

AN ABSTRACT OF THE THESIS OF

Douglas M. Monroe for the Master of Science Degree in Physical Science emphasis in Chemistry presented on December 21, 1991.

Title: DETECTION OF THE DECAY PRODUCTS OF RADON:
ANALYSIS OF LEAD-210, BISMUTH-210 AND POLONIUM-210
BY LIQUID SCINTILLATION

Abstract approved: Alfred J. Evers

The primary purpose of this investigation was to develop a wipe test method for the analysis of the decay products of radon-222 utilizing liquid scintillation detection. Investigations were concerned with the three longer half-life daughters, lead-210, bismuth-210 and polonium-210.

Analyses were performed by wiping a series of standards of known concentration off a glass plate of known area. Calibration curves were then constructed and analyzed by the Quattro Pro spreadsheet program. Unknown samples were then wiped from known areas and observed activities related to concentrations by the slope-intercept method applied to the calibration curves.

Results obtained from the investigation indicated that the method was applicable on a laboratory scale when concentrations of the daughters were above the 2-3pCi/cm² range. Comparison of measured unknown values to actual values at these levels showed that the relative accuracy of the method was approximately 15%. Relative precisions were also approximately 15%. Lower relative accuracies and precisions were obtained at higher concentrations. Application of the method at environmental levels resulted in positive values for the three radon daughters. As concentrations were lower than in the laboratory, high relative standard deviations were observed. No way of determining the accuracies for the environmental samples was available.

**DETECTION OF THE DECAY PRODUCTS OF RADON:
ANALYSIS OF LEAD-210, BISMUTH-210 AND POLONIUM-210
BY LIQUID SCINTILLATION.**

**A Thesis
Presented to
the Division of Physical Sciences
EMPORIA STATE UNIVERSITY**

**In Partial Fulfillment
of the Requirements for the Degree
Master of Science**

**by
Douglas M. Monroe
December 1991**

Charles Gumbel
Approved for the Major Division

Faye N. Vowell
Approved for the Graduate Council

TABLE OF CONTENTS

| Chapter | | Page |
|---------|---|------|
| I | <u>Introduction</u> | 1 |
| II | <u>Experimental Design and Procedures</u> | 6 |
| | Instrumentation..... | 6 |
| | Lead-210 Standard Calibration..... | 7 |
| | Method for Analyses..... | 7 |
| | Preliminary Research..... | 9 |
| | Chromatography of Radium-DEF..... | 9 |
| | Analyses of Wiping Media..... | 10 |
| | Examination of Recovery/Activity vs Time.. | 10 |
| | Enhancement of Scintillation Detergent.... | 10 |
| | Calibration Curves..... | 11 |
| | Statistical Analyses..... | 11 |
| | Environmental Samples..... | 11 |
| III | <u>Results</u> | 13 |
| | Preliminary Research..... | 13 |
| | Chromatography of Radium-DEF..... | 14 |
| | Choice of Wiping Solvent..... | 14 |
| | Examination of Recovery/Activity vs Time.. | 15 |
| | Enhancement of Scintillation Detergent.... | 16 |
| | Calibration Curves..... | 17 |
| | Analysis of Standards..... | 17 |
| | Environmental Samples..... | 18 |
| IV | <u>Discussion</u> | 20 |
| | Recommendations for Additional Study..... | 22 |
| V | <u>Bibliography</u> | 24 |
| | <u>Appendix I</u> | 39 |
| | <u>Appendix II</u> | 40 |
| | <u>Appendix III</u> | 41 |
| | <u>Appendix IV</u> | 42 |

LIST OF TABLES

| Table | | Page |
|-------|---|------|
| I | Chromatographic Separation of the Radon Daughters..... | 25 |
| II | Recovery of Various Wiping Media..... | 26 |
| III | Recovery of Wiping Media as Recorded by Channels..... | 27 |
| IV | Comparison of Experimental Concentrations Versus Known Standard Concentrations..... | 28 |
| V | Data and Results for Environmental Samples | 29 |

LIST OF FIGURES

| Figure | | Page |
|--------|---|------|
| 1 | Lead Standards Placed on Filter Paper..... | 30 |
| 2 | Effects of EDTA Concentration..... | 31 |
| 3 | Sample Activity Compared to Standard..... | 32 |
| 4 | Percent of Standard Activity Observed..... | 33 |
| 5 | Percent Standard Activity by Channel..... | 34 |
| 6 | Addition of EDTA to Scintillation Vials.... | 35 |
| 7 | Combination of Three Calibration Curves.... | 36 |
| 8 | Calibration Curve 5-3-91 by Channel..... | 37 |
| 9 | Activity as a function of Area Wiped..... | 38 |

CHAPTER I

Introduction

In recent years radon gas has gained much publicity in both scientific and public circles. Since it has been considered a leading cause of lung cancer, much concern has been expressed about radon levels in homes (1-5). As a result research effort has been devoted to developing new analytical techniques for low level samples along with determining its effects on both human and other animal populations. What is not usually studied, however, is the eventual fate of radon gas and its decay products.

Radon, being a radioactive element, does not stay as radon-222. Just as it is a daughter in the decay scheme of uranium-238, radon-222 itself decays further to give a series of its own decay daughters (Appendix 1). These include, in order, lead-214, bismuth-214, polonium-214, lead-210, bismuth-210, polonium-210 and lead-206. Lead-206 is stable and thus is the end point in the decay scheme of uranium-238 or the radon-222 daughters.

The problem considered was to develop a method to test for these decay products of radon-222 using samples of known activity and to apply this same method to environmental samples. The idea was that radon gas in the immediate environment would decay. These decay products, which are solids, would not be supported by the atmosphere and so would deposit on dust particles in the air or any solid surface available. The dust or objects could then be analyzed for the radon daughters. The law of secular equilibrium states that if a daughter product has a much shorter half-life than the parent, then the activity of the two will be equal (6). This

law applies in the case of radon-222 and its short-lived daughters, since they are all decay products of a much longer half-life nuclide, radium-226 or uranium-238 if it is traced back to the source. Radon-222 and all of its daughters are in secular equilibrium with radium-226. Basic application of this law tells us that if there is radon in the air, then its decay products should be present also. Therefore, if we could find some way of analyzing dust where radon is present (almost everywhere) we should also find all of its daughters.

Another interesting concept that arises is that since some of the daughters such as lead-210 have a longer half-life than radon-222, it is possible that they could be used to give a better picture of the average radon level in an area. Since radon is a gas, its concentration in a house can be changed by simply opening a window. Outside air, which is typically lower in concentration of radon than inside air, would have a dilution effect on the indoor radon levels (7). Thus, an accurate depiction of the radon concentration in an area might be hard to obtain with only one test. Depending on the time of the test, and the corresponding ventilation of the house, a measurement of the radon levels might be misleading. For example, if windows were open during the testing of radon levels, the results would be artificially low. This may or may not be an accurate indication depending on whether or not the house is always under these same conditions. Tests would have to be done over a longer time span to give more reliable information and an average level. Lead-210, however, has a half-life of over twenty-two years. While radon concentration might vary greatly from month to month, the lead-210 concentration would probably show less variation over a longer time frame such as a year. If this were true, then only a few tests would need to be done for lead-210

while many more tests would need to be done over a much longer time frame for typical radon measurements.

Conceptually, this sounds easy, but several problems present themselves immediately. The method used for radon gas is to trap it on activated charcoal, then analyze for it with some sort of detector. This is not as easy to do with the solid radon daughters. The dust could be filtered from the air onto charcoal with a pump, but the charcoal would absorb most of the radiation that the daughters emit. Preliminary investigations have shown this to be the case.

A new method would have to be developed. Some way by which the daughter products could be removed from the sample (usually dust) and placed in a medium for analyses had to be devised. One major factor which needed to be taken into account was that not all of the daughters decay by the same primary mode of decay. Lead-214 and bismuth-214 are short-lived beta and gamma sources. Polonium-214 is an extremely short-lived alpha emitter. Longer-lived lead-210 and bismuth-210 decay via beta emission, and polonium-210 decays via alpha emission (Appendix II).

In addition, there was some concern over the short half-lives of the first three decay daughters. Since the method developed would most likely involve taking solid samples, some problems could develop. For example, upon isolation from their radon parent, the daughters would continue decaying as normal. This would occur without them being replaced as in the atmosphere. The shorter half-life nuclides would decay and reach undetectable limits fairly quickly. With the short half-lives, analysis would need to be done immediately after sample removal to give an accurate measurement of these daughters' activities. Immediate analysis would not be as efficient as a method which allowed for a reasonable sample preparation time.

Lead-210, and the subsequent daughters, could facilitate this with their longer half-lives. Although lead-210, too, would decay without being replenished, it has a half-life of over twenty years. It would not diminish significantly during the time frame of the analysis. Taking into account the above factors, the scope of the research was changed to just analyze for the three longer half-life daughters of radon-222, which are lead-210, bismuth-210 and polonium-210.

In order to detect these three sources at environmental levels, a very sensitive method had to be developed. As the three daughters decayed by either alpha or beta emission, the method had to involve an instrument capable of detecting these particles. The instrument would also need to be capable of detecting the low energy beta emission of lead-210 and the non-penetrating alpha emission of polonium-210. Liquid scintillation was the most capable of meeting these requirements.

Liquid scintillation involves the placement of the sample in a scintillation solvent. As the sample decays, the emitted particles interact with fluors in the solvent which in turn emit blips of light. One blip of light is produced for each particle the sample emits. The intensity of this light is also related to the energy of the particle absorbed. The liquid scintillator counts these blips and measures their intensity via a phototube and then processes this information in a manner determined by the investigator. By changing the scintillation solvent, different fluors can be introduced which interact with different types of decay products, thus, one can choose the solvent and fluors to suit the particular type of radiation that one is analyzing. In addition, the method of liquid scintillation is extremely efficient since the sample being analyzed is in intimate

contact with the detector, the flour. This enables efficient detection of very low-energy beta and non-penetrating alpha particles.

