

Proceedings of the 1995 API and GRI Naturally Occurring Radioactive Material (NORM) Conference

Exploration and Production Department
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Exploration and Production Department

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TABLE OF CONTENTS

Page

Session 1: Measurement and Survey

Determination of Naturally Occurring Radionuclides in Samples from the Gas and Oil Industry	3
W.A.I. Knaepen, W. Bergwerf, P.F.J. Lancée and G. Jonkers, Shell Research B.V. - Amsterdam	
Detecting NORM Contaminated Tubing During Pulling Operations	17
Thomas M. Williams, Texaco, Inc., EPTD	
High Density Radiological Site Characterization Surveys for NORM Sites	23
C.R. Flynn, M.S. Blair and R.R. Highfill, Chemrad Tennessee Corp.	
Methods of Estimating Dose to Individuals from NORM	35
J.L. Alvarez, Auxier & Associates; R. Gedes, Monsanto Chemical Co.; J. R. Rice, FMC Corporation	
Evaluation of NORM Site Release Criteria for Unrestricted Use	47
Alan McArthur, Central Environmental, Inc.; Simon Schroeder, Environmental Management, Inc.; William Lemons, Core Laboratories, Inc.	
Radioactive Lead: An Underestimated NORM Issue?	59
F.A. Hartog, W.A.I. Knaepen and G. Jonkers, Shell Research B.V. - Amsterdam	

Session 2: Regulatory Issues and Activities

Development of a Comprehensive NORM Program	73
Steve Woods, Steve Abernathy, Halliburton Energy Services; Peter Johnson, CORPEX Technologies, Inc.	
Transportation Regulations for LSA NORM	79
A. Wendell Carriker, Jack G. Albright, Office of Hazardous Materials Technology, Research and Special Programs Administration, U.S. Department of Transportation, Washington, D.C.	

TABLE OF CONTENTS (continued)

	Page
<u>Session 3: NORM Management and Disposal</u>	
Effective Removal of NORM Scale in the Rycroft Field	95
W.G.F. Ford, L.L. Gadeken, T.J. Callahan, Halliburton Energy Services; D. Jackson, Pan Canadian Petroleum Limited	
Technical Profile of the Envirocare of Utah, Inc. NORM Disposal Operation	103
Kevin C. Fuller, Envirocare of Utah, Inc.	
Modeling of NORM Injection in a Layered Geologic System	109
G.P. Williams, K.P. Smith, D.L. Blunt, Argonne National Laboratory; C.L. Tebes, University of Illinois Champaign/Urban	
<u>Session 4: NORM Scale Prediction and Control</u>	
Controlling Barium Sulfate Scale Deposition by Inhibitor Squeeze at the Guerra Well in South Texas: A Case Study	125
Michael J. McKenzie, Texaco E&P; Shiliang He, Rice University; Charles Hinrichsen, Texaco, Inc.; John E. Oddo, Water Research Institute; Anthony J. Gerbino, OLI Systems, Inc.; Amy T. Kan, Mason B. Tomson, Rice University	
A Laboratory and Field Study of the Mitigation of NORM Scale in the Gulf Coast Region of the United States	133
John E. Oddo, Xiliang Zhou, Water Research Institute; Jorge P. Gamez, Gas Research Institute; Shiliang He, Mason B. Tomson, Rice University	
Control of NORM at Eugene Island 341-A	143
P.J. Shuler, Chevron Petroleum Technology Company; D.A. Baudoin, Chevron U.S.A.; D.J. Weintritt, Weintritt Consulting	
A Comparison of NORM Scale Dissolvers	163
Darrell L. Gallup, Unocal Corporation	
Remediation of Normally Occurring Radioactive Material (N.O.R.M.): A Field Trial	179
J.D. Orum, D. Baudoin, AMBAR Laboratories; O. Briscoe, AMBAR, Inc.	

TABLE OF CONTENTS (continued)

	Page
Computer Aided Prediction of NORM Associated Scale	189
A.J. Gerbino, S.J. Saunders, M. Rafal, OLI Systems, Inc.	

