

The Monitoring of Benzene Exposure by Air Sampling

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The information presented shows that it is not possible to ensure compliance with an absolute ceiling threshold limit for benzene.

The ANSI standard can be slightly modified to provide practicable working criteria of 10 ppm time-weighted average and not more than 2% probability of exceeding 25 ppm. It would, however, be simpler for the hygienist to work to a daily exposure dose limit of 5000 ppm-minutes, which is proposed here.

This criterion is tested against some typical log-normal distributions and is submitted to toxicologists for consideration.

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Introduction

BENZENE HAS BEEN described as radiomimetic. It would seem that this description is appropriate to the incidence of chronic disease which manifests itself by blood changes comparable to those produced by ionizing radiation and, as recently reported, by chromosomal damage. While both benzene and radiation can cause acute poisoning, the effects are not similar—perhaps due in part to the differing routes of access to the body.

While the effects of overexposure to benzene are commonly assessed by hematology, the possible risk arising from occupational exposure can be determined by one or more of three techniques. These are determination of the concentration of benzene in the air the man breathes, measurement of concentration of benzene in the breath he exhales after exposure may have occurred, and analysis of his urine for the principal metabolite—conjugated phenol. The first is a measure of his exposure, the second a measure of the amount absorbed, the third a measure of the amount of benzene metabolized in the body.

Improvements have been made in these techniques and have been evaluated by a

test exposure to a controlled concentration;¹ typical application in a field study using personal air samplers has been described.²

This paper considers the problem facing the practicing hygienist in the interpretation of air sampling results using current criteria. It does not present new toxicological information, and toxicity aspects are introduced only insofar as they influence air sampling criteria.

Air Sampling

At the present time the industrial hygienist is presented with a bewildering assortment of standards for assessing exposure. Some of the various national standards recently reported³ show limits varying from 6 to 100 ppm.

In the United States all the following standards are currently valid:

Threshold Limit (1970) ⁴	25 ppm ceiling
ANSI Standard (1969) ⁵	50 ppm peak
	25 ppm ceiling
	10 ppm time-weighted average

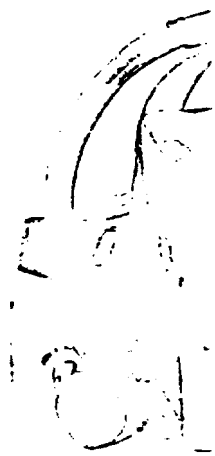


FIGURE 1. Intrinsic

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Hygienic Guide (1970)⁴ 25 ppm ceiling
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While it is entirely proper that threshold limits "should be interpreted and applied only by a person trained in this discipline,"⁴ some guidance from toxicologists on the significance of these values appears desirable.

Summary of Air Sampling Technique

As concentrations of substance in workplaces typically vary widely in space, it is necessary to collect the air from the breathing zone if results are to represent the air inhaled by a man moving about at work.

As concentrations typically vary markedly in time, it is necessary to take a series of measurements over a long enough period of time to represent average and extreme conditions.

Representative samples have been collected by means of a modification of the original personal air sampler⁷ which is intrinsically safe (Figure 1). This draws

air at a constant rate of 1 liter/min through cartridges containing silica gel attached to a harness on the shoulder.

Cartridges are changed every 10 or 15 minutes, and the benzene subsequently is eluted with ethanol in the laboratory. The quantity of benzene is determined by gas chromatography, and the concentration in the air calculated for each sample period.

The Distribution of Air Sample Results

It has already been shown⁷ that a series of observations of air concentrations of a radioactive aerosol arising in workplaces can be described reasonably accurately by a log-normal distribution, though this does not represent the sequence of samples with respect to time. This is also observed in outdoor pollution studies⁸ and could be expected for vapor concentrations which men breath while at work.

The average concentration to which a man is exposed is best described by the arithmetic mean of all the samples collected, weighted for the duration of the samples unless this is held constant throughout the period.

The variability of the concentration, which may be at least as important a factor, is best described by the geometric standard deviation (GSD) which is applied to the median (geometric mean) to determine the probability of a given concentration's being exceeded.

