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OPERATIONAL EVALUATION OF THE ELECTRET ION CHAMBER (EIC) METHOD FOR DETERMINING RADON-IN-WATER CONCENTRATIONS

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ABSTRACT

A recent trend in radon measurement has been the adaptation of traditional radon in air measurement technology for performing radon-in-water measurements. The U.S. EPA Office of Radiation and Indoor Air-Las Vegas (ORIA-LV) is the principle laboratory responsible for evaluating methods of radon measurement involving both new and existing technologies. The electret-ion-chamber (EIC) method is an approved technology for the measurement of radon in air and has been used successfully in a variety of indoor radon measurement applications. A new EIC application for the measurement of radon-in-water has been developed by the manufacturer and subjected to a comprehensive operational evaluation at the ORIA-LV Radon Laboratory. The measurement technique was subjected to radon exposures ranging from appoximately 500 to 60,000 pCi/L and evaluated for precision, bias, and practicality for performing field measurements. The results of the evaluation, while promising, indicate that additional development of the technique may be warranted.

INTRODUCTION

Between July and November 1992 ORIA-LV conducted an operational evaluation of the EIC method for determining radon-in-water concentrations as developed by Rad Elec, Inc. (REI) of Frederick, Maryland. In order to determine the ability of the method to measure radon over a wide range of concentrations, the evaluation was performed in two phases and involved the analysis of approximately 160 water samples for their content of ^{222}Rn. The ^{222}Rn target values for phase-one ranged from 777 to 19,977 pCi/L while target values for phase-two began at a substantially higher concentration of 63,400 pCi/L and ranged down to 478 pCi/L. Water samples used in the evaluation were collected by REI from the water-supply well of a municipal office building in Reading, PA known to contain a highly elevated concentration of ^{222}Rn and subsequently provided to ORIA-LV for analysis. Radon target values for the water samples were obtained by ORIA-LV using liquid scintillation (LS) analysis.

METHODOLOGY

The first phase of the evaluation was divided into four exposure sets referred to as 1A, 2A, 3A, and 4A. Each exposure set consisted of 25 individual EIC measurements of radon laden water along with 10 control EIC exposures using distilled water that was void of any measurable quantity of radon. In order to examine the possible influence of background radiation and EIC stability as well as any potential interference from humidity or other external factors, the controls were subdivided into two groups consisting of five closed EIC's and five open EIC's.

Exposure set 1A was exposed to water samples with a measured target value of 19,977 pCi/L of ^{222}Rn while exposure sets 2A, 3A, and 4A were exposed to 7,935, 2,625 and 777 pCi/L respectively. All phase-one exposures including the controls were made using a sample volume of approximately 67ml of water. The water samples were collected by REI in small 67ml leak tight glass bottles with teflon lined caps over a period of approximately one hour, using a protocol published in *Two Test Procedures for Rn in Drinking Water* (EPA Report number 600/2-87/082:1987), and numbered sequentially. To reduce any variability in dissolved radon concentrations that may have been introduced during the lengthy period of collection, sequential runs of samples collected over a relatively short period of time were used for each exposure set.

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In order to expand the operational range over which the method was evaluated, a second exposure phase was conducted with the initial exposure set beginning at 63,400 pCi/L of ^{222}Rn. Since phase-two of the evaluation was initiated with a substantially higher concentration of radon it was divided into six separate exposure sets referred to hereafter as exposure sets 1B through 6B. Due to the increased number of exposure sets being performed during phase-two, each exposure set was reduced to ten individual EIC water sample measurements along with six control exposures (three opened and three closed EICs). The phase-two exposure sets 1B through 6B were exposed to water samples containing 63,400, 22,500, 7,247, 1,984, 1,416 and 478 pCi/L of ^{222}Rn respectively. Exposure sets 1B through 5B were made using 67ml water samples. Per the REI protocol for performing measurements of low concentrations of radon, exposure set 6B was made using larger 134ml water samples along with an increased exposure time. Exposure parameters for set 6B control samples were adjusted accordingly.

