

BIOASSAY CORRELATION WITH BREATHING ZONE SAMPLING

Roger Caldwell
Thomas Potter
Edward Schnell

Health and Safety Department
Nuclear Materials and Equipment Corporation
Apollo, Pennsylvania

Prepared for Presentation at the 13th Annual
AEC Bioassay and Analytical Chemistry Conference

1967

Bioassay Correlation with Breathing Zone Sampling

Roger Caldwell, Thomas Potter and Edward Schnell

Nuclear Materials and Equipment Corporation
Apollo, Pennsylvania

The Purpose of Air Sampling

Because airborne radioactivity is the chief hazard in a nuclear fuel plant, the health physicist expends much effort in air sampling. He expects his air sampling program to accomplish several ends, ^(1,2,3) such as:

- 1) warning when high air levels exist,
- 2) measuring the effectiveness of control measures,
- 3) determining general room air levels or
- 4) following contamination trends.

But the primary purpose of air sampling is:

- 5) the determination of personal exposure.

Indeed, since federal regulations ⁽⁴⁾ are written in terms of individual exposure, the law compels the health physicist to estimate the radioactivity which each radiation worker inhales.

The problem is interpreting air samples. It is very easy to take an air sample, but it is often very difficult to know what the results mean. A few years ago, Harry Schulte ⁽⁵⁾ of Los Alamos made the point that "air sampling is not a science but an empiric art."

There are two basic kinds of air sampling: fixed station, commonly called general area (GA) sampling, and breathing zone (BZ) sampling. Industrial hygienists have been taking BZ samples for decades by holding sample heads close to the worker's nose. ⁽⁶⁾ Recently, battery powered lapel samplers have been developed which are worn by the worker. ⁽⁷⁾

Most nuclear fuel facilities we know about either assign fixed station concentrations to workers or depend on bioassay to estimate exposure. This paper will show that personal breathing zone air samplers not only yield exposure data which correlates with bioassay results, but most often are the only accurate means of measuring individual exposure.

The Nature of Industrial Radioaerosol Exposure

Some vague ideas about the nature of industrial airborne activity have led to elaborate fixed station air monitoring systems.⁽⁸⁾ One common notion is that air activity takes the form of a rather large cloud which disperses throughout a room until it settles out on horizontal surfaces. Another concept is that, except in accident conditions, air activity consists of isolated particles randomly distributed in the room air.

Our experience presents a completely different picture. We believe almost all industrial radioaerosol exposures are extremely localized in space. An example of what we mean is shown in figure 1. Here an analytical chemist, moving a contaminated beaker from one hood to another, is exposed to a small local cloud. He is wearing a lapel sampler which should detect the release. Nearby a fixed station air sampler is operating. This sampler, if it detects the release at all, will surely underestimate the exposure. We have also found the mere withdrawal of contaminated gloved hands from a hood can expose workers.

Plutonium glove box releases especially follow steep concentration gradients. Figure 2 shows a typical example. The operator is coming out of the gloves to check his hands on the alpha meter. He will find them contaminated because a hole developed in the left box glove.

As soon as he is aware of the contamination, he will put on a respirator, cover the glove port, survey the area and change the glove. But he will have

already been exposed by the small cloud generated when the glove was inverted. We have found that fixed station samplers, like the one in the background, rarely detect these local releases. Also many times the releases are not accompanied by any appreciable floor fall out contamination. This type of release is common in plutonium facilities. One major plant⁽⁹⁾ reports 900 glove failures per month.

Another common source of exposure is contaminated protective clothing. A worker wearing contaminated clothing generates a cloud or radioaerosol around himself. Figure 3 shows how the health physicist views the typical radiation worker. NUMEC experiments⁽¹⁰⁾ suggest that the function and design of protective clothing needs reevaluation.

Although these photographs were staged with MSA smoke tubes, NUMEC experience with lapel samplers strongly suggests that uranium and plutonium aerosol clouds, although invisible, take exactly the same shape. The typical airborne release is a small cloud which quickly disperses to unmeasurable concentrations with relatively little surface contamination. In our experience floor contamination does not necessarily mean you have an air inhalation problem, but it surely means you had one earlier.

NUMEC Experience with Breathing Zone Sampling

NUMEC uranium and plutonium workers have worn lapel samplers for two years. We find these samplers usually indicate higher concentrations than stationary samplers. Often the difference is orders of magnitude.

Figure 4 gives a two year comparison of lapel samplers with fixed station air samplers. It shows the lapel to fixed station ratio distribution for 594 BZ samples at our plutonium laboratory and 459 at our uranium plant. The sample durations were for single shifts, an eight hour workday. The fixed station concentration is either the average of those in the worker's

area or the one closest to his work station. Actually we found little difference between fixed station "breathing zone" samplers and those intended to cover general areas. The interval of general BZ - GA agreement (+ 100%, - 50%) covered 27% of the plutonium BZ samples and about 19% of uranium plant BZ samples. Notice that almost 9% of Pu BZs are less than 50% lower than the GAs. Sixty-four per cent of Pu BZ's exceeded the GA concentration by a factor of 2 or more, 23% by more than a factor of ten. The highest ratio we've ever detected was 9,870. Thirty-five per cent of uranium plant BZ concentrations exceeded 10 times the fixed station concentrations. While the median of these ratios is less than 10 for both plants, the very skewed distribution makes high level exposures very important in computing the average exposures.

