting from division by zero when adjusting intensity ratios to account for duty cycle as described above.

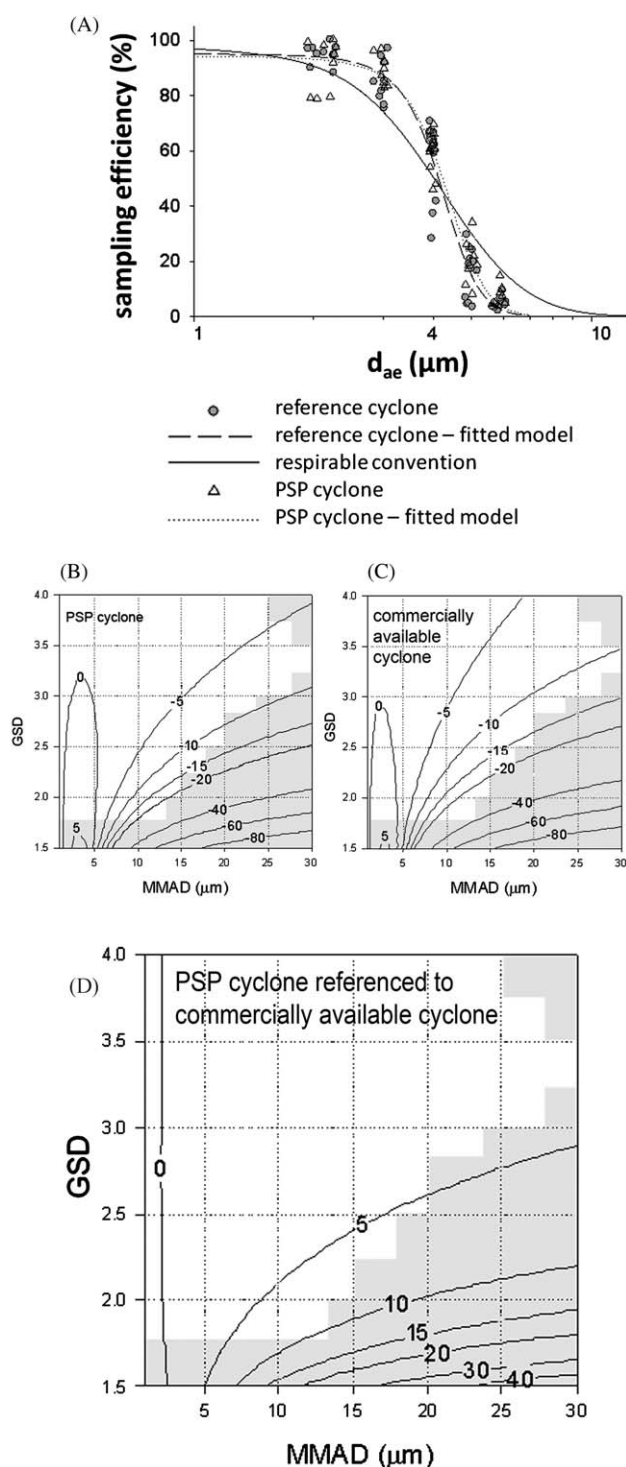
$$y = \frac{94.44}{1 + e^{-(x-4.29/-0.53)}} \quad (1)$$

$$y = \frac{95.20}{1 + e^{-(x-4.18/-0.49)}} \quad (2)$$

The above sigmoidal equations were used to plot the efficiency curves shown in Fig. 7A. Eqn. (1) was used for the PSP sampling head plot. Eqn (2) was used for the reference plot.

According to European Standard EN13205,<sup>32</sup> only particles having certain size distributions are to be considered when evaluating the performance of respirable samplers. The Committee's criteria includes those particles of most interest for workplace aerosol sampling, specifically the respirable fraction when it amounts to at least 5% of the total aerosol mass. Table A2 of EN13205<sup>32</sup> presents these findings as a two dimensional matrix, MMAD *versus* GSD, which for the respirable fraction includes 224 size distributions. Fig. 7B–D overlay the excluded aerosol size distributions as shaded regions on bias maps, so that the practical merits of both the modified and the commercially available cyclones may be readily seen. These bias maps not only confirm that both samplers exhibited similar performance, but also reveal that for nearly all the size distributions of concern both cyclones exhibited between –20% and +5% bias relative to the respirable convention. Moreover, the efficiencies of the two cyclones differed by <10% from each other over these same size distributions. Any influence that duty cycle may have had on performance requires further examination, however, since the PSP plots in Fig. 7 were generated using data combined from experiments that energized the valves for all of the 8 non-zero duty cycles.