Field Results

Personal air samples, each of 15-minute duration, have been collected during a variety of operations where men are exposed to benzene vapor. Each series of samples has covered a whole working shift or a complete operation, with total times varying from 2 to 8 hours. Each set of results has been placed in rank order and plotted on logarithmic probability paper according to the system of Larsen.¹⁰ Some typical results are shown in Figures 2 and 3 with the computed distribution curves superimposed.

In Figure 2, which refers to exposure to pure benzene, curve 1 shows the marked

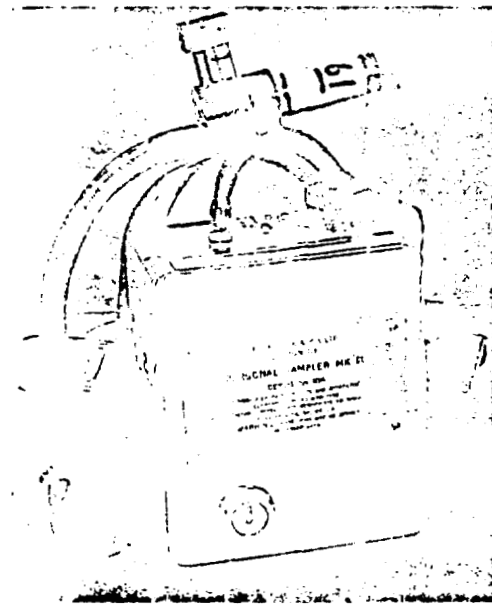


FIGURE 1. Intrinsically safe removal air sampler.

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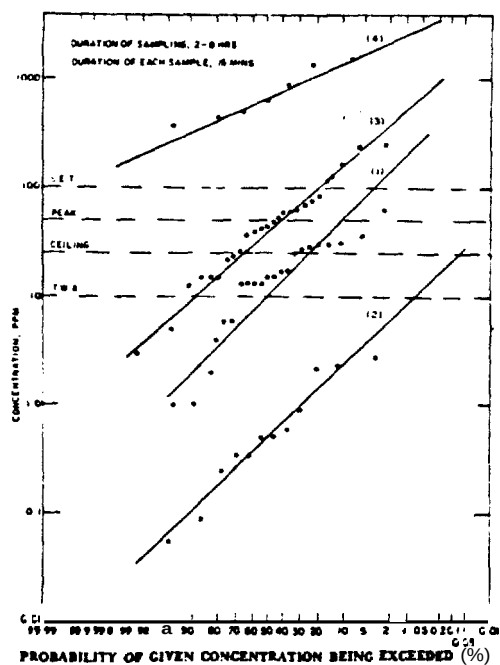


FIGURE 2. Distribution of air sample results for four men exposed to benzene in various operations.

variability (GSD 3.7) found when a man moves around point sources of emission in the open air approaching each very closely. Curves 2 and 3 show slightly lower values of GSD (3.4 and 2.9) for similar operations undertaken in more windy conditions. In contrast, curve 4 shows the small variability (GSD 1.6) when a man works in a confined space.

Figure 3 shows exposure when a hydro-

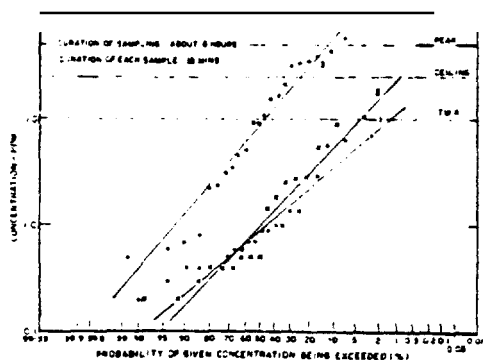


FIGURE 3. Distribution of air sample results for three men exposed to benzene in hydrocarbon mixtures.

carbon mixture containing benzene is emitted from a single source in a partially enclosed area. For two men working around the opening (curves 1 and 2) the GSD's are 2.9 and 3.8. For one man working in a confined space below the source of emission the GSD is 4.5 (curve 3).