Figure 5 presents a clearer idea of how important personal samplers are when high level exposures occur. The BZ/GA ratio data for all plutonium exposures exceeding 10 MPC for an eight hour shift is plotted against the breathing zone concentration.

The first impression from this graph is the extreme variability of the BZ/GA ratio for a given BZ concentration. As an example, for those BZs between 40 - 50 d/m/m³ the fixed station concentration varied from one half to one eight hundredth of the BZ concentration. It is difficult from this data to pick out a suitable factor (such as the UKAEA^(3,11,12) has done) by which to multiply the GA concentrations to obtain individual exposure.

Another thing to notice is the upward trend in the BZ/GA ratio as the BZ concentration increases. Basically this means the worse the problem is the wronger the fixed station data.

We have drawn in the line where the fixed station air sample would indicate the soluble MPC_a for Plutonium. For all data above the line the GA was less than MPC. Only those GA's below the line even indicated that a

hazard existed. This is an important point. Many industrial radioaerosol exposures are going unnoticed because the nuclear industry is depending on fixed station air sampling.

The inability of stationary air samplers to indicate hazardous conditions in a uranium plant is shown on Figure 6. Nearly 73% of the time the GA sampling network failed to warn when greater than permissible exposure was occurring. Lest NUMEC be accused of not knowing how to place fixed air sample heads, please remember the location of the fixed air samplers in Figures 1 and 2. It has been our practice to place these sampling heads as close to the breathing zone in high risk areas as is possible. There are 55 such samplers in our 20,000 ft.² plutonium laboratory and 34 in our 40,000 ft.² uranium plant.

It might be thought from reviewing this data that radiation control practices at NUMEC plants are not up to industry standards. We honestly don't think this is the case. In the first place we didn't have enough lapel samplers to continuously sample the breathing zone of all our workers. Consequently, we have chosen to use our available lapel samplers as diagnostic tools in areas where we feel that local "micro-climates" of radioaerosol may exist. Thus the high percentage of BZ samples above MPC_a is misleading. Our feeling is that it is the industry air sampling standards that are not adequate.

The Correspondence of Breathing Zone Sampling with Early Fecal Clearance

The lapel sampler data would not be relevant, if it did not represent true exposure. For this reason, whenever an exposure occurred, the operator was removed from radiation work and both fecal and urine samples were collected. Figure 7 gives the correlation between BZ sampling and early fecal clearance for plutonium exposures. The eight cases shown were selected from almost a

hundred exposures because total fecal and urine data was available for the first seven days post exposure and because there was no recent prior exposure to complicate interpretation. Early fecal clearance was chosen as the exposure criteria because of earlier experience with urine and fecal sampling at our Plutonium laboratory. (13)

There is remarkable agreement between the proposed ICRP lung model (14) and the lapel sampler data. The line represents expected 72 hour lung clearance for insoluble one micron MAD PuO_2 particles.

The failure of fixed station samplers could not be more graphic. We might also add that urine sampling did not demonstrate these exposures. Except for the highest plutonium exposures, no perturbation in urine excretion could be detected.

An example of inhalation inventory balancing is shown in Figure 8. A single UO_2 exposure was detected. We were fortunate enough to be able to follow this exposure unperturbed by subsequent exposures. Several items are interesting.

First, the activity inhaled as estimated by GA samples is less than the first day's fecal elimination. Secondly, the balance between the BZ estimate and that excreted over 50 days is noteworthy. Even the overage on excretion is consistent since the individual had a history of recent exposure.

The early clearance half time of 1.5 days is interesting in that it agrees well with the lung model. It means that individuals must be removed from any possible exposure for at least seven days before fecal data can be used to estimate long time lung burdens.

Fecal Sampling In a Uranium Plant

Figure 9 shows the importance of routine fecal sampling in a uranium plant. The excretion rates via feces and urine are plotted against each

other for all cases where the samples were collected the same day. The traditional permissible urine level is 75 d/m/day. The permissible fecal excretion rate was calculated to be 50 d/m/day assuming the ICRP recommended 380 day half time for chronic UO_2 exposures.

Many of the fecal data represent early clearance and are not necessarily unpermissible. However it is obvious that urine data by itself gives a false impression of actual exposure. We were able to use this data to estimate the effectiveness of half face respirators. We found that they are not very effective unless their use is closely supervised.

The Necessity for Breathing Zone Sampling

Many radiation protection workers feel that breathing zone sampling is too fussy or is impossible. We believe it is absolutely necessary and can be easily done successfully with personal air samplers. Figure 10 demonstrates our concept. Nearly all radioaerosol sources are small; generally the worker's hands are the major aerosol generator. In static room air conditions concentrations will fall off with the inverse cube of the distance. If the distance from the hands is doubled, the concentration will be lower by a factor of eight. We have verified this by experiment.