Starting with these basic considerations and concluding with a method of analyzing for the long-lived daughters of radon-222 was the scope of this research. The remainder of the research project was concerned with the development and implementation of this analysis.

CHAPTER II

Experimental Design and Procedures

Instrumentation

The primary method of analysis in this research was liquid scintillation. The instrument used was a LKB-Wallace RackBeta model 1217-811 liquid scintillator. The instrument was an automated model with sample changing capabilities. All data acquisition and manipulation were controlled by an IBM PC attached to the scintillator. The instrument is capable of monitoring four channels simultaneously.

Channel I was set to analyze over the entire energy range. Channel II was set to analyze for the lower-energy radiation. Channel III was set for the middle-energy range of radiation and channel IV was set to monitor the higher-energy radiation.

These channels also roughly correspond to the energies of radiation given off by the three radon daughters. Lead-210 decays at a lower energy and corresponds to channel II. Bismuth-210 is of a middle-energy decay and corresponds to channel III. Polonium-210 decays at a higher energy and corresponds roughly to channel IV. Channel I would record the total activity. The efficiency of the scintillation detector was calculated by comparing known standard activities with their experimentally determined activities. A lead-210 standard was used. The activity recorded by the scintillator was nearly three times that expected from the standard itself. This was due to the secular equilibrium of bismuth-210 and polonium-210 which had been established with the lead. All three exhibited equal activities. As expected, the detector was found to be nearly 100% efficient.

The scintillation solvent used was Ecolume, a biodegradable, nonhazardous detergent specific for beta and alpha decay. It contains the primary flour 2,5-diphenyloxazole (PPO) and the secondary flour 1,4-bis-2-(5-phenyloxazole)-benzene (POPOP) (Appendix 3). The object of the primary flour is to interact with the decay particles and emit a blip of light. The secondary flour shifts the wavelength of this light to a region which can be detected by a phototube. Utilizing this system, the radiation from substances which emit beta or alpha particles can be detected.

Lead-210 Standard Calibration

A single lead-210 standard was used throughout the entire research. It contained lead-210, bismuth-210 and polonium-210 at a concentration of 113pCi/uL with a standard deviation of plus or minus 2pCi/uL for each. This was prepared from a standard solution of carrier-free lead-210 nitrate (Appendix 4). Approximately 100uL of the standard was diluted with water to a volume of 10ml. Since the original standard's concentration was a solution that had been last assayed in 1961, it was calibrated against a standard of bismuth-210 via analysis on a Geiger-Muller tube (Appendix 4).

Method for Analyses

All analyses were done by wiping a known area of glass to remove analytes. Standards and knowns were prepared on a large glass plate which was marked into sample sections of known area. An area of 50cm² (5x10cm) was found to be the most convenient, it provided the largest area for wiping without being too

inconvenient to handle. Known amounts of standard were transferred to the glass plate by Eppendorf pipet and allowed to dry for approximately ten minutes in an oven at 110°C. (Care was taken to wear gloves at all times when working with the standard.) All analyses were done on glass because it proved to be the most reproducible surface in the lab.

Wipes of the standards were then performed by wetting an approximately 1cm² piece of filter paper with a suitable solvent. Preliminary work showed that 0.1g ethylenediaminetetraacetic acid (EDTA) per one milliliter of pH10 ammonia-ammonium chloride buffer worked most satisfactorily. The buffer was prepared by dissolving 6.75g of ammonium chloride in 57ml of concentrated ammonia and diluting with water to a volume of 100ml (8). Whatman #40 ashless filter paper was used as the wiping pad.

After wetting with solvent, the filter paper was then rubbed smoothly over the entire sample area; care was exercised to wipe the entire sample. The most reproducible results were obtained if the filter paper was used to wipe the sample once and then turned over and wiped again. The moist pieces of filter paper were then placed directly into scintillation vials without drying. Each scintillation vial contained four milliliters of the scintillation detergent.

Preliminary work indicated that an equilibration time was needed between sample preparation and analysis. A time of 24 hours was found to be adequate. After waiting for this period following placement in the scintillation liquid, the samples were then analyzed for total alpha and beta emissions and energy of radiation with the scintillation detector.

Analyses of Wiping Media

Samples of lead standard were placed on a glass plate in 50cm² areas at a volume of 50uL each. Wipes were done on each square with various wiping media. These wipes were placed directly into the vials for analysis. One vial was also used with 50uL standard pipeted directly into it. The activity of this vial was then assumed to be the highest obtainable value. Wipe activities were then compared to this value to determine their percent activity of the actual.

Ethylenediaminetetraacetic acid (EDTA) was chosen as the solvent, and its optimum concentration was then determined. Lead standard was placed in 50cm² sample areas on a glass plate in 10uL portions. These areas were then wiped with concentrations of EDTA solution varying from 0.02g/ml to 0.15g/ml in pH10 buffer.

Examination of Recovery/Activity vs Time

One vial was set up with 25uL of lead-210 standard pipeted directly into it. Another contained a 1cm² piece of filter paper upon which was placed 25uL of the standard. The stop was removed from the liquid scintillator so that the two vials activities would be recorded one after the other until the program was stopped. Analysis on each vial took approximately 10 minutes.

Enhancement of Scintillation Detergent

To each of four pieces of filter paper 100uL of lead-210 standard was applied. These were then placed in scintillation vials along with 5ml of scintillation detergent. A solution of EDTA, 0.1g/ml in pH 10 buffer, was then added in 100, 500

and 1000uL portions to three vials. No EDTA solution was added to the fourth vial. These vials were then analyzed after waiting one hour.

Calibration Curves

Lead-210 standard solution was micropipetted onto 50cm² areas on a glass plate in varying volumes. The plate was then dried for ten minutes in an oven at 110^oC. The samples were then wiped with the 0.1g/ml EDTA solution in pH10 buffer and placed in the scintillation detergent for analysis.

Statistical Analyses

Known concentrations of the lead standard were placed on a glass plate and dried in an oven. These were then wiped with the 0.1g/ml EDTA solution and analyzed. A calibration curve also was constructed and analyzed along with these standards. The slope and intercept of this calibration were then calculated utilizing the Quattro Pro spreadsheet program and used to determine the concentrations of the known samples. These experimental concentrations were then compared with expected values. All determinations of knowns were done in groups of three.

Environmental Samples

Environmental samples, with the exceptions noted, were taken in the same manner as described in the method section. Instead of using a glass plate, however, samples were taken directly from windows or other glass surfaces in the

area being analyzed. A piece of plastic was cut with a 50cm^2 rectangle in the middle to facilitate wiping a constant area. EDTA in pH10 buffer at a concentration of 0.1g/ml was used as the wiping medium. A calibration curve was prepared for each set of samples analyzed. Environmental concentrations were then calculated from this calibration by using the best straight line equation. An ATT model 6310 computer was used with the Quattro Pro spreadsheet for all calculations.

CHAPTER III

Results

Preliminary Research

Several factors needed to be examined to develop a standard method for analysis of the radon daughters, and to determine if the development of the method were even feasible. The method would need to involve some way of picking up a trace solid sample and transferring it into a scintillation detergent for analysis.

It was decided that a satisfactory way to do this would be by wiping the sample to be analyzed with some sort of wiping media placed on a wipe pad. This would then be placed in the scintillation detergent for analysis. Since filter paper was available, reasonably cheap, and sturdy when wet, it was chosen as the wiping pad.

As a preliminary test, samples of a lead, bismuth, and polonium standard were introduced onto filter paper and analyzed on the liquid scintillator. This was done simply to see if the samples could leave the filter paper and be detected in the detergent. The results of this can be seen in Figure 1. The plot relating concentration to observed activity is linear within statistical uncertainty. This shows that samples on filter paper can give reasonable quantitative results. The activity of a sample placed on filter paper versus the same amount of sample placed directly into a scintillation vial was also examined. It was found that 61% of the sample from the filter paper could be detected.

Chromatography of Radium-DEF

The fact that each of the three daughters emitted radiation of different energies suggested that each daughter might correspond to a separate channel on the scintillator for detection. This was tested by separating the lead standard into its three components by a standard method known as chromatography radium-DEF (9). Each daughter was then analyzed on the scintillator to determine which channel its radiation corresponded. The results are presented in Table I. The daughters do not totally correspond to one channel, but are isolated slightly in each. The lead-210 showed 77% of its radiation in channel II. The bismuth-210 corresponds as expected with channel III, but only 57% of its radiation occurred there. Polonium is divided nearly equally between channels III and IV.

This does lend itself to describing channel II output as coming mainly from lead-210, but does not apply as well to the bismuth and polonium. It should also be noted that there is probably some tailing between the samples. This means that there was most likely some overlap of compounds on the filter paper, especially between the two compounds with similar R_f values, bismuth and polonium. It does say that these are probably not the best results this method is capable of obtaining. Overlap of the three standards caused some of the overlap in the channels. If pure standard samples were obtained and analyzed we might find that the radiation from each sample were isolated even more in one particular channel.

Choice of Wiping Solvent

After deciding on filter paper as the wiping pad, the choice of wiping media was investigated. Samples of standard were placed on a glass plate and wiped using

filter paper wetted with various solvents. Their activity was then compared with a known amount of standard injected directly into the scintillation detergent. From this percent recoveries for the wipe technique were determined. The results of this can be seen in Table II.

Table III breaks the recoveries down by channels. It is interesting to note that different solvents picked up varying amounts of sample in each channel. Since these channels roughly correspond to the three different daughters being analyzed, it is feasible to suggest that different solvents could be used to wipe for different daughters. For the rest of this analysis, EDTA was chosen as the wiping media. This was due to the fact that it picked up almost 94% of the channel II activity that was due mainly to lead-210, making it the most efficient solvent for the longest lived daughter in the decay chain.