As specified by REI, all measurements employed the use of an E-Perm System S-chamber in conjunction with either a long or short-term EIC measurement device. For each exposure set, the selection of long or short-term EIC's, duration of exposure, and sample volume size (67ml or 134ml) were made in accordance with the radon-in-water measurement protocol published in part II of REI's *E-PERM System Manual.* EIC selection, exposure times, sample sizes, and target values for phase-one and phase-two exposure sets are presented in tables 1 and 2 respectively. In addition, all exposures were conducted in a climate controlled laboratory setting with a mean ambient air temperature of approximately 23°C and a background atmospheric radon concentration of less than 0.5 pCi/L.

The EIC exposure protocol used for performing radon-in-water measurements was taken directly from the REI *E-PERM System Manual.* The following is a brief synopsis of the procedure: A water sample is collected in a small glass sample bottle and placed in the bottom of a 3.8 liter glass wide-mouth analysis jar lying on its side. The sample bottle is held in place by a metal clip fixed to the bottom of the analysis jar. The pre-exposure voltage of the EIC being used for the analysis is measured. The EIC (s-chamber) is then opened to the "on" position and attached to a plastic clip on the lid of the analysis jar in such a manner that it will be suspended in the air-phase of the analysis jar throughout the period of exposure. The water sample bottle is opened; the analysis jar lid is screwed on and the entire assembly is turned upright thereby distributing the water sample over the bottom of the analysis jar. A rubber collar in conjunction with a large metal hose clamp is used to form an air tight seal between the jar and the lid. With gentle agitation of the analysis jar the radon quickly reaches an equilibrium between the water and air phases. After the specified exposure period, the EIC is removed and a post-exposure voltage is measured. Figure 1 illustrates the measurement set-up as described above.

Using the EIC voltage drop that occurred during the analysis exposure, duration of the exposure, appropriate decay factors, and constants (eg. volume of water sample and analysis jar) a radon concentration for the air-phase of the analysis jar can be calculated and then converted into a dissolved radon concentration for the water sample at the time of collection. For those interested, a detailed discussion of this calculation can be found in *Electret Ion Chamber Radon Monitors Measure Dissolved ^{222}Rn In Water* (Kotrappa and Jester: Health Phys. 64:397-405; 1993).

Target values were obtained by removing three representative samples of water (generally the first, middle, and last samples) from each sequential run being used for a particular exposure set and subjecting them to liquid scintillation (LS) analysis at the ORIA-LV Radon Evaluation Laboratory using EPA method 913.0 (draft). For example, if the sequential run used for an exposure set consisted of water samples 1 through 28, then samples number 1, 14, and 28 were removed for LS analysis and their results averaged to obtain a representative target value for the exposure set. In addition, for several of the samples, target values were independently confirmed through split sample analyses with the EPA Environmental Monitoring Systems Laboratory-Radiation Quality Assurance Group at Las Vegas. Results for all samples analyzed by the two labs were within ±5% of each other.

RESULTS AND DISCUSSION

Results for both phase-one and phase-two exposure sets are summarized in tables 3 and 4 respectively. Included in the summary tables are the mean radon-in-water concentrations obtained via EIC measurement along with their associated precision and bias for each exposure set.

The precision for the phase-one exposure sets ranged from one sigma values of ±3.5% (set 3A) to ±15.2% (set 2A). The precision for three out of the five exposures sets were within ±5%. However, exposure set 2A with a precision of ±15.2% was influenced by a single unexplained high measurement of nearly twice the mean measurement value. Omittance of this high measurement value would result in a one sigma precision of ±6.2% for exposure set 2A and is more representative of the overall consistency of the individual measurement values for the set.