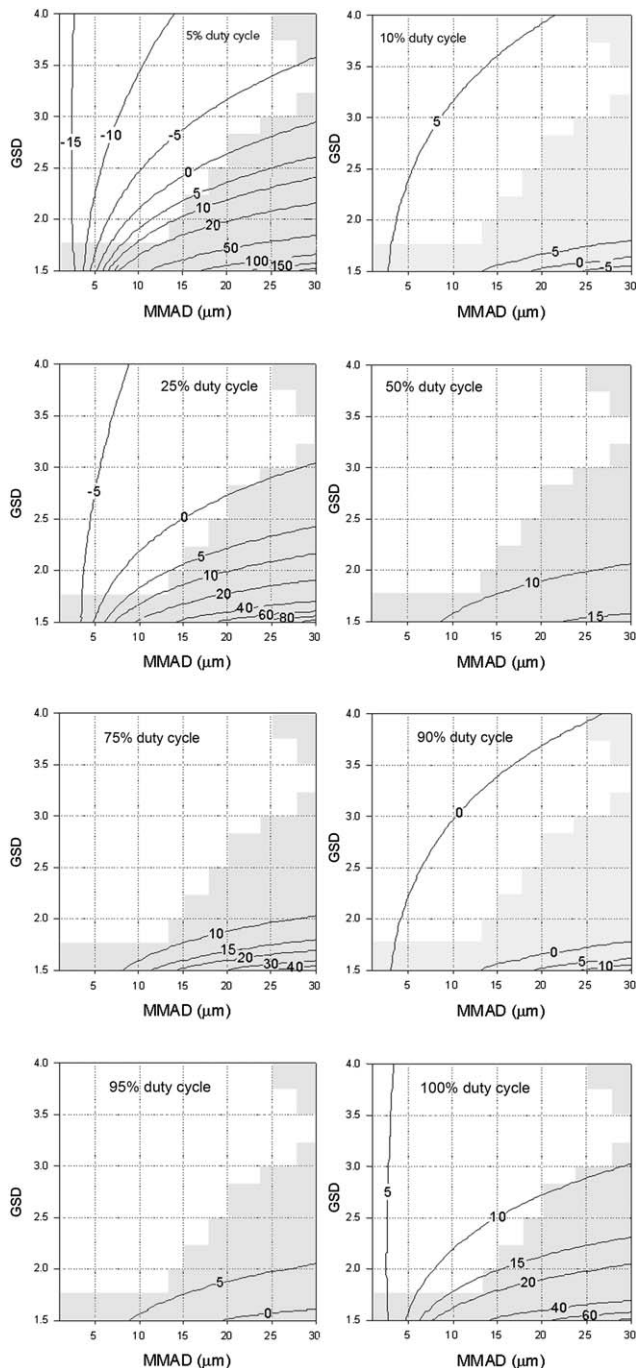
Bias maps comparing the performance of the 2 cyclones for the non-zero duty cycles tested are shown in Fig. 8. For the size distributions of interest, performance exhibited <10% bias, except for the 5% duty cycle case, when the duration of the air perturbations occupied the largest proportion of each sampling interval, that is, about 20% to 30% of the 5% duty cycle. Performance did exhibit bias up to 20% for some size



**Fig. 7** Performance of 2 aluminium cyclones: one modified for use in the PSP sampling head and the other as commercially offered. Airflow was  $2.2 \text{ L min}^{-1}$ . (A) Sampling efficiency for the two cyclones compared to the ISO/CEN/ACGIH respirable convention. Each data point represents the average of 3, 4 or 6 replicates of the parameters tested. Data are from all the experiments that exercised non-zero duty cycles. Bias of (B) the modified cyclone, and (C) the reference cyclone, each referenced to the convention. (D) Bias of the modified cyclone referenced directly to the reference cyclone. Per European Standard EN13205,<sup>32</sup> size distributions shaded on the bias maps are to be excluded when evaluating performance.

distributions at the 100% duty cycle, but this is of no practical consequence since the maximum inhalation rate would rarely, if ever, be experienced. Penetration data for each duty cycle plot are available ESI†.

Fortunately, errors introduced by sampling at duty cycles <10% or so may be avoided in 3 ways. Firstly, the scaling factor



**Fig. 8** Bias maps comparing the performance of the modified cyclone used in the PSP sampling head to the reference cyclone used in the conventional manner. The reference cyclone was always sampling, *i.e.*, 100% duty cycle. Per European Standard EN13205,<sup>32</sup> size distributions shaded on the bias maps are to be excluded when evaluating performance.

may be decreased from 45.5. Doing so is certainly practical, as the 100 L min<sup>-1</sup> inhalation rate that was assigned to the 100% duty cycle provides a dynamic range greater than needed. By decreasing the scaling factor, the duty cycle corresponding to any particular inhalation rate increases. Secondly, instead of using the same scaling factor for all subjects being sampled, the resting inhalation rate of a user may be determined, entered into the PSP, and programmatically assigned to the 10% duty cycle. This would force operation between 10% and 100% duty cycles while still accommodating a 10 to 1 operating range mentioned by Satoh *et al.*<sup>15</sup> Thirdly, as a person's inhalation rate changes, the duty cycle of the valves must be updated to keep sampling proportional to inhalation, hence the term "update interval." For our experiments we used a 1 s update interval. When inhalation rates are very low, instead of incurring the greater error observed with the associated short duty cycles, *e.g.*, a 5% duty cycle, the PSP could simply not sample at all for one or more of these update intervals, but instead, could add these sampling durations to a subsequent update interval, thereby forcing all sampling to duty cycles >10%.

## Conclusions

Traditional sampling pumps cannot follow inhalation, but operate at a constant, preset airflow that allows sampling with a predictable collection efficiency. Older designs for physiologic sampling pumps attempt to follow inhalation by varying airflow, but in so doing, make collection efficiency indeterminate. The method presented herein preserves a predictable collection efficiency while sampling in proportion to inhalation, and provides a technology to better assess personal exposure to airborne hazards.

## Disclaimer

The mention of any company names or products does not imply an endorsement by NIOSH or the Centers for Disease Control, and nor does it imply that alternative products are unavailable, or unable to be substituted after appropriate evaluation. The findings and conclusions in this report are those of the author(s) and do not necessarily represent the official position of the Centers for Disease Control and Prevention.

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