The usual turbulent condition is more complicated. Still, the concentration gradient will be steep and any fixed station air sampler a few feet away will underestimate the man's exposure.

Breathing zone sampling is usually recommended on the vague intuition that the closer the sampler is to the nose the better. NUMEC and UKAEA^(1,3,11,12,15) experience provides a more convincing basis for personal air sampling. The worker handling radioactive materials lives in a "micro-climate" which must be sampled if the health physicist is to detect industrial radioaerosol exposure.

BIBLIOGRAPHY

1. B.A.J. Lister, Health Physics Aspects of Plutonium Handling, AERE-L151, 1964.
2. J. Pomarola, et.al., Assessment of Individual Risk During Airborne Contamination by Plutonium, IRPA Congress, Rome, 1966.
3. W.A.Langmead and D.T. O'Connor, The Significance of Radioactive Aerosol Measurements made in the Working Environment, IRPA Congress, Rome, 1966.
4. Code of Federal Regulations, Title 10, Part 20.103.
5. H.F. Schulte, The Contribution of Industrial Hygiene to the Protection of the Radiation Worker, 1966.
6. E.C. Hyatt and H.F. Schulte, Air Sampling Procedures in Evaluating Exposures to Uranium, HASL-58, 1958.
7. R.J. Sherwood and D.M.S. Greenbough, A Personal Air Sampler, Annual Occupational Hygiene, 2:127, 1960.
8. M. Sanders, Innovations in Air Monitoring Techniques for Large Scale Programs, Y-KB-78, 1966.
9. E.A. Putzier, Plutonium Hazards and Accident Experiences, RFP-621, 1965.
10. E. Schnell and R. Crosby, Breathing Zone Air Levels as Caused by Contaminated Clothing, unpublished NUMEC report, 1966.
11. R.J. Sherwood, On the Interpretation of Air Sampling for Radioactive Particles, American Industrial Hygiene, 27:2, pp 98-109, 1966.
12. R.T. Brunskill and S.T. Hermiston, The Detection and Measurement of Plutonium Airborne Contamination in Major Plutonium Facilities, IRPA Congress, Rome, 1966.
13. R. Caldwell, The Detection of Insoluble Alpha Emitters in the Lung, AEC Conference on Bioassay and Analytical Chemistry, Gatlinsburg, Tenn., 1966,
14. Task Group on Lung Dynamics, Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract, Health Physics, 12:173, 1966.
15. D.C. Fraser, Health Physics Problems Associated with the Production of Experimental Reactor Fuels Containing PuO₂, Health Physics 13:1133, 1967.