The effect of concentration of the EDTA solution was also examined. Figure 2 shows how the activity varied with the concentration of the EDTA used on the wiping pad. Statistically, it appears that the concentration of the EDTA does not significantly change the recovery of the samples with the exception of at the highest concentration. This corresponds to changing the wiping medium concentration from 0.1g/ml EDTA to 0.15g/ml. This could possibly be explained by a change in pH brought about in the buffer by the added EDTA since the acid form of EDTA was used. A lowering of the pH could change the affinity of the metals for the EDTA (10).

Examination of Recovery and Activity vs Time

During the first few experiments, it was noted that widely varying results

were obtained for the recovery of the standard via the wipe tests. Perhaps some of the standards remained on the filter paper, or some sort of equilibrium needed to be reached for optimum analysis. This was examined by wiping a known amount of sample and comparing its activity with the sample placed directly into the scintillation detergent.

Figure 3 shows the activity of the sample wipe and the known activity as a function of time. It can be seen that some time is required for the radon daughters to equilibrate with the scintillation detergent. Figure 4 shows similar information expressed as percent recoveries versus time. It can be seen here that the recovery is very much dependent on how much time elapsed after wiping prior to analysis. Figure 5 expresses the results of Figure 4 by channels. The percent recovery increases in all channels with time. These results indicate that there is a minimum time needed for efficient recovery by this wipe method. An equilibration time of twenty-four hours was chosen for the sample, before analysis in order to give the best efficiency possible. No significant change in percent recovery was observed after a twenty-four hour equilibration period.

Enhancement of Scintillation Detergent

Given the long waiting times needed prior to analysis, some way was sought to speed the equilibrium of the sample between the filter paper and the scintillation detergent. It was thought that the long equilibrium times might be due to the strong affinity of the analytes for the EDTA solution on the filter paper. This was expected to be overcome by adding the same EDTA solution to the detergent, thus decreasing the equilibrium times. Known volumes of EDTA solution were added to the

detergent and the sample wipes placed in them. The results of this can be seen in Figure 6. As can be seen, the addition of EDTA to the detergent has negative effects on the analysis. This is probably explained by the fact that the EDTA is hydrophilic while the scintillation detergent is fairly hydrophobic. The EDTA solution is probably attracted to the paper, and likewise the radon daughters to the EDTA, thus increasing the affinity of the standards for the filter paper.

Calibration Curves

The preliminary research showed that the filter paper was capable of holding and releasing the samples linearly with concentration. It was necessary to find out if this was true when a wipe test was done. Varying amounts of standard were analyzed using the wipe method. Their activities were then plotted against the amount wiped. The results of several representative calibration curves can be seen in Figures 7 and 8. Figure 7 shows only total activity for several curves, while Figure 8 is broken down by channels for one. As can be seen, the results are linear within reasonable uncertainties. In addition, the data was taken over a wide range of concentrations (0-100,000 counts/min), so the linearity should prove applicable in almost any situation which would be encountered. Since they are "linear", when analyzing an unknown sample a calibration curve can be constructed, and the unknown concentration simply calculated from the slope and intercept in much the same way as Beer's law is applied to UV-VIS spectroscopy.

Analysis of Standards

Applying all the factors discussed above, the analysis of an unknown was

attempted. A calibration curve was constructed from standards, then known samples were wiped and their calculated concentrations compared. The results of a few of these can be seen in Table IV.

Concentrations were calculated using the slope-intercept method. Values for relative errors ranged from -45.7% to +63.2%. The average absolute value for the errors was approximately 13.0%. The standard deviation for the errors was plus or minus 20.0%. Collectively, the experimental values averaged out to 2.8% above the expected. This suggests that, with a large number of trials, the method uncertainties tend to average out. Although the statistical data does not appear great for this method, it should be noted that most of the error is due to just five runs out of a total of thirty-three. Excluding these, the relative errors range from -16.0% to 18.8%, and the average deviation of the errors is only 7.9%.

It seems likely that the runs in question involve some sort of experimental error. Some error and poor reproducibility can be attributed to those inherent in the scintillation instrument and Eppendorf pipets. The largest error is probably due to the natural errors inherent in the wiping method itself.

Environmental Samples

Using the methods which were developed in this research, several environmental samples were obtained and analyzed. Although all samples showed activity above background, good reproducibility was not obtainable. Wipe data taken from various sources in Cram Science Hall can be seen in Table V. The environmental sample concentrations were found to range from 0.09 to 0.34pCi/cm². In addition, a plot of activity vs area wiped for some of the environmental samples was constructed.

This is shown in Figure 9. A plot of the last three areas appears linear, but the uncertainty is very large. The low concentrations of the daughters in the environmental samples proved to be a major problem. Even with longer counting times, which would improve the statistics associated with the randomness in decay, poor relative precision was still a problem.

CHAPTER IV

Discussion

This investigation was directed toward developing a method for analyzing the three long-lived daughters of radon-222, and examining possible application to environmental samples. The daughters were transferred to a scintillation detergent by utilizing a wipe test with EDTA solution on filter paper. The sample was then analyzed utilizing liquid scintillation.

The feasibility of this method when applied to relatively high concentrations of the analytes was evident. The response of activity versus concentration of the analytes was linear or nearly linear in all cases. This applied to both the direct addition of the standard to the detergent or when wipe tests were done to obtain a sample. This means that samples could be wiped and their activities compared to a calibration curve to get a value for the concentration of the analytes.

Recoveries for the parent-daughter species investigated also varied by channel with different wiping media. Since each channel roughly corresponds to one of the radon daughters, this raises the possibility that different daughters could be analyzed separately by changing the wiping media. Results of the wipe test also support its application as a test method for the radon decay products. The wipe tests yielded good to average reproducibility and accuracy when applied to samples of known activity. All of the above factors indicate that the method could be applied with good reliability to the analysis of an unknown sample of relatively high concentration.

Application of the method to low levels of radiation in the environment did

not prove as feasible. The main problem was the poor reproducibility of the wipe tests. The accuracy of the tests was not obtainable on the environmental level since the concentrations of the analytes were not known. Values for the indicated concentration ranged greatly in the same sample in many cases. This is probably due to the fact that such small concentrations are being considered. The relative precisions do not seem to be constant over the entire concentration range. It seems more likely that the absolute precision remains constant or change only slightly for any given concentration.

The method may be good to plus or minus 1pCi whether the amount wiped is 10pCi or 100pCi. This would cause relative errors to increase as lower concentrations of analyte are examined. Another factor may be that different forms of the analytes were encountered in the environmental samples. In the prepared lab samples, all of the analytes existed in their cationic forms with nitrate as the corresponding anion. In the environment, things are not likely to be as simple. Obviously all of the analytes would not exist as the nitrate form. There may be complexes which form with the dust that behave much differently than what was found for nitrates in a controlled laboratory situation. There are probably interactions with many different species which were not present in the lab samples. This could also change the reproducibility of the method and the results when applied to environmental samples. Poor reproducibility for the environmental samples could also be due to the fact that the dust may not be distributed evenly on surfaces. This would probably mean that the radon daughters were not distributed evenly either. Any of the above factors could account for the poor precision observed.

Several positive factors of the method as applied to the environmental samples

were found. All environmental wipes did yield positive values for the radon daughters in the samples. Even with the poor reproducibility, the analytical method indicates that there is something there and at a detectable level. Samples that were expected to have high activities did have higher activities than other samples with the exception of the 10cm² wipes. The wipes inside a cabinet where uranium ore samples are stored did yield higher activity values than outside the cabinet. A glass plate on top of a uranium sample gave even higher results. In addition, the side closest to the ore registered higher activity than the opposite side. These seem to indicate that the method does work in a general sense. Further, the fact that the method did prove feasible in the laboratory samples at higher concentrations was positive. This leaves the possibility that if it could be validated on a smaller concentration level, it would prove to be an effective method for an analysis of the radon daughters in the environment.

Recommendations for Additional Study

Further work needs to be done in several areas. The causes for the poor reproducibility of the environmental samples needs to be investigated. The method needs to be modified in order to increase the precision at lower concentrations. If this difficulty is overcome, another area of interest might be resolution of each daughter's concentration from one sample wipe. If the distribution of radiation for each daughter in each channel were characterized better, some sort of curve fitting could be done to determine exactly what amount of radiation in a sample is attributed to each daughter. Utilizing this information, one could determine the concentration of each of the three daughters and possibly the total

average radon concentration from just one wipe analysis.

CHAPTER V
Bibliography

- (1) Otto G. Raabe and McDonald E. Wrenn, "Analysis of the Activity of Radon Daughter Samples by Weighted Least Squares." Health Physics, 17, (1969), pages 593-595.
- (2) James Bolger, "Radon: The Invisible Enemy." Safety and Health, January (1988), pages 42-44.
- (3) Bruce J. Srachta, "Radon is a Killer." Safety and Health, July (1987), pages 49-51,79.
- (4) Anthony Nero, "Earth, Air, Radon and Home." Physics Today, April (1989), pages 32-39.
- (5) Daniel M. Downey and Glenn Simolunas, "Measurement of Radon in Indoor Air." Journal of Chemical Education, 65:12 (1988), pages 1042-1044.
- (6) Herbert M. Clark and Ralph T. Overman, Radioisotope Techniques, McGraw-Hill. 1960. pages 305-310,333-336.
- (7) Julie A. Evans, "Radon: Reducing Risks." School and College, December (1988), pages 29-34.
- (8) R. A. Day, Jr and A. L. Underwood, Quantitative Analysis, Prentice Hall. 1991. pages 622-623.
- (9) Grafton D. Chase and Joseph L. Rabinowitz, Principles of Radioisotope Methodology, Burgess Publishing. 1967. pages 346-347.
- (10) John H. Kennedy, Analytical Chemistry Principles, Harcourt Brace Jovanovich. 1984. page 294.
- (11) Jerome L. Duggan, Laboratory Investigations in Nuclear Science, The Nucleus, Inc. 1988. page 160.
- (12) Chart of the Nuclides, General Electric Company. 1984.
- (13) Beta Disc Sources Model 2942 Instruction Manual, Picker X-Ray Corporation. 1962. page 5.