The precision for the phase-two exposure sets ranged from one sigma values of ±2.0% (set 1B) to ±11.6% (set 2B). The precision for five out of the six exposure sets were within ±5%. As in the case of exposure set 2A, the precision of exposure set 2B was influenced by a single high measurement.

The mean percent bias for phase-one exposure sets ranged from -26.3% to -31.3% while phase-two exposure sets ranged from -22.8% to -27.6%. This bias is clearly illustrated in figures 2 and 3 which show the mean EIC value in pCi/L plotted against the target value for each exposure set. Although the biases for all phase-one and phase-two exposure sets were significantly below their respective target values, they displayed a remarkable consistency. A regression analysis using the least squares method was applied to the combined results of the phase-one and phase-two exposure sets and is presented in figure 4. Although a best fit regression line was calculated and plotted, an analysis of variance showed no significant variance between EIC bias and the concentration of radon being measured. Therefore the mean percent difference of approximately negative 26% may be applied uniformly to all of the EIC exposures performed.

The combined phase-one and phase-two controls produced a mean voltage drop of 0.6 volts for the closed EIC exposures while the open EIC exposures produced a mean voltage drop of 1.5 volts. This slight increase in voltage drop for the open controls was not considered excessive and may have been the result of environmental conditions such as the high humidity within the analysis jar. The mean voltage drops for both types of controls (open/closed) were considered insignificant when compared to the much larger voltage drop of the actual EIC measurements.

In order to examine possible inter-laboratory variability, simultaneous analyses of identical water samples were performed by REI in Frederick, MD on four separate occasions during phase-two of the evaluation using the REI EIC method of analysis. The water samples analyzed by REI were performed in duplicate and corresponded to the same exposure times and durations as evaluation sets 1B, 3B, 4B, and 5B. These samples were all from the same water collection batch provided to ORIA-LV for phase-two of the evaluation. The results of these simultaneous analyses are presented in table 5. While both the ORIA-LV and REI results showed a consistent but negative bias, the REI bias on average was half the bias obtained by ORIA-LV. The difference between the ORIA-LV and REI results may be due to factors such as the difference in elevation between the two analysis sites or possible leakage of radon during transportation of the water samples to ORIA-LV. At this time it is not possible to assign a definitive cause or causes to this potentially serious discrepancy without conducting additional carefully designed exposures.

With regard to the practicality of this method for performing field measurements, no significant difficulties were encountered performing the steps required for the set-up and analysis of water samples. However, the authors believe that the exposure set-up is slightly cumbersome requiring a certain amount of speed and manual dexterity on the part of the user. Additionally, particular care must be exercised when tightening the sealing collar on the analysis jar as not to severely puncture one's hand with the screwdriver. With some slight design modifications, for example a sealing collar that does not require the use of a screwdriver, and a moderate amount of practice by the user most of these limitations can easily be overcome. Finally, as with any method requiring collection of water samples for analysis of radon content, strict adherence to the sample collection protocol must be maintained in order to ensure the validity of the sample.

SUMMARY

The primary objective of this evaluation was to assess the ability of the EIC method to accurately measure radon-in-water over a wide range of concentrations. In order to determine the accuracy of the method, both precision and bias were evaluated. The secondary objective was to evaluate the practicality of the method for performing measurements in the field.

The evaluation established that the EIC method demonstrates good precision when measuring radon in water over a wide range of concentrations. In addition, it was shown that with a moderate amount of practice the method should be fairly easy for most technicians to perform. However, with regard to bias, additional investigation is needed to resolve the following problems: (1) the cause of the large but consistent negative bias, and (2) the significant discrepancy in bias observed between analyses performed at ORIA-LV and REI. Until these issues concerning bias are adequately addressed and the method re-evaluated, the accuracy of any measurements made in the field are suspect.

SELECTED REFERENCES

E-PERM® system manual. Rad Elec Inc., Jan. 1, 1991.