Fig 1

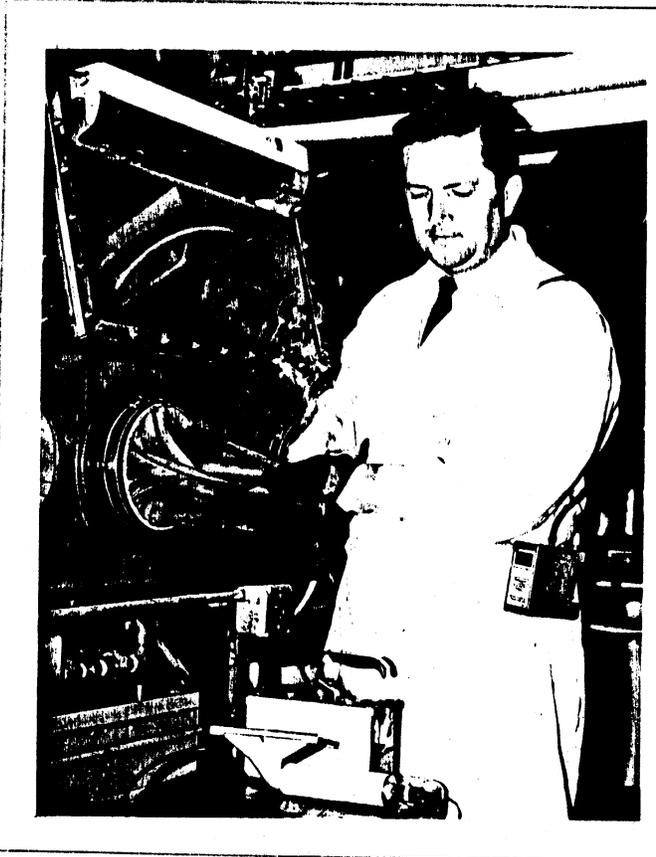


Fig 2

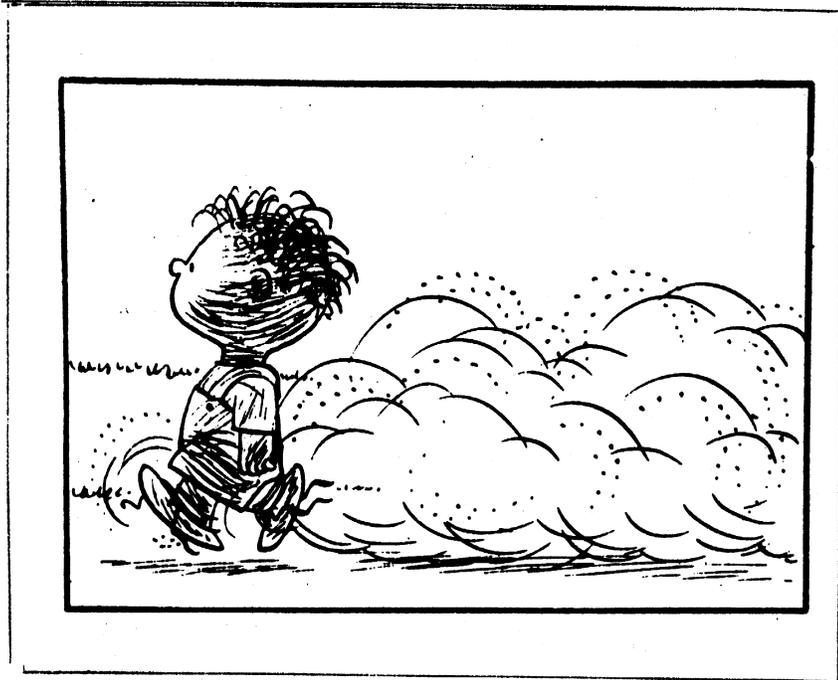


Fig 3

COMPARISON OF LAPEL (BZ) TO FIXED
 STATION (GA) AIR SAMPLING
 NUMEC NUCLEAR FUEL FACILITIES
 1966-1967

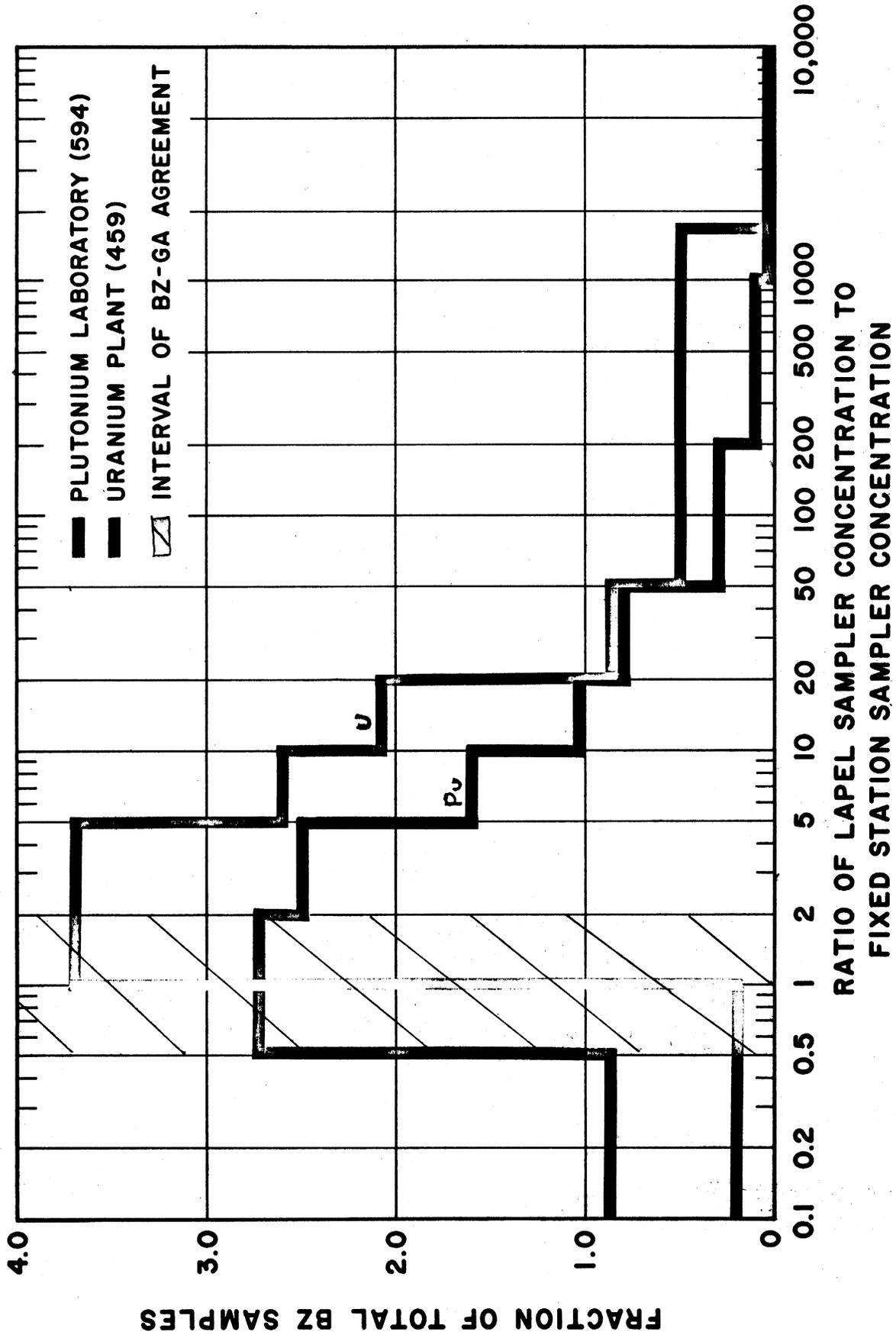


Figure 4

COMPARISON OF LAPEL (BZ) TO FIXED STATION (GA) AIR SAMPLING

NUMEC PLUTONIUM LABORATORY