Table I
Chromatographic Separation of
the Radon Daughters

Total Counts by Channel:

| | I | II | III | IV |
|---|------|------|------|------|
| <hr style="border-top: 1px dashed black;"/> | | | | |
| ^{210}Bi | 7742 | 1073 | 4386 | 2282 |
| ^{210}Pb | 5927 | 4586 | 996 | 345 |
| ^{210}Po | 3778 | 142 | 1987 | 1647 |

Percent Activity by Channel:

| | | II | III | IV |
|---|--|------|------|------|
| <hr style="border-top: 1px dashed black;"/> | | | | |
| ^{210}Bi | | 13.9 | 56.7 | 29.5 |
| ^{210}Pb | | 77.4 | 16.8 | 5.8 |
| ^{210}Po | | 3.8 | 52.6 | 43.6 |

Table II
Recovery of Various Wiping Media

| Wipe Media | Activity | Background | Corrected | %Recovery |
|-----------------------|----------|------------|-----------|-----------|
| conc HNO ₃ | 3649.4 | 60.4 | 3589.0 | 11 |
| EDTA soln | 16893.5 | 101.2 | 16792.3 | 53 |
| 1.0M HCl | 14498.0 | 212.4 | 14285.6 | 45 |
| sat NaCl soln | 16486.6 | 60.7 | 16425.9 | 52 |
| 6M HNO ₃ | 11563.2 | 202.9 | 11360.3 | 36 |
| Methanol | 13547.0 | 361.2 | 13185.8 | 41 |
| Water | 17292.0 | 213.9 | 17087.1 | 54 |

100% Recovery= 31872.1cpm

Table III

Recovery of Wiping Media as
Recorded by Channels

| Sample | Channel: | | | |
|-----------------------|----------|----|-----|----|
| | I | II | III | IV |
| conc HNO ₃ | 11 | 26 | 7 | 3 |
| EDTA soln | 53 | 94 | 53 | 10 |
| 10% NaOH | 49 | 62 | 65 | 11 |
| 1.0M HCl | 45 | 71 | 50 | 10 |
| sat NaCl soln | 51 | 57 | 47 | 53 |
| 6M HNO ₃ | 36 | 57 | 42 | 5 |
| Methanol | 42 | 47 | 35 | 49 |
| Dist Water | 54 | 75 | 64 | 15 |

Table IV

Comparison of Experimental Concentrations Versus
Known Standard Concentrations

| Experimental Concentration | Actual Concentration | Relative Deviation |
|-------------------------------|-------------------------|-----------------------|
| 186pCi | 114pCi | 63.2% |
| 153 | 114 | 34.2 |
| 123 | 114 | 7.9 |
| 117 | 114 | 2.6 |
| 510 | 570 | -10.5 |
| 481 | 570 | -15.6 |
| 557 | 570 | - 2.3 |
| 492 | 570 | -13.7 |
| 614 | 570 | 7.7 |
| 714 | 570 | 25.2 |
| 886 | 570 | 55.4 |
| 585 | 570 | 2.6 |
| 622 | 570 | 9.0 |
| 598 | 570 | 4.9 |
| 309 | 570 | -45.7 |
| 653 | 570 | 14.6 |
| 581 | 570 | 1.9 |
| 1276 | 1140 | 11.9 |
| 958 | 1140 | -16.0 |
| 1053 | 1140 | - 7.7 |
| 1111 | 1140 | - 2.6 |
| 1049 | 1140 | - 8.0 |
| 959 | 1140 | -15.9 |
| 980 | 1140 | -14.0 |
| 1075 | 1140 | - 5.7 |
| 1130 | 1140 | - 0.9 |
| 1071 | 1140 | - 6.1 |
| 3373 | 2840 | 18.8 |
| 2844 | 2840 | 0.1 |
| 3009 | 2840 | 6.0 |
| 2860 | 2840 | 0.7 |
| 5075 | 2840 | -10.8 |
| 11618 | 11400 | 1.9 |

Average= 2.8%

Standard Deviation= 20.0%

Table V

Data and Results for Environmental Samples

| Sample | Activity | Area | pCi | pCi/cm ² |
|---------------------------------|----------|-------------------|---------|---------------------|
| ----- | | | | |
| Basement Window: | | | | |
| (Room 17, Cram Hall) | | | | |
| | 38.0cpm | 10cm ² | 17.3pCi | 1.73 |
| | 23.8 | 25 | 10.8 | 0.43 |
| | 26.6 | 50 | 12.1 | 0.24 |
| | 31.5 | 100 | 14.3 | 0.14 |
| Glass Plate in Basement: | | | | |
| | 20.3 | 50 | 9.1 | 0.18 |
| Storage Cabinet Door: | | | | |
| Inside | 16.0 | 50 | 7.2 | 0.14 |
| Outside | 10.3 | 50 | 4.6 | 0.09 |
| Uranium Ore Cover Plate: | | | | |
| Bottom | 18.7 | 25 | 8.4 | 0.34 |
| Top | 13.3 | 25 | 6.0 | 0.24 |
| Outside Window: | | | | |
| | 12.3 | 50 | 5.5 | 0.11 |