Hypothetical Distributions

To enable comparisons to be made with the criteria published in the United States, three hypothetical distributions have been plotted in Figure 4. All three have a time-weighted average value (arithmetic mean) of 10 ppm and have geometric standard deviations of 1.5, 3.0, and 4.5 to represent least, typical, and most variable concentrations likely to be met in practice. The limit line proposed by Jones and Brief,¹¹ which is also shown, is discussed later.

Interpretation of Results Using Current Criteria

Provided that sufficient numbers of samples are taken under conditions representing long-term exposure, no difficulty should arise in interpretation using the time-weighted average value of 10 ppm; should be noted that in a log-normal distribution the arithmetic mean is not identical with the geometric mean (median).

The interpretation of results using short-term criteria is less straightforward. Specifically, the fact that results can be

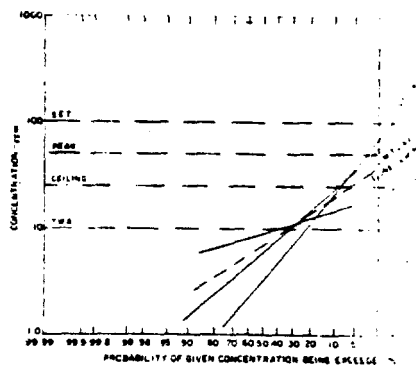


FIGURE 4. Typical and extreme values for series of air samples having an arithmetic (time-weighted average) of 10 ppm.

scribed by log-normal it is impossible to approach a value, as there is a finite probability of concentrations being exceeded to the level of the ceiling, which is about 100 ppm.

The evaluation of risk must therefore be made in terms of acceptable risk—that is, the probability of a given concentration being exceeded to an acceptable level.

Such a limit can be set by amendment of the ANI standard. A maximum for peak reasonable to protect against a 25 ppm average may be exceeded only once in a 10-hour working period. If 10-minute periods during the day, this can be restated in terms of the form that the probability of exceeding 25 ppm over a 10-minute period should not be greater than 1%.

Such a criterion makes it possible to assess short-term exposure on a log-normal plot or on a sample results. If the distributions having a time-weighted average of 10 ppm shown in Figure 4, by this standard, it can be seen that the area where the air concentration is relatively uniform (GSD 1.5) of the ceiling value's being exceeded is less than 1%. In more typical outdoor work the probability of exceeding the ceiling and therefore the corresponding peaks is more important than the time-weighted average.

From Figure 4 it can be seen that for concentrations having a time-weighted average of 10 ppm and a probability that the arithmetic mean of 100 ppm would be exceeded of 0.5% (one sample in 200 for extreme case of a GSD of 4.5), the probability is 1% (that is, one sample in 100) for a GSD of 3.0. It is not possible to set a limit on the biological significance of the distribution.

In a recent paper, Jones and Brief¹¹ proposed a rational procedure for preparing air sampling results.

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tribed by lognormal distributions makes it impossible to apply an absolute ceiling value, as there is a finite probability of any concentration's being exceeded (up to that corresponding to the saturated vapor pressure, which is about 100,000 ppm at 20°C).

The evaluation of short-term exposure must therefore be made on the concept of acceptable risk—that is, restricting the probability of a given concentration's being exceeded to an acceptable value.

Such a limit can be developed by slight amendment of the ANSI criteria for the acceptable maximum for peaks. It would appear reasonable to propose that a concentration of 25 ppm averaged over 10 minutes may be exceeded only once during an 8-hour working period. As there are 48 10-minute periods during such a working day, this can be restated with adequate accuracy in the form that the probability of exceeding 25 ppm over any 10-minute period should not be greater than 2%.

Such a criterion makes it immediately possible to assess short-term exposure from a log-normal plot or calculation of the air sample results. If the hypothetical distributions having a time-weighted average of 10 ppm shown in Figure 4 are assessed by this standard, it can be seen that in an area where the air contamination is relatively uniform (GSD 1.5) the probability of the ceiling value's being exceeded is less than 1%. In more typical conditions of outdoor work the probability is about 10%, and therefore the control of short-term peaks is more important than control of the time-weighted average concentration.