SESSION 1

MEASUREMENT AND SURVEY

DETERMINATION OF NATURALLY OCCURRING RADIONUCLIDES IN SAMPLES FROM THE GAS AND OIL INDUSTRY.

Validation of Analytical Methods through an Interlaboratory Test Programme

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SUMMARY

The build-up of Naturally Occurring Radionuclides (NOR's), leading to the production of various types of Naturally Occurring Radioactive Material (NORM), is often encountered during gas and oil production *c.q.* treatment. This gives rise to increased health hazards to personnel and to an environmental waste management problem. Both the NOR's considered to be relevant for E&P samples and their analytical results in terms of activity concentration were found to be strongly dependent on the analytical contractor laboratory selected. This is caused by the lack of standardized procedures for the analysis of E&P NORM. In anticipation to future, more stringent NORM legislation on disposal and associated quantitative risk assessment projects, which may become endorsed by national authorities, the gas and oil industry is helped by quality assured, standardised NORM analysis procedures. Therefore, the identification of relevant NOR's and the validation of their analysis in various types of E&P NORM samples has been investigated at Shell Research Amsterdam by carrying out a literature study and an (dutch) interlaboratory test programme, respectively. The certification of the analytical results has been performed by a foreign (*i.e.* non-Dutch) institute. Only after the implementation of corrective actions as agreed by the participants, standardised analytical methods for E&P NORM samples could be defined. The standardised methods arrived at are described in general terms in this paper.

INTRODUCTION

NORM (Naturally Occurring Radioactive Materials) is a acronym generally used for substances in which Naturally Occurring Radionuclides (NOR's) show up in concentrations above about 0.1 Becquerel per gramme (Bq/g). In the gas and oil industry, the term NORM is commonly used to describe the products and by-products in which the radioactive progeny of the two long-lived parent radionuclides, ^{232}Th and ^{238}U either show up or become enriched due to production or treating processes. During oil and gas production process this progeny may, to some extent, be brought to the surface together with natural gas, with well-fluids like natural gas liquids, crude oil and water, and with solid particles.

As such, NORM will constitute a radiological hazard, *i.e.* delivering a so-called TENR (Technologically Enhanced Natural Radiation) dose to human beings. Two path ways for exposure can be distinguished:

- enhanced *external* radiation fields near and in production and treatment facilities *c.q.* equipment may deliver a TENR dose to (contractor) *personnel*, and
- *internal* contamination of (contractor) *personnel* during maintenance and repair practices (inhalation or ingestion of NORM) and of the *public at large* through uncontrolled disposal of NORM or NOR contaminated equipment into the environment (ingestion of NOR's via human food chain).

Any TENR dose delivered to (contractor) personnel can be minimised by bringing in place Standard Operational Procedures (SOP's)¹, whereas any TENR dose delivered to the public at large has to be controlled by competent national authorities regulating NORM disposal practices. Presently, there is an increasing international environmental concern on the occurrence and build-up of NORM in the Non-Nuclear Industries (NNI). In the Malaysian federation, in various gas/oil producing US states as well as in the European Union (EU), stringent guidelines to control NORM in oil and gas production and in the processing/treating facilities are implemented^{2,3}, or are being drafted^{3,4} and will be implemented into national legislation in the forthcoming years. Besides, a group of international bodies⁵ has issued 'basic radiation safety standards', which in the absence of national radioactive substance legislation may be utilised as NORM guidance.

This type of legislation either addresses NNI NORM specifically, or includes NNI NORM. This is achieved by defining notification, intervention and/or clearance activity concentration levels of the individual NOR's present. Besides, also schemes for compulsory, regular external and internal surveys of E&P facilities and equipment, may be an integral part of this type of legislation. Though activity concentrations forms the heart of the legislation, references to standardized (analytical) methods on sampling, sample pre-treatment and NOR analysis are neither included, nor recommended. To the best of our knowledge no such standardized methods have been issued yet for E&P NORM samples, neither have analytical contractor laboratories been identified who are working according to (quality assurance) certified procedures.