All samples taken July 10, 1991

Figure 1
Lead Standards Placed on Filter Paper

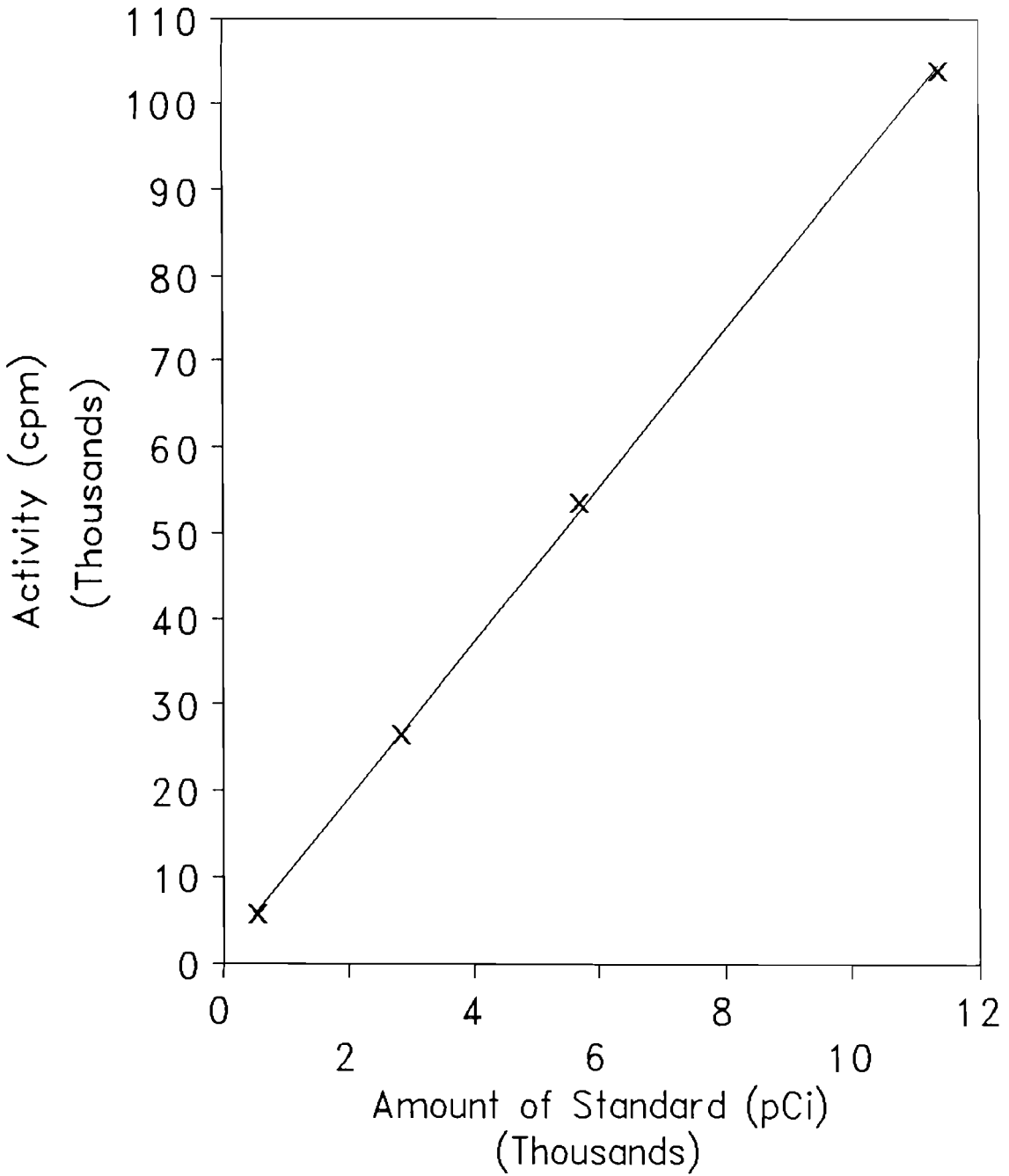
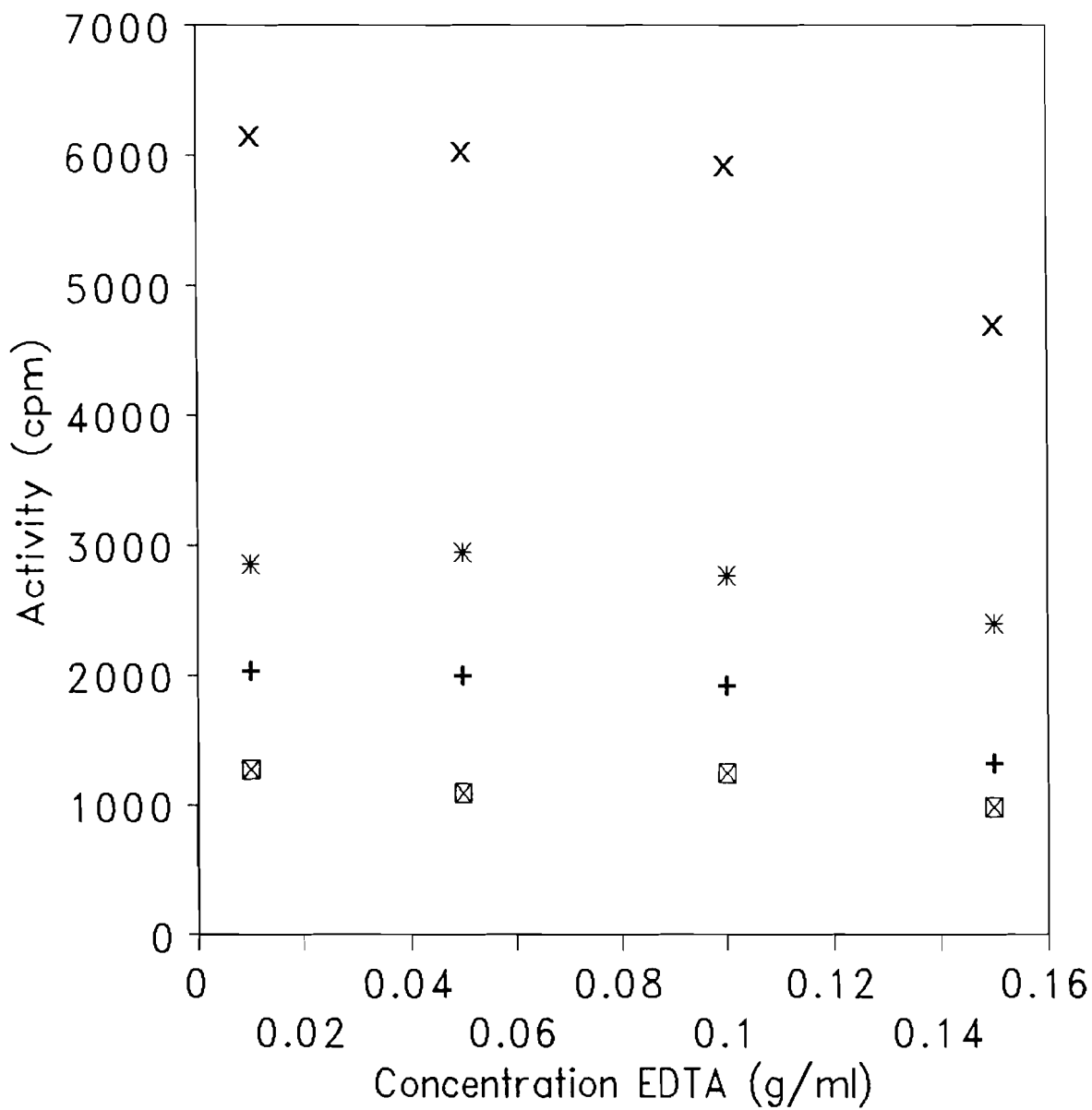
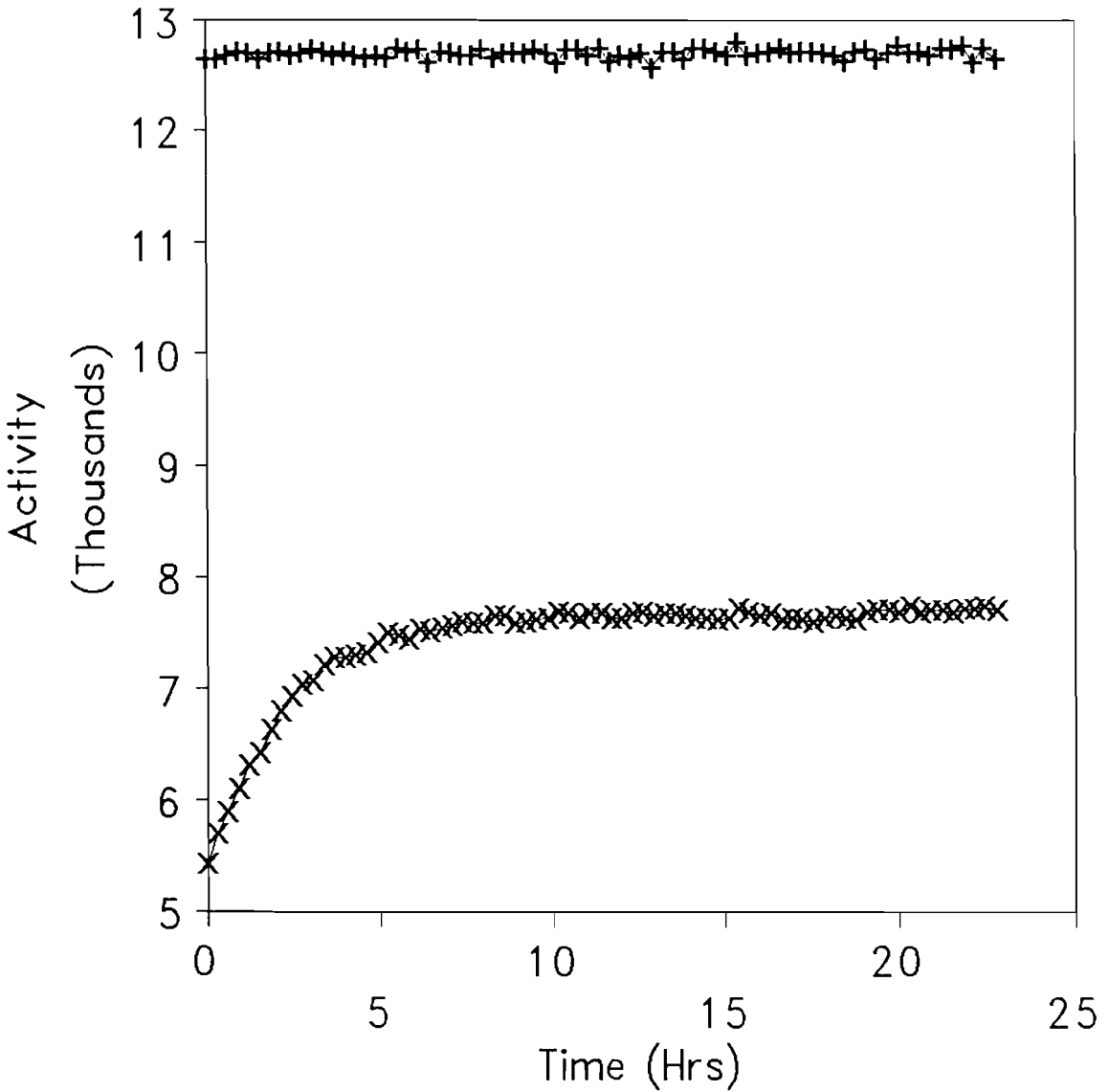