From Figure 4 it can also be seen that for concentrations having a time-weighted average of 10 ppm and a GSD of 3.0, the probability that the AIHA short-term tolerance of 100 ppm would be exceeded is 0.5% (one sample in four shifts); for the extreme case of a GSD of 4.3, the probability is 1% (that is, one sample in two shifts). It is not possible to comment on the biological significance of such values.

In a recent paper, Jones and Brief¹¹ have proposed a rational procedure for comparing air sampling results with the ANSI

criteria using the 10-ppm time-weighted limit and the 50-ppm maximum for a single 10-minute value. They make use of Larsen's method for plotting log-normal curves and have taken account of the effect of sampling time on geometric standard deviation which has been ignored here.

The limit line they derive for 15-minute sampling periods is also shown in Figure 4; it indicates that by this criterion the time-weighted average limit is more significant than the ceiling value only for distributions having GSD's below 2.3. For typical distributions the short-term limits for exposure are more restrictive.

From this analysis based on typical personal air sampling results it is apparent that the need to limit peak exposures is likely to be more restrictive than the need to limit the long-term average. It is difficult to assess the biological significance of controlling a chronic hazard in this way.

The Significance of Short-Term Peak Concentrations

The principal reason for limiting peak concentrations would appear to be to prevent the irritation of the upper respiratory tract and eyes, and to control the risk of acute effects of intoxication. In the Hygienic Guide it is stated that a concentration of 3000 ppm is irritant, but the author believes that 5000 ppm can be tolerated on a purely subjective basis.

The concentration that produces narcotic symptoms is less clearly defined. It is certainly dependent on the duration of exposure and is probably best expressed as an exposure dose—that is, the multiple of the concentration to which exposure occurs and the duration of the exposure. Review of published literature suggests that the danger limit for intoxication from short-term exposure is of the order of 100,000 ppm-minutes. It therefore appears desirable that for any exposure exceeding about 2 minutes and less than 8 hours the permissible concentration should be inversely proportional to the duration of exposure.

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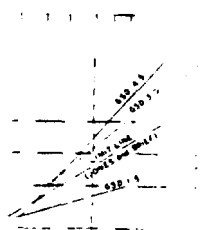


Figure 4. The relationship between the time-weighted average concentration (TWA) and the probability of exceeding a short-term peak concentration for different Geometric Standard Deviations (GSD). The curves show that for a given TWA, the probability of exceeding the tolerance increases as the GSD increases.

There remains the question of whether exposure to occasional short-term **peak** concentrations can cause chronic benzolism. This is suggested in the contribution made by Gerarde in Patty's *Industrial Hygiene and Toxicology*¹² which is not precisely quoted in the Hygienic **Guide**. Review of the literature does not reveal any specific evidence that chronic disease **has** been produced by short-term peak exposures in the absence of a significant time-weighted exposure. Where brief exposures to high concentrations have occurred, these are likely to have been repetitive and accompanied by more prolonged exposure to low levels.

Although a sudden death has been reported following an unknown short exposure¹³ (and the post mortem results suggest that a longer period of exposure must have occurred), the principal acute effect to be controlled is commonly taken to be narcosis. Recovery following a severe exposure may be complicated by later **sequelae**,¹⁴ though Elkins¹⁵ suggests that the risks of severe aftereffects following a single severe exposure are less than with most of the organic halogen compounds.

If it can be assumed that the chronic blood dyscrasias can be related to the amount of benzene taken up and retained in the bone marrow, it would appear unlikely that short-term exposure alone can make a significant contribution.

From breath sampling results following experimental and occupational exposure (to be reported), there appears to be a body compartment in which benzene is very slowly absorbed and excreted. There is reason to believe that this is fatty tissue and that it probably includes the bone marrow in which the blood cells develop. The excretion half-life **has** been found to be of the order of 24 hours, and it is likely that uptake is similarly slow. In dog experiments Schrenk et al.¹⁶ showed long-term retention of benzene during and following long exposure.