The need for standardization has been clearly experienced in practice. On several occasions it has been observed that results from E&P NORM analysis on pre-treated, homogenised samples from the gas/oil industry by Dutch authorized institutes with experience and expertise in NORM analysis, showed large mutual scatter for various analyses results. In view of the great (economic) impact the use of unreliable working procedures might have for E&P industry; the lack of certified procedures for E&P NORM sampling and analysis; the strongly growing awareness of the need for standardisation in environmental screening matters, and the potential liabilities, it is needless to state that both sampling and analysis methods, which both are reliable, standardized and acceptable to the competent authorities, are a critical need.

In practice, the availability of adequate and proper NORM sampling and analysis methods would allow: 1) to enable accurate determination of the strict radiological levels on-site, 2) to qualify and to process production facility parts as well as secondary streams and waste, 3) to enable reporting to authorities of NORM activity concentrations involved, and 4) to carry out reliable quantitative risk assessments. The availability of these methods will also strengthen the negotiating position of the E&P industry in discussing NORM levels with national authorities.

The importance of sampling, being the first step in the analysis sequence, has not been neglected nor has it been underestimated. In fact, poor sampling degrade all values of subsequent activities as it wastes time and effort thereby requiring repeat samples to confirm uncertain results. Despite its important contribution to the uncertainty of the NORM results, the standardization of NOR analysis of samples stemming from the E&P industry (i.e. gas, hydrocarbon condensate, water, scale and sludge) has been the subject of a research project at Shell Research Amsterdam. Because of the highly specific character of these sample types with respect to the sample composition and the NOR's present therein, and because of the impact of the NOR activity concentration on the measures following from national legislation, standardization of NORM analysis techniques was considered to be essential. An interlaboratory test programme on NORM analysis using pre-treated, homogenised actual samples from the gas and oil production and treatment facilities has been set up, in order to achieve this goal. The way in which the interlaboratory test programme has been conducted, has extensively been described⁵. Next to a short summary, the results obtained will in particular be described in this paper.

⁵ FAO [Food and Agriculture Organisation], International Atomic Energy Agency [IAEA], ILO [International Labour Organisation], NEA/OECD [Nuclear Energy Agency of the Organisation for Economic Cooperation and Development], PAHO [Pan American Health Organisation] and WHO [World Health Organisation].

E&P NORM CHARACTERISTICS

Naturally Occurring Radionuclides (NOR's) which are omnipresent on the planet *earth*, can be subdivided into cosmogenic (e.g. ^7Be , ^{14}C) and primordial (e.g. ^{40}K , ^{87}Rb , ^{138}La , ^{190}Pt , ^{232}Th , ^{238}U) radionuclides. While cosmogenic radionuclides are continuously formed in the outer atmosphere by cosmic irradiation, primordial (or terrestrial) radionuclides have been present ever since the *earth* (life-time 5.5×10^9 y) has been formed.

Uranium (U) and Thorium (Th) are spread throughout *earth's* crust, at an average concentration of 4.2 and 12.5 ppm(w/w)⁶. The naturally occurring U- and Th-isotopes being ^{235}U (natural abundance: 0.72%), ^{238}U (99.28%) and ^{232}Th ($\approx 100\%$), are radioactive. Due to its low isotopic abundance, ^{235}U is rarely encountered in practice and therefore this series will not be further discussed here. Consequently, 'radioactivity' is also wide-spread in the environment, on average 0.05 Bq[^{238}U]/g and 0.05 Bq[^{232}Th]/g.

The primordial radionuclides ^{232}Th and ^{238}U , are special radionuclides, as they each are heading a series of successive nuclear decays (Figs. 1 and 2). Both these NOR's decay through essentially the same elements, all of which are radioactive. As such, these radioactive daughters can be present in gas and oil bearing formations as NOR's, where next to gas and oil also formation water is present in varying amounts.