Figure 2
Effects of EDTA Concentration



x CH I + CH II * CH III x CH IV

Figure 3
Sample Activity Compared to Standard



—+— Standard —x— Sample

Figure 4
Percent of Standard Activity Observed

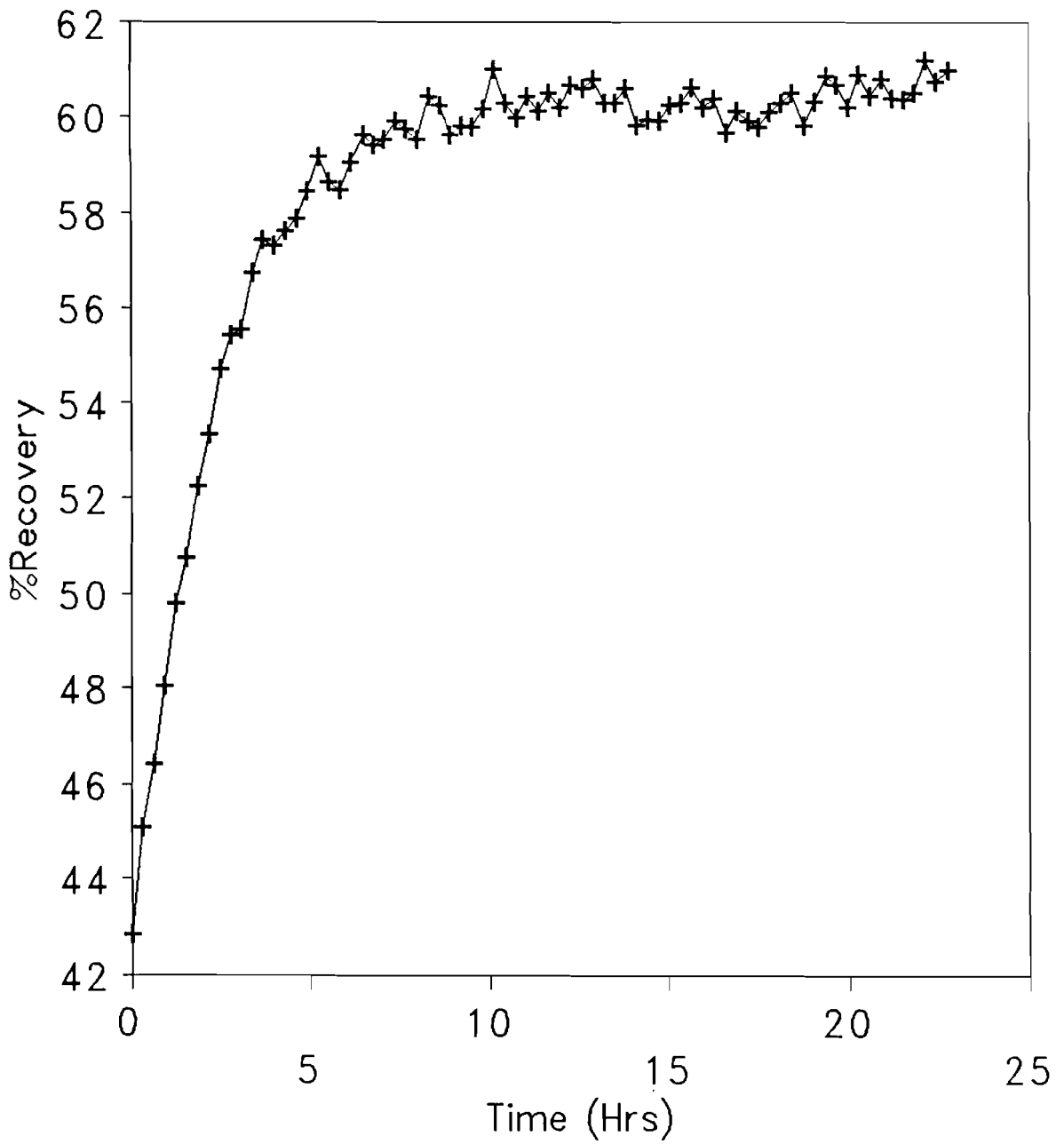


Figure 5
Percent Standard Activity by Channel

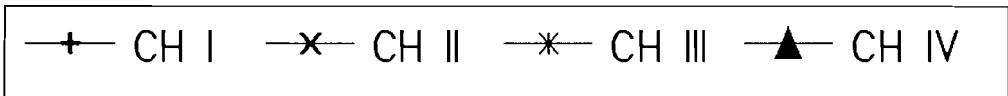
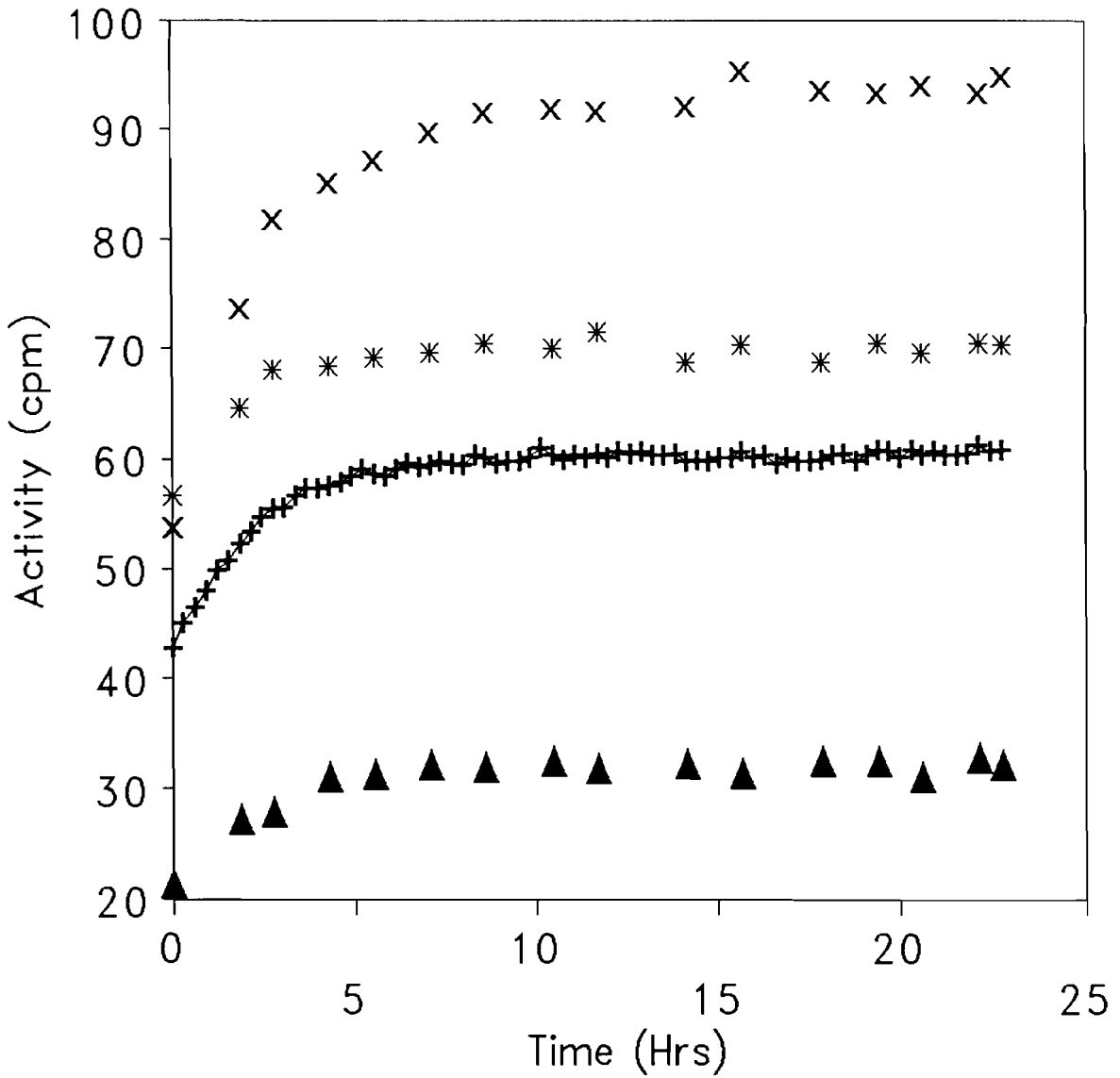


Figure 6
Addition of EDTA to Scintillation Vials

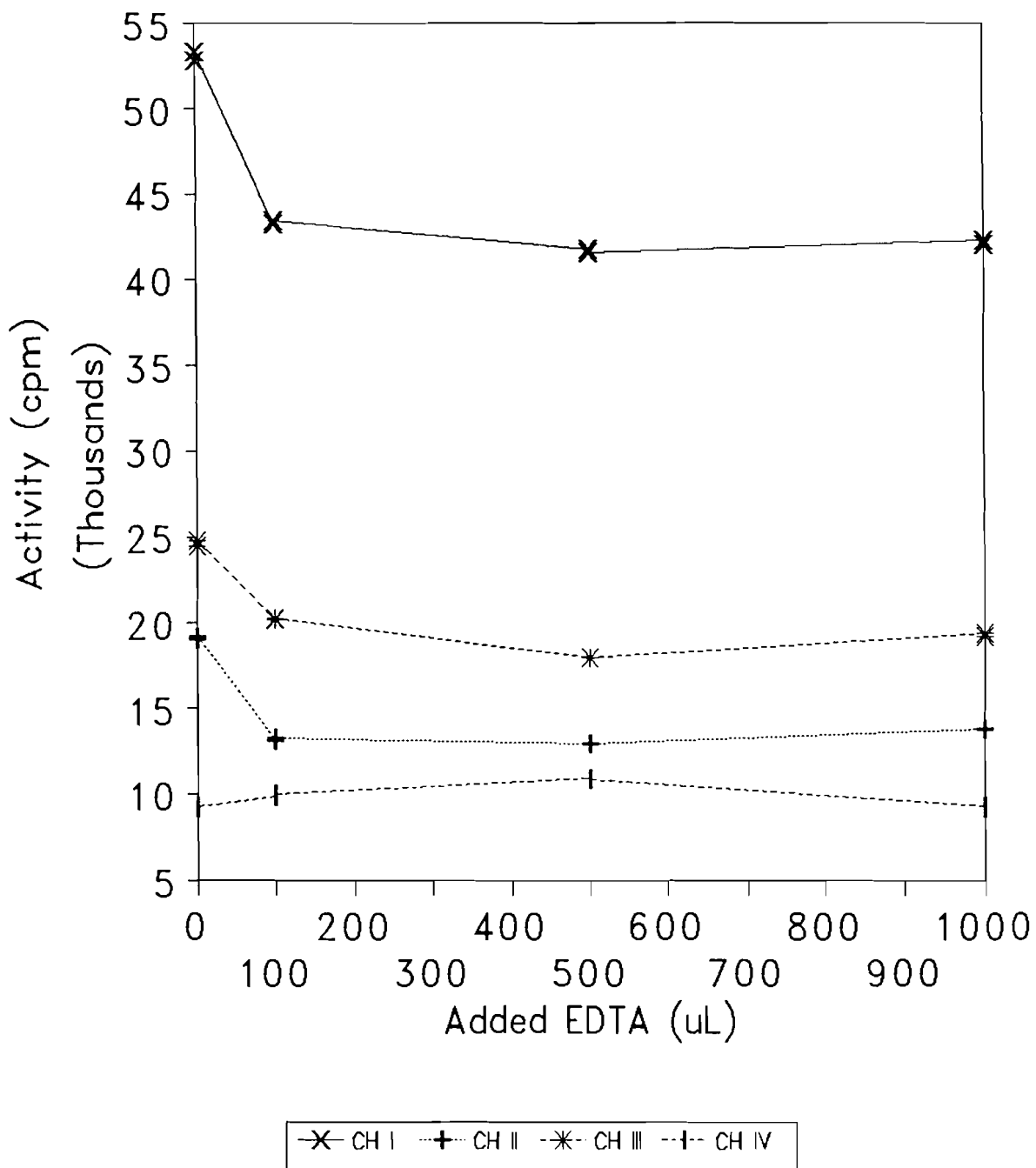
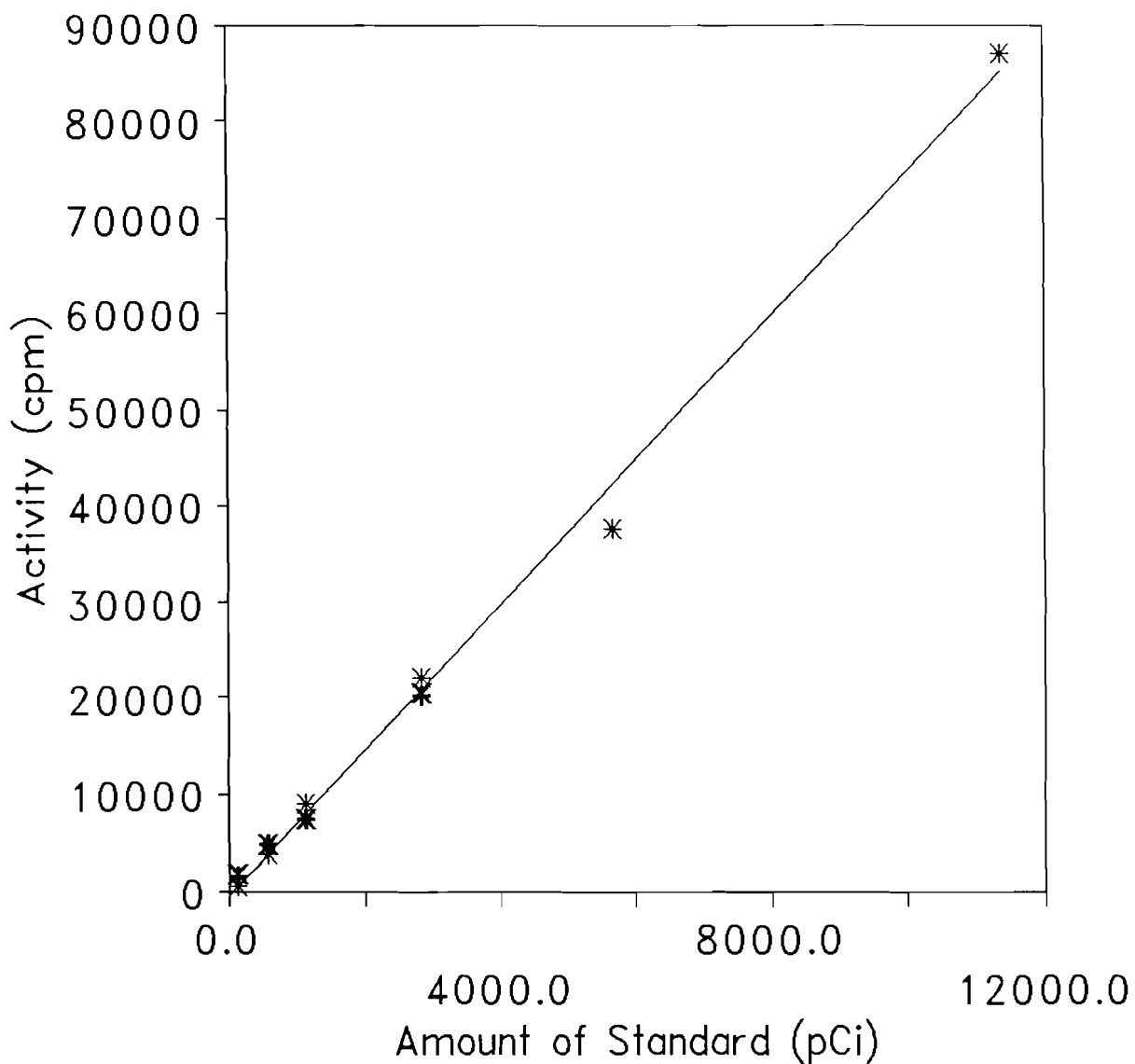