Kotrappa, P.; Jester, W. A. Electret ion chamber radon monitors measure dissolved ^{222}Rn in water. Health Phys. 64:397-405; 1993.

U.S. Environmental Protection Agency, Environmental Monitoring Systems Laboratory-Las Vegas. Determination of radon in drinking water by liquid scintillation counting. Method 913.0 (Draft). May 1991.

U.S. Environmental Protection Agency. Two test procedures for Rn in drinking water. Inter-laboratory study. EPA/600/2-87/082: 1987.

DISCLAIMER

Mention of trade names or commercial products in this document does not constitute EPA endorsement or recommendation for their use.

TABLE 1. Phase-One Measurement Configurations

Exposure Set	Target Value (pCi/L)	EIC	Sample Size (ml)	Exposure (Hours)
1A	19,977	long-term	67	26
2A	7,935	long-term	67	26
3A	2,625	short-term	67	26
[a] 4A (part-1)	777	short-term	67	53
[a] 4A (part-2)	777	short-term	67	73

[a] Exposure set 4A was subdivided into a 53 and 73 hour exposure. Both part-1 and part-2 exposures were started simultaneously but part-2 was allowed to accumulate an additional 20 hours of exposure. The exposure set was divided approximately equal: 12 measurements with 4 controls (2 open and 2 closed) in part-1; 12 measurements with 6 controls (3 open and 3 closed) in part-2.

TABLE 2. Phase-Two Measurement Configurations

Exposure Set	Target Value (pCi/L)	EIC	Sample Size (ml)	Exposure (Hours)
1B	63,400	long-term	67	24
2B	22,500	long-term	67	24
3B	7,247	short-term	67	24
4B	1,984	short-term	67	24
5B	1,416	short-term	67	24
6B	478	short-term	134	48

TABLE 3. Summary of Phase-One Results

Exposure Set	Number of Replicates	Avg Radon Conc EIC (pCi/L)	Target Value (pCi/L)	EIC Precision (1 sigma)	EIC Bias (%)
1A	25	13,716	19,977	±4.6%	-31.3
2A	25	5,760	7,935	±15.2	-27.4
3A	24	1,935	2,625	±3.5%	-26.3
4A-1	12	567	777	±10.4%	-27.0
4A-2	12	573	777	±4.5%	-26.3

TABLE 4. Summary of Phase-Two Results

Exposure Set	Number of Replicates	Avg Radon Conc EIC (pCi/L)	Target Value (pCi/L)	EIC Precision (1 sigma)	EIC Bias (%)
1B	10	45,904	63,400	±2.0%	-27.6
2B	10	16,669	22,500	±11.6%	-25.9
3B	9	5,593	7,247	±3.5%	-22.8
4B	10	1,510	1,984	±4.7%	-23.9
5B	10	1,054	1,416	±4.3%	-25.6
6B	9	355	478	±3.2%	-25.7

TABLE 5. Comparison Of Simultaneous ORIA-LV/REI Radon Analyses

Exposure Set	Target Value (pCi/L)	LVF EIC Value (pCi/L)	LVF EIC Bias (%)	REI EIC Value (pCi/L)	REI EIC Bias (%)
1B	63,400	45,904	-27.6	56,850	-10.3
3B	7,247	5,593	-22.8	6,388	-11.9
4B	1,984	1,510	-23.9	1,713	-13.7
5B	1,416	1,054	-25.6	1,212	-14.4

FIGURE 1.
E-PERM® SYSTEM RADON-IN-WATER MEASUREMENT
(Modified from P. Kotrappa & W.A. Jester, written commun.)