1966-1967

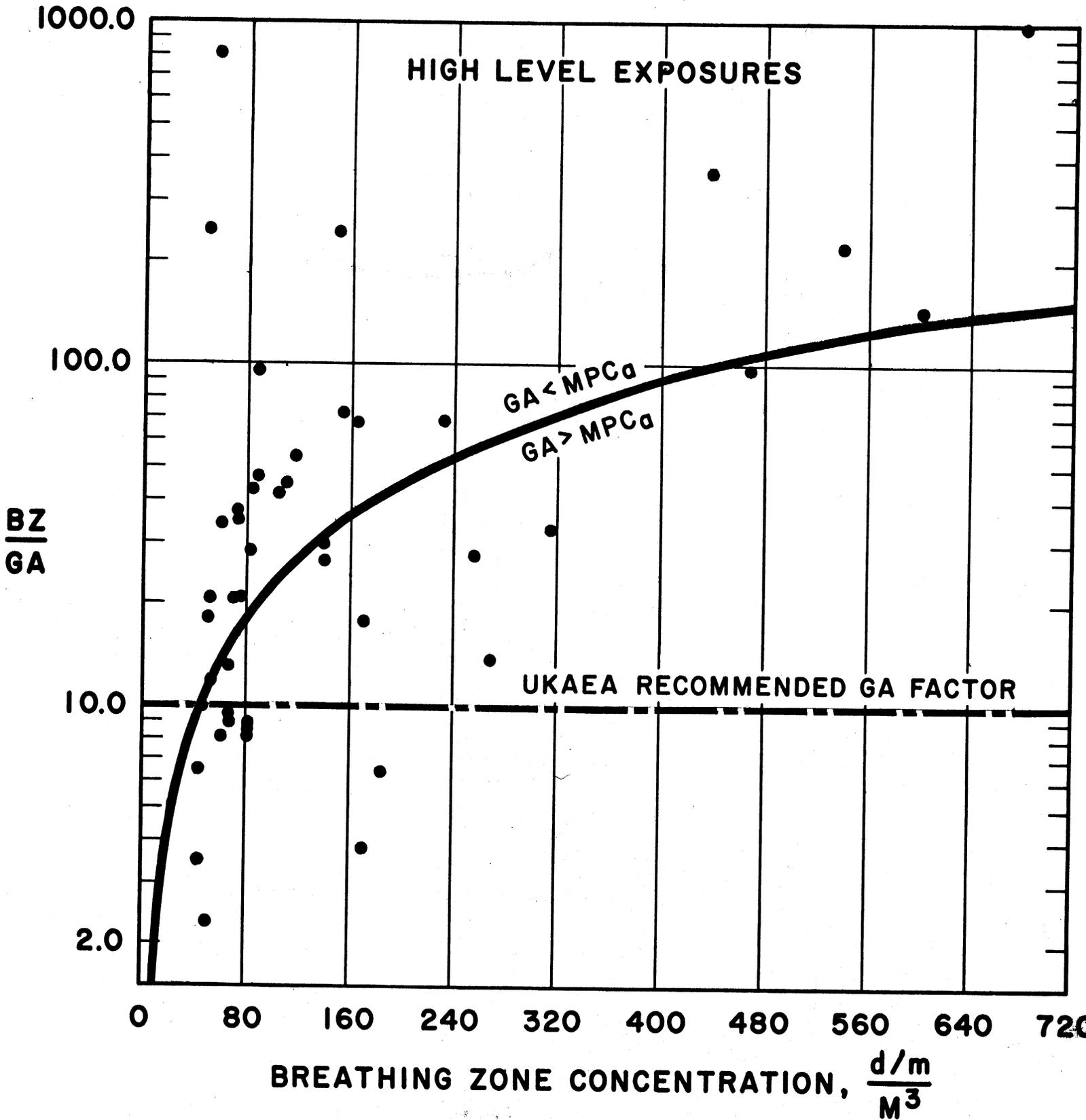


Figure 5

HAZARD INDICABILITY

Lapel Samples (BZ) vs. Fixed Station Samples (GA)

NUMEC URANIUM PLANT

1966 - 67

CONDITION INDICATED	NUMBER RECORDED	FREQUENCY
BZ > MPC GA < MPC	300	.654
BZ > 10 MPC GA < MPC	33	.072
BZ < MPC GA < MPC	54	.118
BZ < MPC GA > MPC	2	.004
BZ > MPC GA > MPC	70	.152

