

Direct sampling of organic solvents in expired breath with a new solid sorbent sampling device

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Monitoring exhaled breath is a useful technique for estimating the total amount of volatile organic solvents absorbed by individual workers through both inhalation and dermal routes. The use of this procedure for routine industrial hygiene applications has been limited by the lack of techniques to collect and concentrate trace levels of solvents found in expired breath directly.

A new device that uses solid sorbents for the direct sampling of solvents present in mixed-expired breath has been designed (Glaser & Arnold, unpublished results). The samples, once collected, may then be transported back to the laboratory and stored for later analysis. The sampler, constructed of stainless steel, operates on the same principle as an air-purifying respirator. Subjects bite the mouthpiece and inhale purified breathing air through conventional charcoal inhalation canisters. The exhaled air is sampled from the mainstream on 45-mm wafers of charcoal cloth sorbent or from the sidestream on other sorbents.

The goal of the research reported in this paper was to compare concentrations of m-xylene in breath obtained with this device to those obtained with a conventional end-expired sampling procedure following controlled exposures of six subjects to 75 ppm of m-xylene in air. Mixed-expired breath was simultaneously sampled for m-xylene from the mainstream with the charcoal cloth sorbent and from the sidestream with Tenax TA. End-expired breath was sampled for m-xylene with the use of bags. The carbon dioxide concentrations of the mixed- and end-expired samples were also determined.

Methods

Subjects and exposures

Six male volunteers, ranging in age from 20 to 29 years and weighing from 62.6 to 113 kg, participated in the study. The subjects had no history of occupational exposure to solvents. Prior to the exposures, each subject received a complete medical examination. Since m-xylene is largely metabolized by the liver (3, 4), this examination included a complete screen of liver function for preexisting liver disease.

Three separate exposure sessions were held. At each session, two subjects were exposed to 75 ppm of m-xylene in air for 4 h in a controlled environmental exposure chamber maintained at 70°C and at 50 % relative humidity. Only sedentary activities were permitted during the exposures.

Sampling and analysis

After each 4-h exposure session, breath samples were obtained at approximately 0, 5, 10, 15, 30, 45, 60, 75, 90, 120, 150, 180, 210, and 240 min. Each subject contributed a breath sample at each of the 14 scheduled sample intervals. Samples were collected in an alternating nonrandom fashion. While one subject contributed an end-expired sample, the other contributed the mixed-expired samples; the sampling order was then reversed. End-expired breath was sampled in bags according to the 30-s breath holding procedure of Jones et al (2).

Mixed-expired breath was sampled for m-xylene from the mainstream with a four-wafer primary sorbent bed and a two-wafer backup bed of charcoal cloth. The volumes sampled ranged from 5.5 to 22 l. Mixed-expired carbon dioxide was sampled in a gas-sample bag attached downstream of the stainless steel sampler. Sidestream samples were collected at 50 ml/min, with a 100-mg primary bed and a 50-mg backup bed of Tenax TA.

All the m-xylene samples were analyzed with gas chromatography with flame ionization detection. The bag samples were analyzed with a gas chromatograph equipped with a gas sample loop. The charcoal cloth samples were desorbed in carbon disulfide. The Tenax TA samples were thermally desorbed. The carbon dioxide concentrations were determined on site with a medical gas analyzer.

Results and discussion

The breath concentrations of m-xylene obtained for all six subjects are plotted in figure 1. On the average, each breath sampling and analytical method indicated approximately an order-of-magnitude decrease in the breath concentrations of m-xylene throughout the 240-min desaturation period. The concentrations of m-xylene in the end-expired breath samples were comparable to those reported previously following exposures to the same amount of p-xylene (1).

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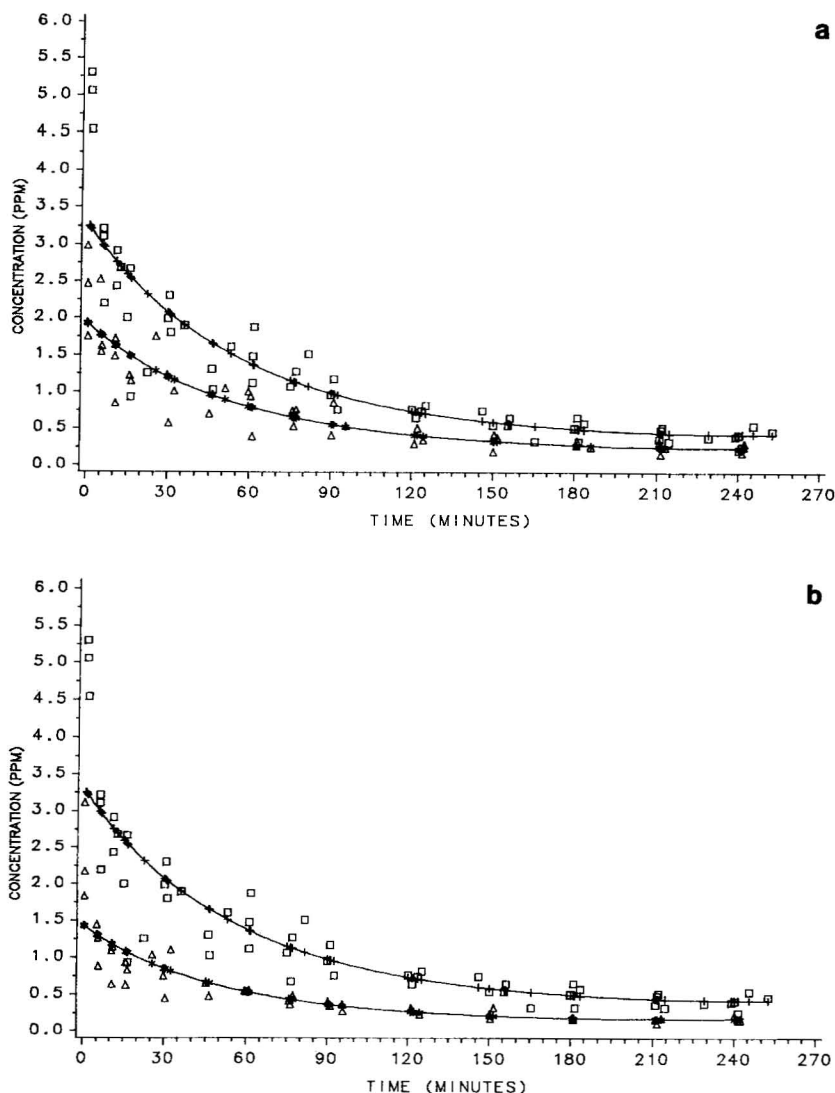