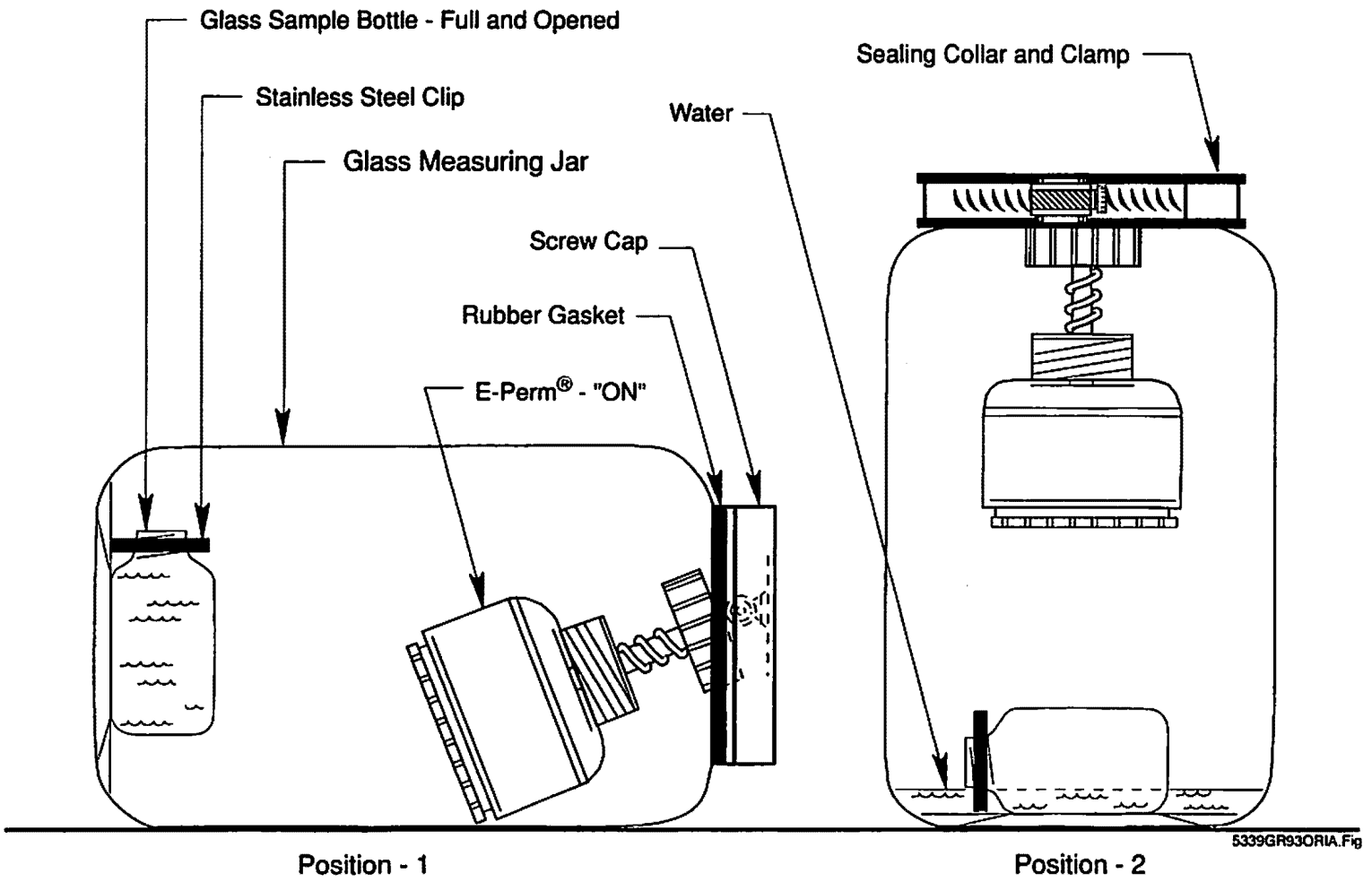

FIGURE 2

PHASE-1 RESULTS