Total BZ Samples 459
 Total GA > MPC 72
 Total BZ > MPC 403

Figure 6

CORRELATION OF FECAL BIOASSAY
WITH AIR SAMPLING
NUMEC PLUTONIUM LABORATORY
1966-1967

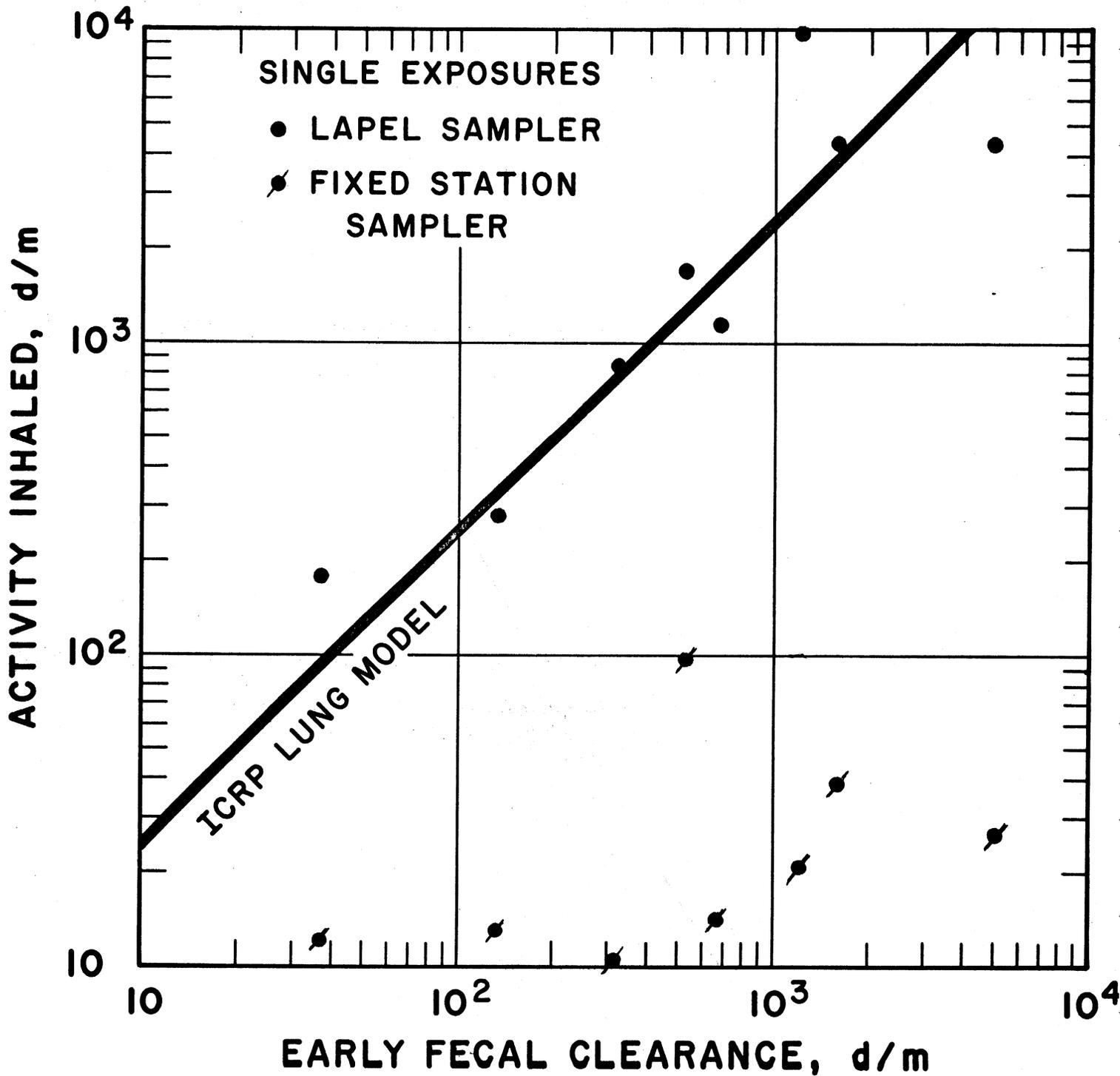


Figure 7

URINE vs FECAL EXCRETION RATES