Figure 7
Combination of Three Calibration Curves



| | | | | | | | |
|---|------|---|-----|---|-----|---|-----------|
| + | 7-21 | x | 5-3 | * | 6-5 | — | Regressio |
|---|------|---|-----|---|-----|---|-----------|

Figure 8
Calibration Curve 5-3-91 by Channel

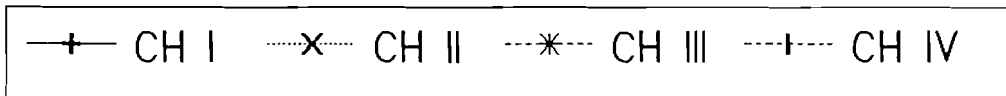
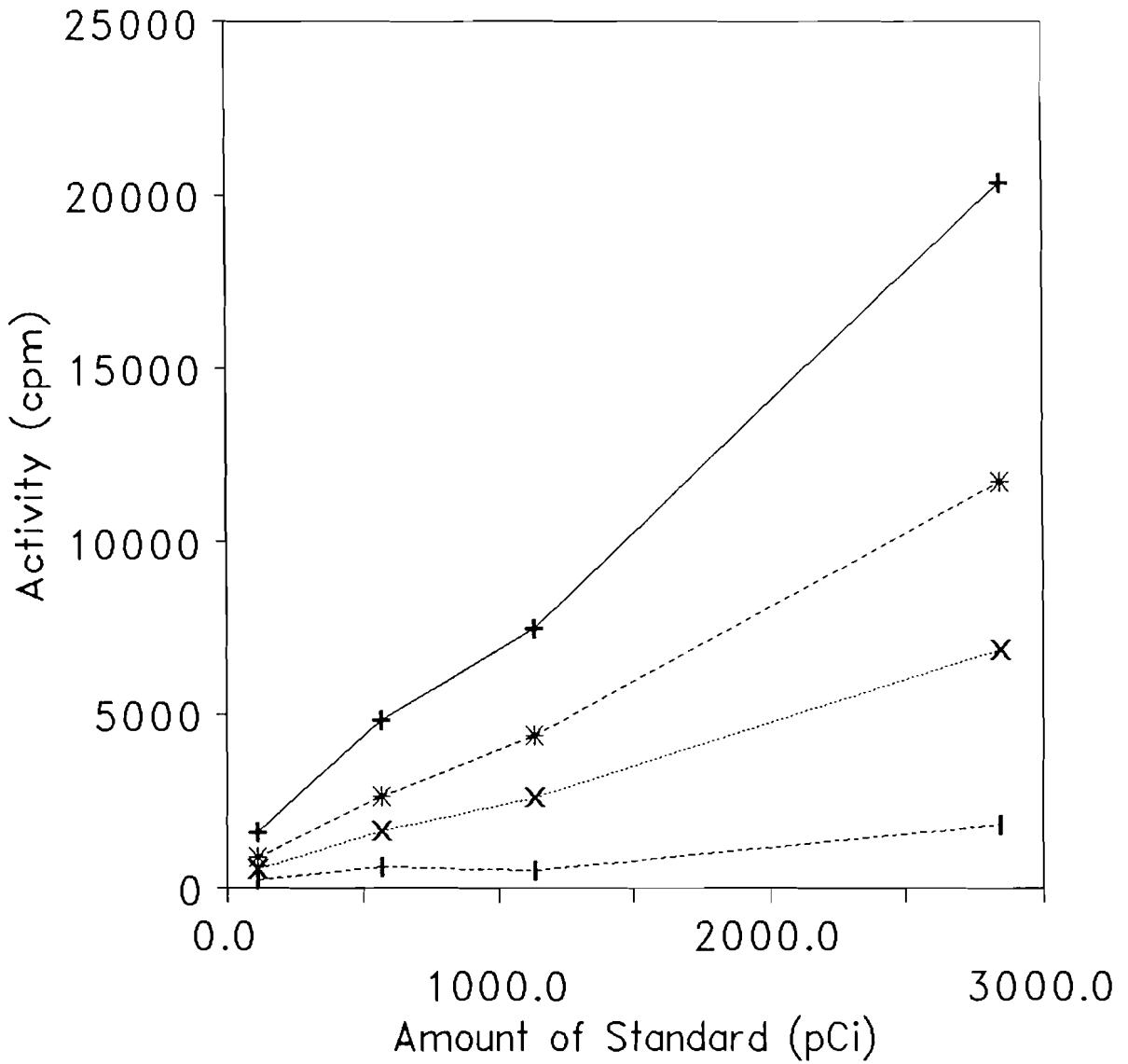
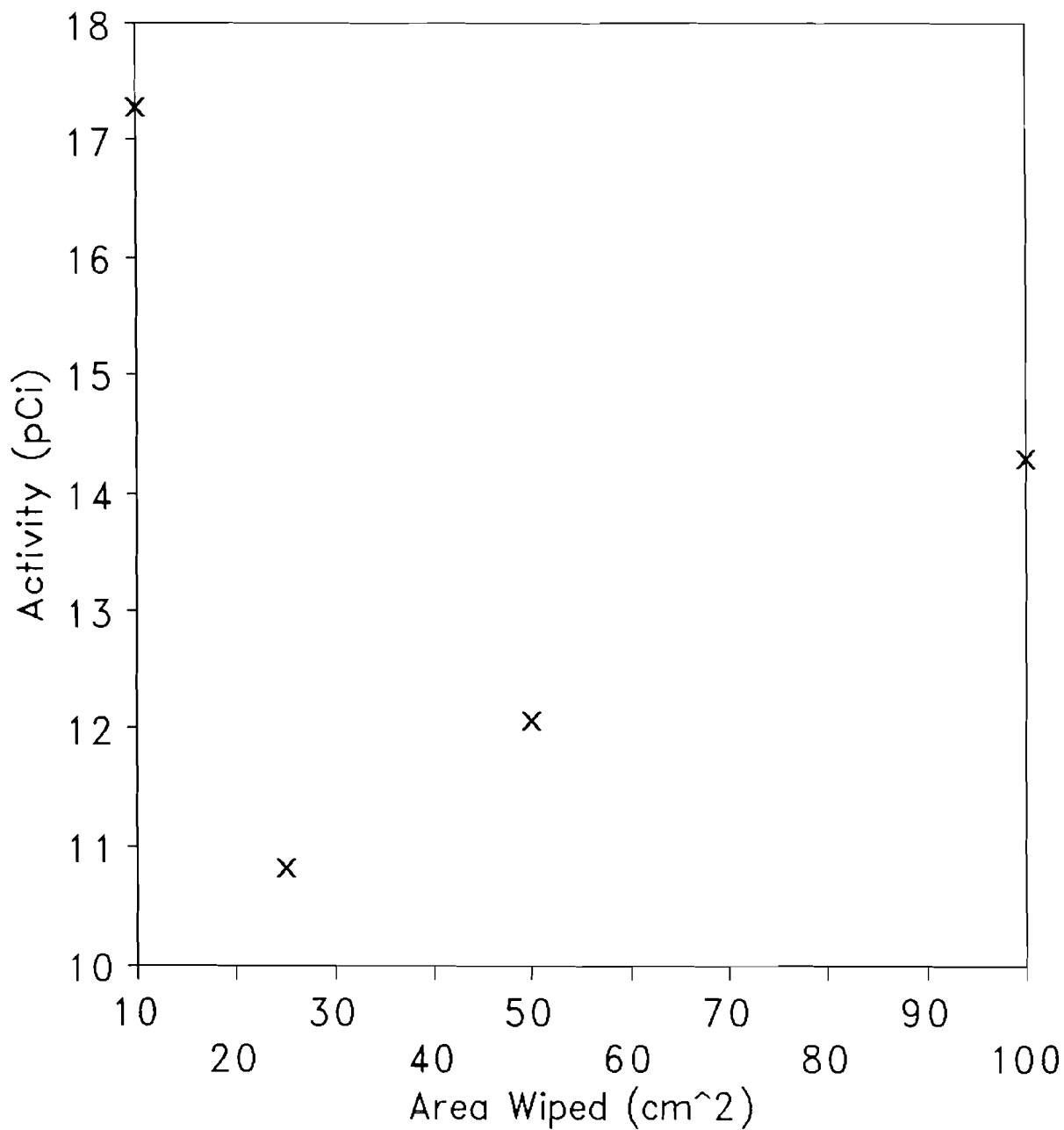
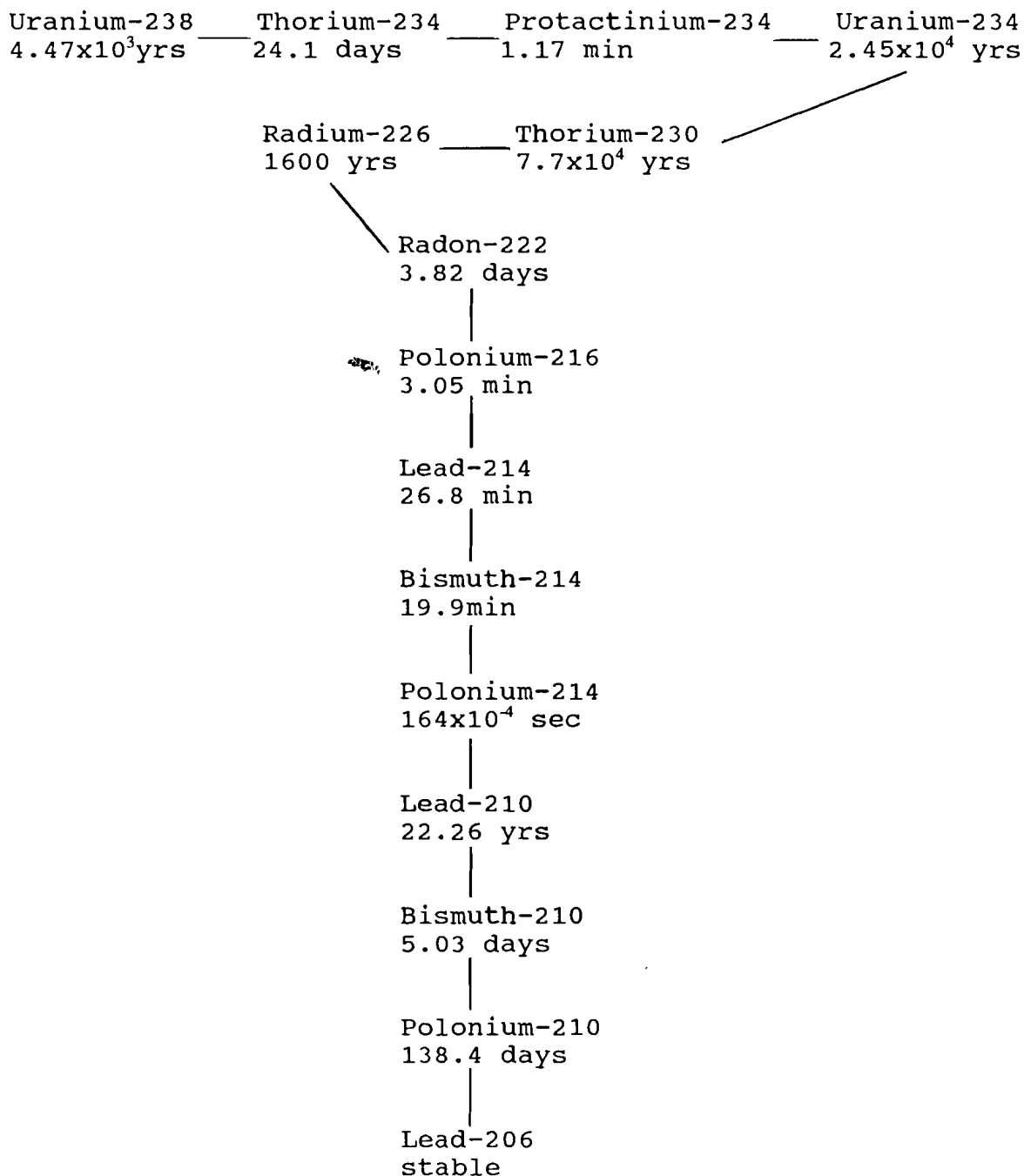