While benzene uptake by the blood is very rapid, and fairly rapid absorption occurs in muscular and well-perfused tissues, the uptake by bone marrow is probably very

slow and dependent on time-weighted average exposure.

A Single Criterion to Control Chronic and Acute Hazards

To meet varying environmental conditions it at first appears necessary to quote a range of appropriate acceptable concentrations for various exposure periods.

However, a single criterion for exposure dose can be derived from the present time weighted average limit, which would give adequate control of both chronic and acute hazards. Thus, an exposure-dose of 5,000 ppm-minutes corresponds closely to the 10 ppm time-weighted average limit for an 8-hour exposure, and for short exposure periods is well below the concentration known to produce narcotic effects.

For single daily exposures of this order for periods as short as 1 minute the limiting parameter may be irritation, but, in view of the difficulty of safely controlling so short a time limitation, exposure of men without respiratory protection could not be a considered good practice.

If benzene is indeed radiomimetic, then consideration might be given to controlling exposure over much longer periods. It is to be noted that radiation standards have changed from a ceiling value concept expressed in roentgens per minute in 1928 to exposures averaged over three months, one year, a generation, and even a lifetime.

It is pertinent to ask whether the fundamental standards for benzene exposure should be set on similar long-time periods, even if short-term criteria to control the risk of acute poisoning are also required. It is certainly economically desirable that some relaxation of present criteria be allowed for intermittent operations involving benzene.

The Application of the Proposed Criterion to Field Sampling

The use of the single exposure-dose criterion permits routine monitoring of individual men at risk by means of single samples collected over the work shift. For a given sampling rate the exposure-dose is

directly related to the amount of benzene collected by the sampler. If the concentration exceeds 5000 ppm-minute 1 exceeded, then the risk of disease is unlikely.

If short-term exposure (a probability of a single exposure exceeding 500 ppm-minutes) can be determined from Figure 1 of an extremely variable

is no more than 0.05 average is not to be exceeded corresponds to a six months; for more concentrations the probability is less than once per year.

A further advantage of the criterion is that, for a fixed amount of sample of given flow rate, it is possible to construct a simple warning if exceeded.

From a series of studies it appears that an exposure of 5000 ppm-minutes could produce a concentration of phenol in urine in excess of 200 mg/liter. This is below the 200 mg/liter proposed by the Committee¹⁷⁻¹⁹ for the control of benzene exposure but sufficient to show that exposure can be controlled by end-of-shift sampling for phenol.

Although end-of-shift sampling provides a very sensitive indication of quantitative exposure, it is assessed from samples collected during the shift.

that is, immediately after the shift. An exposure dose of 5000 ppm-minutes could be expected to result in 0.2 ppm in breath at the end of the shift.

The Significance of Single Samples: Biological Aspects

Roach²⁰ has suggested that the time might be set at a fixed half-life and, assuming a distribution of results, the

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directly related to the quantity of benzene
collected by the sampling device. If the
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ceeded, then the risk of chronic and acute
disease is unlikely in these conditions.

If short-term samples are taken, the
probability of a single 10-minute sample's
exceeding 500 ppm (exposure-dose = 5000
ppm-minutes) can be approximately de-
termined from Figure 4. Even in the case
of an extremely variable concentration this
is no more than 0.05% if the time-weighted
average is not to exceed 10 ppm. This
corresponds to a single sample every two
months; for more typically varying con-
centrations the probability corresponds to
less than once per year.

A further advantage of using this type
of criterion is that, as the limit corresponds
to a fixed amount of benzene trapped in a
sampler of given flow rate, it should be pos-
sible to construct simple monitors to pro-
vide warning if excessive exposure occurs.

From a series of studies of occupational
exposures for periods from 1/2 to 3 hours,
it appears that an exposure dose limit of
5000 ppm-minutes could be expected to
produce concentrations of conjugated
phenol in urine in the range of 20 to 60
mg/liter. This is below the value of 70 to
200 mg/liter proposed by various auth-
ors¹⁷⁻¹⁹ for the control of chronic hazard
but sufficient to show that risk of acute
exposure can probably be adequately moni-
tored by end-of-shift urine sampling for
phenol.