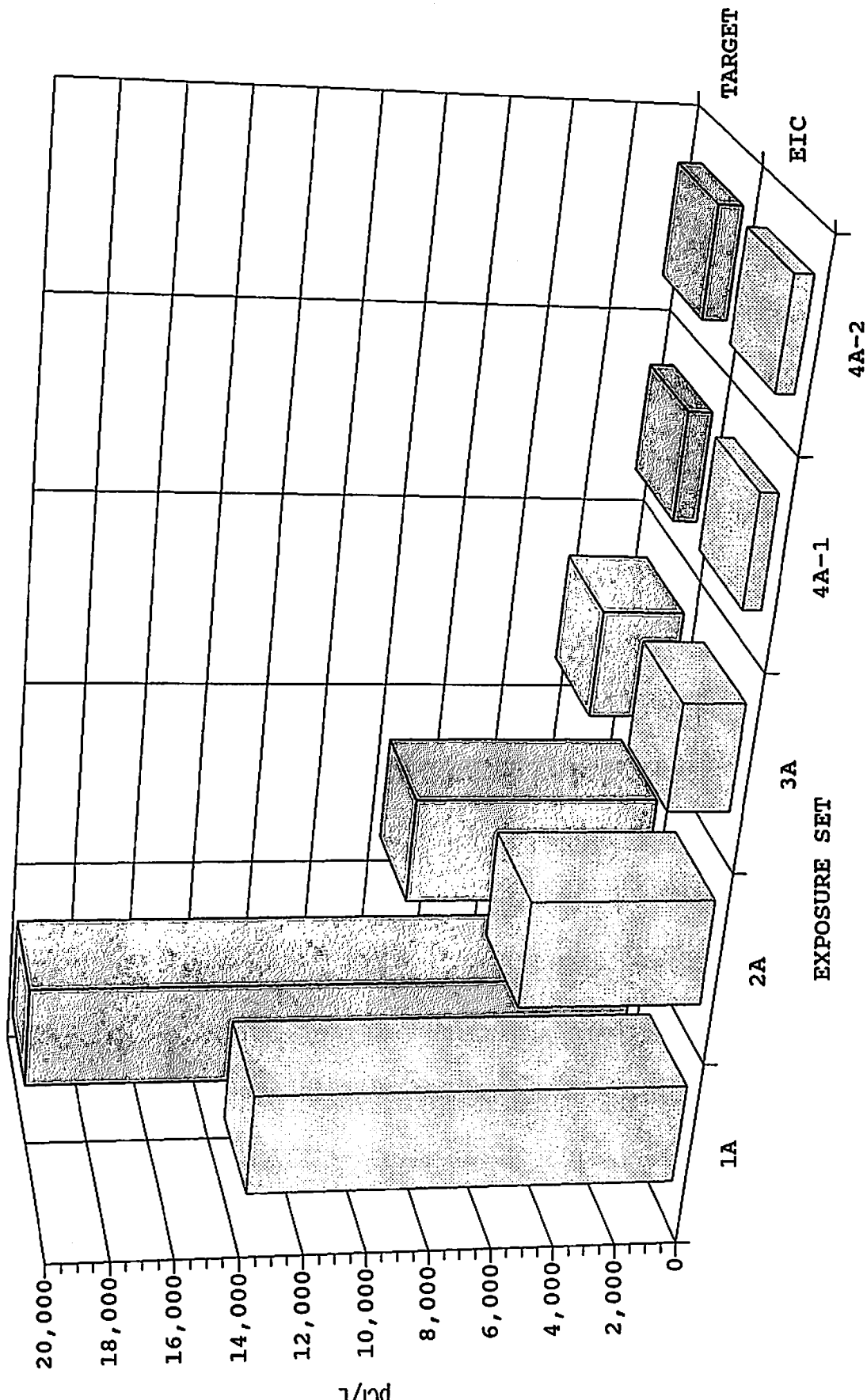

FIGURE 3

PHASE-2 RESULTS