Figure 1. Plots showing the decrease in concentration of m-xylene in the breath of six subjects following exposure to 75 ppm of m-xylene for 4 h. Figure 1a compares the charcoal cloth and bag sampling results. Figure 1b compares Tenax and bag sampling results. For times of 1 min or less equation 1 requires an indicator variable to fit the data correctly. ($\square$ = concentrations obtained via bag sampling, Δ = concentrations obtained via charcoal cloth or Tenax sampling, —+— = the line fitted to the bag data, —*— = the line fitted to the charcoal cloth or Tenax data)

No significant capacity problems were encountered with the charcoal cloth for mainstream sampling. Six percent of the sidestream samples collected with Tenax TA exhibited significant breakthrough; significant corrections to the sidestream concentrations for blanks were required.

Linear regression analyses of the data obtained using each breath sampling method showed that the release of m-xylene via the breath was best described by the quadratic exponential decay function:

$$Y = e^{(\alpha + \beta t + \gamma t^2 + E)}, \quad \text{equation 1}$$

where Y is the concentration at time t and α , β , and γ are constants and E is the residual error of the de-

saturation curve. Quadratic exponential decay curves were fitted to the breath concentration data obtained for each subject for all three sampling and analytical procedures. The bias of each method relative to the others was determined from this regression analysis. Ratios of mixed- to end-expired m-xylene concentrations determined by charcoal cloth relative to bag sampling ranged from 0.48 to 0.67 (mean 0.58, relative SD 0.13). These ratios agreed well with the individual ratios of mixed- to end-expired carbon dioxide, which ranged from 0.55 to 0.70 (mean 0.61, relative SD 0.08).

For Tenax TA relative to bag sampling, the ratios of mixed- to end-expired m-xylene concentrations

ranged from 0.33 to 0.56 (mean 0.43, relative SD 0.24). Neither the overall ratio nor the individual ratios agreed with the respective ratios of mixed- to end-expired carbon dioxide.

The residual errors of the fitted desaturation curves for each subject provided estimates of the relative standard deviations of each sampling and analytical method. These estimates were pooled to provide an estimate of the relative standard deviation of the overall sampling and analytical methods of 0.13, 0.19, and 0.14 for mainstream, sidestream, and end-expired sampling, respectively.

For all three sampling and analytical procedures, nonlinear regression analysis of the breath data also showed that loss of the m-xylene from the body via the breath could be described with a three-compartment pharmacokinetic model of the form

$$Y = Ae^{-k^1t} + Be^{-k^2t} + Ce^{-k^3t} + E, \quad \text{equation 2}$$

where Y is the concentration at time t and A , B , and C are constants and k^1 , k^2 , and k^3 are the approximate desaturation rates of the individual compartments and E is the residual error of the desaturation curve. For the three methods, k^1 ranged from 0.23 to 0.43 min^{-1} , k^2 ranged from 0.013 to 0.016 min^{-1} , and k^3 was 0. Estimates of the sampling and analytical method bias and precision obtained from this statistical analysis were comparable to those obtained with the linear least squares analysis.

We conclude that mainstream sampling of mixed-expired breath with charcoal cloth was as precise as sampling end-expired breath with bags. On the basis of physiological considerations, the bias of mainstream mixed-expired relative to end-expired m-xylene concentrations was also reasonable. The higher variability

and negative bias of sidestream relative to mainstream sampling were likely due to an inaccurate estimation of the volume sampled via the sidestream and to an overcorrection of certain of the sample masses for blanks that were actually Tenax TA artifacts. It may be possible to circumvent these problems by sampling with other sorbents and measuring the concentration of mixed-exhaled carbon dioxide as an internal standard.

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