Although end-of shift breath samples pro-
vide a very sensitive indication of exposure,
quantitative exposure can really only be as-
sessed from samples taken 18 hours later
—that is, immediately before the following
shift. An exposure dose of 5000 ppm-min-
utes could be expected to produce about
0.2 ppm in breath at that time.

The Significance of Sampling Time

Biological Aspects

Roach²⁰ has suggested that the sampling
time might be set at one-tenth the biologi-
cal half-life and, assuming a nonnal distri-
bution of results, has shown that, if the

mean concentration is maintained at a level
1 standard deviation below the threshold
limit, the risk of the threshold body burden
being exceeded at any time would be less
than one in a million. This is perhaps an
overconservative approach. From Salz-
mann's work²¹ it can be deduced that fluc-
tuations in the quantity of the vapor
retained in the body are adequately rep-
resented if sampling time is one to two
times the biological half-life.

The biological half-life for benzene in
the brain is not well established, and it does
not appear possible to set an appropriate
sampling period to control the hazard of
acute poisoning. If, however, the suggestion
to establish a maximum exposure of 5000
ppm-minutes is accepted, then a sampling
system could readily be developed to give
warning when such an exposure may occur.

Observations of benzene elimination (to
be reported) show a half-life for excretion
of phenol sulfate in urine of about 7 hours,
which may be related to the concentration
of benzene in the liver. If this is a signif-
icant biological parameter, then sampling
periods as long as a whole shift will reveal
fluctuations adequately.

The significance of long-period retention
of benzene in fatty tissue to air sampling
procedure has already been discussed.

Statistical Aspects

Samples of air are taken to estimate the
concentrations of substances "sampled" by
workers; it is too commonly assumed that
they are identical. Personal sampling re-
duces errors due to variation of concentra-
tion with distance; statistical methods are
needed to determine the likely magnitude
of errors due to fluctuations in time.

While a single sample over a shift can
provide a measure of a man's exposure
during that shift, it provides no indication
of likely fluctuation. If conditions do not
change significantly between shifts, then
an estimate of the possible error in assessing
exposure can be made by observing fluctua-
tions during a shift. This is commonly done
by a simple rule based on the range of the
results observed which assumes their nor-
mal distribution, but it is better made by

calculation of standard error as described by Gale" and applied to radioactive particle sampling.

Unless preliminary results show that the level of exposure is likely to be very far away from the criterion, at least 25 samples will be required to obtain adequate statistical accuracy. If routine monitoring is found to be necessary, it is important that proper statistical control of the program be maintained.

Conclusions

The practical difficulties found in the application of a ceiling limitation to the threshold limit for a chronic poison leads to the proposal that threshold limit values for acute and chronic exposure be separately assessed. By introducing the concept of a permitted day's exposure based on a time-weighted average threshold limit of 10 ppm, it appears that both acute and chronic hazards can be controlled by a single limit of 5000 ppm-minutes.

Air samplers to provide warning of acute overexposure could readily be constructed, and long-period personal air samplers will provide adequate information on which to assess the chronic hazard.

As the introduction of this single criterion would greatly simplify monitoring exposure, it is hoped that toxicologists will consider its acceptability.

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Received June 25, 1971

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Introduction

DETECTOR TUBES for industrial hygiene simple measurements of certain gaseous pollutants in the air of a... the speed and ease with which measurements are made with undeniable advantage such measurements have

With the objectives of providing aid to hygienists about accuracy and precision by encouraging tube manufacturers to improve the performance of detectors, the Bureau of Occupational Safety and Health of the U.S. Public Health Service is conducting a detector tube project. The project is a pilot program and is not a certification program.

The Perchloroethylene Vapor System

Perchloroethylene vapors were generated dynamically by the pressure method (Figure 1), the carrier gas, was water, double-effect evaporation of perchloroethylene liquid