Figure 9
Activity as a Function of Area Wiped



APPENDIX I

Decay Scheme of Uranium-238¹¹



APPENDIX II

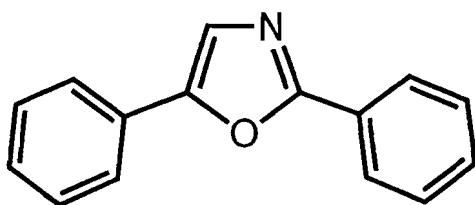
Half-life and mode of decay for
Radium-226 and its daughters¹²

| Isotope | half-life | primary mode of decay | energy |
|-------------------|--------------------------|--------------------------|--------------|
| ²²⁶ Ra | 1600y | alpha | 4.78MeV |
| ²²² Rn | 3.82 days | alpha | 5.49MeV |
| ²¹⁸ Po | 3.05min | alpha | 6.00MeV |
| ²¹⁴ Pb | 26.8min | gamma | 242-352KeV |
| ²¹⁴ Bi | 19.9min | gamma | 609-1120KeV |
| ²¹⁴ Po | 164x10 ⁻⁴ sec | alpha | 7.69MeV |
| ²¹⁰ Pb | 22.26y | beta | .017-.061MeV |
| ²¹⁰ Bi | 5.03days | beta | 1.161MeV |
| ²¹⁰ Po | 138.4days | alpha | 5.304MeV |
| ²⁰⁶ Pb | stable | ----- | ----- |

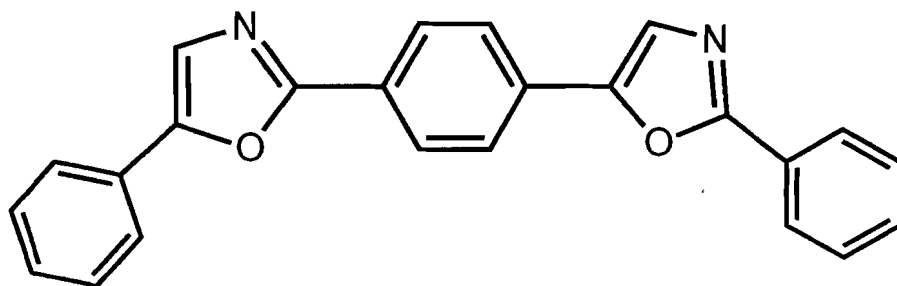
APPENDIX III

Structure of Primary and Secondary Flour Used in Liquid Scintillation

POP: 2,5-diphenyloxazole



POPOP: 1,4-bis-2-(5-phenyloxazole)-benzene



APPENDIX IV

Information on Bismuth-210 Source Used for Calibration of Lead-210 Standards¹³

Beta Source BP-4

| | |
|--------------------|------------------------|
| Color of Source: | Red |
| Isotope: | Bismuth-210 |
| Serial Number: | BP-4040 |
| Activity: | 0.0216 Microcurie |
| Half-life: | 19.4 years |
| Max. Beta MeV: | 1.16 |
| Gamma MeV: | None |
| Specific Activity: | Weightless |
| Absorber: | 7.5 mg/cm ² |
| Range: | 490 mg/cm ² |

Physical Characteristics: The source consists of an essentially weightless amount of lead nitrate on the copper planchet, covered with an aluminum absorber approximately 7.5 mg/cm² in thickness.

Basis for Calibration: National Bureau of Standards supplies calibrated amounts of Bi-210 activity mounted on a silver plate. A known fraction of the radiolead solution used in the preparation of the Bi-210 reference sources was deposited on another silver plate and the counting rates of the plates compared. Allowing for an 8% correction because the National Bureau of Standards source contained appreciable amounts of lead carrier, the stock solution was thus calibrated. Precise aliquots of this solution were prepared in identical fashion to that used for the Bi-210 reference sources and used as calibration standards. The accuracy is estimated at plus or minus 10%.

I, Douglas M. Monroe, hereby submit this thesis to Emporia State University as partial fulfillment of the requirements for an advanced degree. I agree that the Library of the University may make it available for use in accordance with its regulations governing materials of this type. I further agree that quoting, photocopying, or other reproduction of this document is allowed for private study, scholarship (including teaching) and research purposes of a nonprofit nature. No copying which involves potential financial gain will be allowed without written permission of the author.

Doug Monroe

Signature of Author

Dec. 9, 1991

Date

DETECTION OF THE DECAY PRODUCTS OF RADON:
ANALYSIS OF LEAD-210, BISMUTH-210 AND POLONIUM-210
BY LIQUID SCINTILLATION

Jacqui Saltzer

Signature of Graduate Office Staff Member

Dec 10, 1991

Date Received