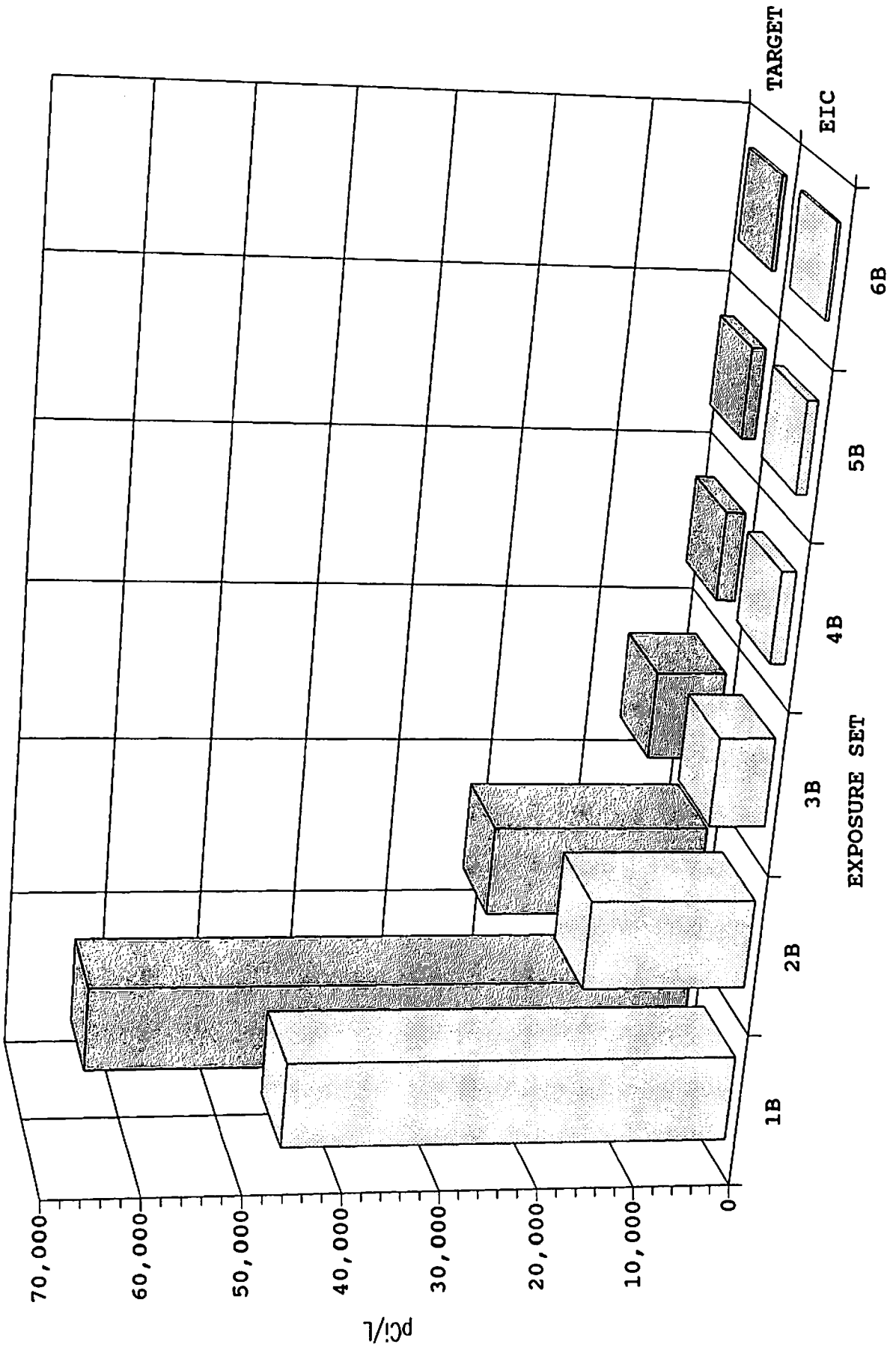

FIGURE 4

BIAS VS CONCENTRATION (PHASE 1 and 2)