CHRONIC ^{234}U OXIDE EXPOSURES
NUMEC URANIUM PLANT

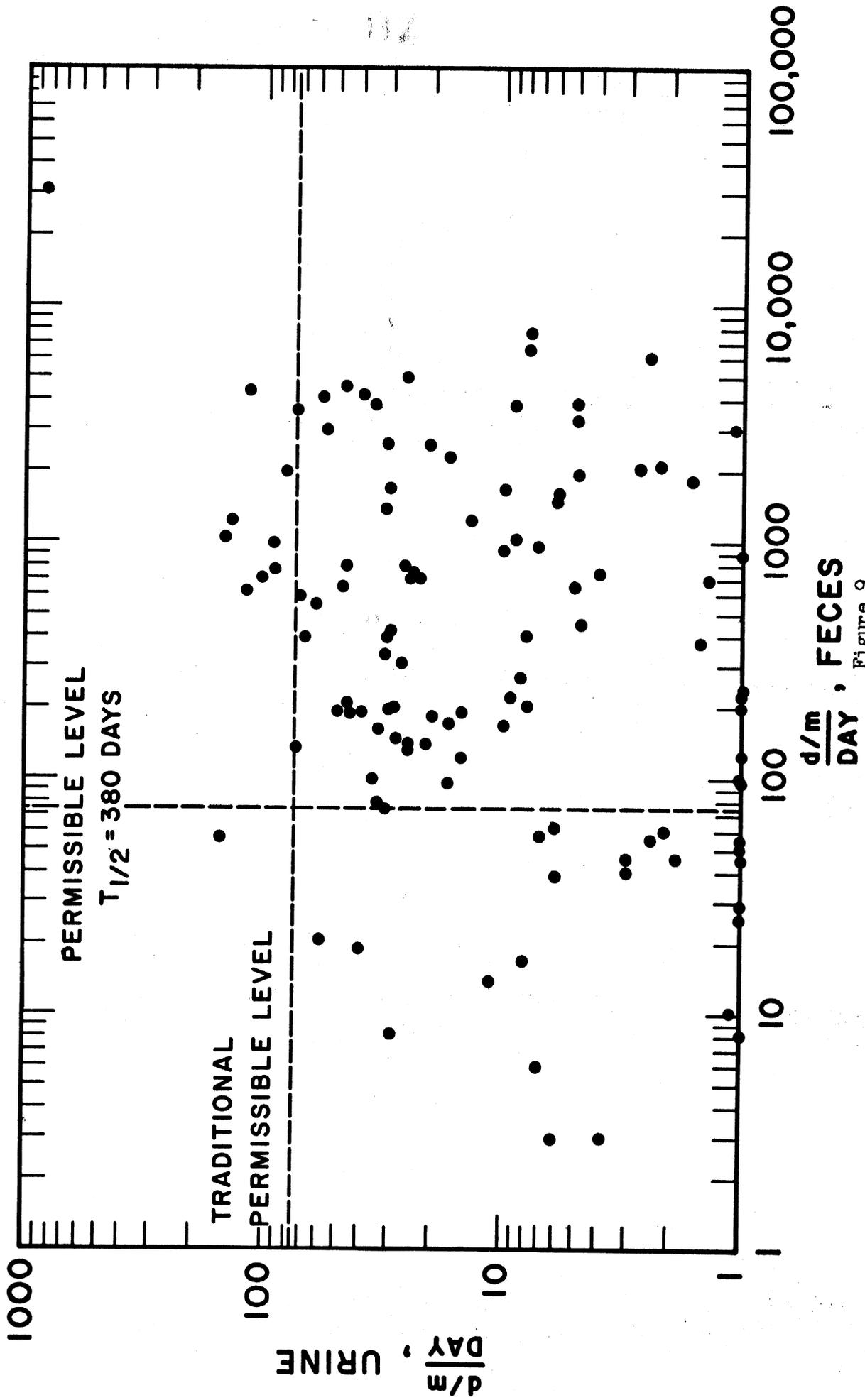


Figure 9

**UO₂ EXPOSURE AND BIOASSAY DATA
(W.S.) ACUTE EXPOSURE - May 22-23, 1967
NUMEC URANIUM PLANT**

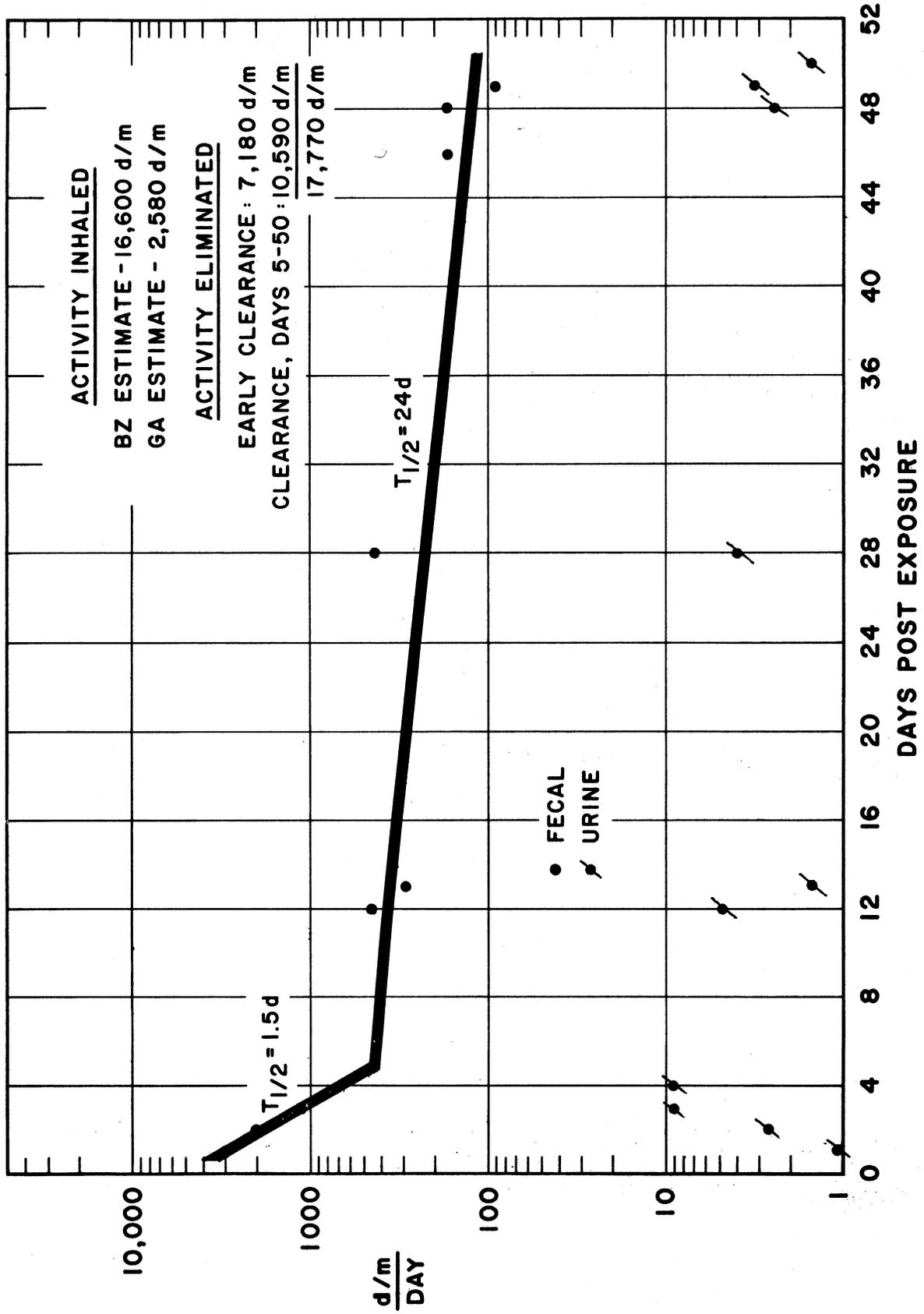
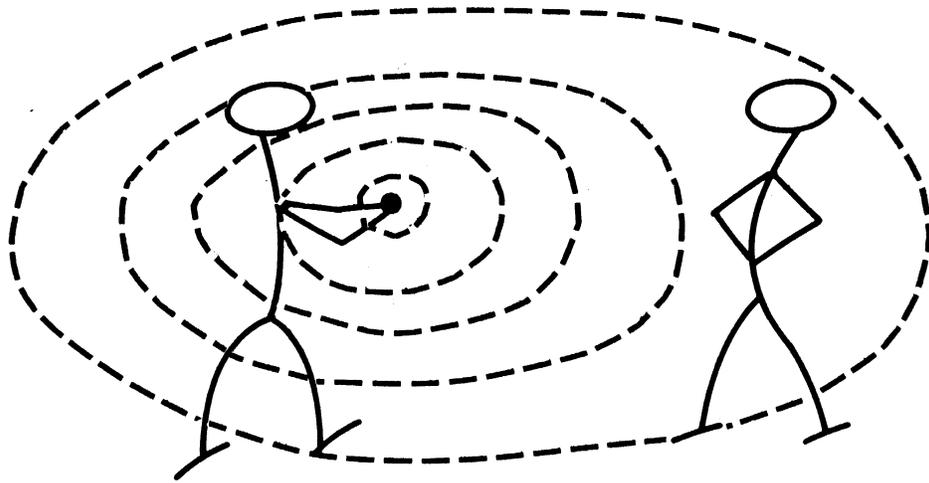


Figure 8

NECESSITY FOR BZ SAMPLING

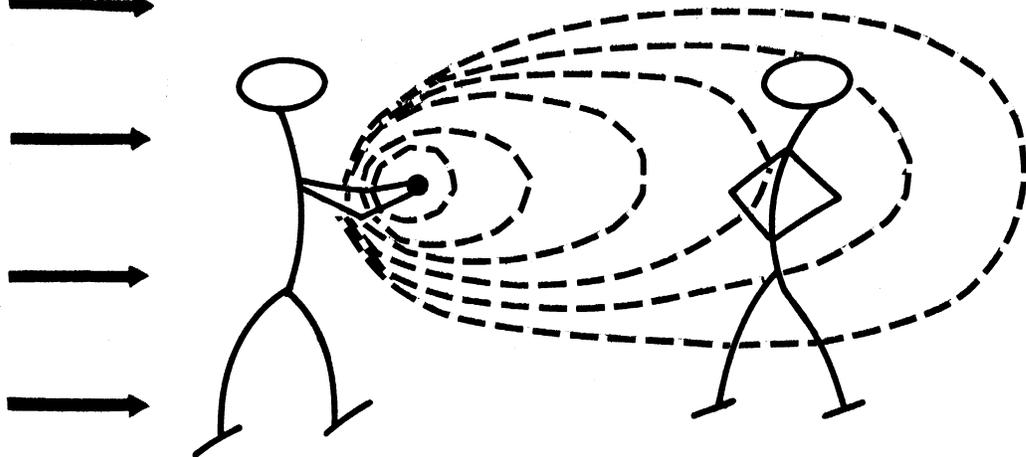
STATIC



$$C \propto \frac{1}{R^3}$$

TURBULENT

DRAFT →



$$C \propto \frac{1}{x^2} e^{-\frac{k}{x^2}}$$

Figure 10