If a geological formation containing ^{232}Th and ^{238}U has not been disturbed ('closed system') for more than 10^6 years, all members of both decay series will have the same activity concentration (Bq/g) and the series is said to be in *secular equilibrium*. However, as the chemical concentration (mole/g) is proportional to the half-life time ($t_{1/2}$) of a radionuclide, their number of atoms varies considerably. When a formation is not 'closed' to radionuclide migration, e.g. ^{226}Ra becomes mobilised and is 'deposited' somewhere outside the formation, the secular equilibrium will be disturbed. At the site of deposition the only way the ^{226}Ra concentration can stabilise or increase is by influx of (geo)chemical/physical transport phenomena. No ingrowth of ^{226}Ra via radioactive decay of its ancestors will occur as all radionuclides above ^{226}Ra (cf. Fig. 2) will stay close ^{238}U . In this way at the site of deposition ^{226}Ra is *unsupported*, i.e. ingrowth is not supported by radioactive decay.

The (geo)chemical/physical properties of all radium isotopes, i.e. ^{226}Ra , ^{228}Ra and ^{224}Ra , are identical. Differences between these isotopes are caused by their radiation decay characteristics and hence by the associated potential radiation hazards. A NOR should 'live' long enough to develop the (geo)chemical properties of the element. In gas and oil production two typical time scales are important, a *geological* (say, far longer than 1,000 years) and a *production time scale* (order of magnitude: days to decades). Therefore, for processes taking place on a time scale of one day, only isotopes of thorium, radium, lead, uranium, radon and polonium may become unsupported. From considerations with respect to the formation of hydrocarbons from source rock under reducing conditions⁷, from considerations with respect to geochemical transport phenomena⁸ and from an extensive literature survey⁹, only a limited number of NOR's actually may appear unsupported in E&P (by-)product streams.

The survey showed that both ^{238}U (and its immediate daughters: ^{234}Th , $^{234\text{m}}\text{Pa}$, ^{234}U and ^{230}Th) and ^{232}Th were only present in minute quantities (≈ 0.05 Bq/g), which is in agreement with transport phenomena under reservoir conditions. Consequently in E&P samples the NOR's mentioned do not constitute a radiological hazard.

Furthermore, it was observed that radium isotopes actually mark a clear separation line for the encounter of NORM in gas/oil production, treating or processing facilities. The most likely cause for this separation is a distinct difference in geochemical properties (mobility under reservoir conditions) in combination with the effective^b half-life of ^{238}U and ^{232}Th on one hand, and that of ^{226}Ra and ^{228}Ra and their progeny

^b The effective half-life of a radionuclide under reservoir conditions is defined as a linear combination of its physical half-life ($t_{1/2}$) and its 'mean residence time' in a phase during production from the reservoir or even during maturation, migration or accumulation processes in the subsurface.

on the other hand. As in the ^{232}Th series in between ^{228}Ra and ^{224}Ra , rather long-lived (with respect to a production time scale) and (geo)chemically immobile ^{226}Th is encountered, ^{228}Ra and ^{224}Ra are normally not in secular equilibrium, and hence for radiological assessment studies, production water and production water associated deposits have to be analyzed both for ^{228}Ra and ^{224}Ra .

For relatively dry natural gas wells, ^{222}Rn forms a second distinct separation line for the potential encounter of NORM. Most likely the ^{222}Rn stems from (reservoir) rock near the perforation. Very often it is observed that with increasing depletion of gas reservoir due to production also increasing ^{222}Rn concentrations are found.

From the survey it was also observed that also ^{210}Pb may mark an independent separation line in the ^{238}U decay series, as this radionuclide can be encountered unsupported. When carried with the water phase, this is likely caused by its specific (geo)chemical properties, but also indications exists that ^{210}Pb is transported through the gas phase¹⁰.

Only one case of unsupported (highly radiotoxic) ^{210}Po in hydrocarbon condensate has been reported, notwithstanding the fact that one should be aware that this radionuclide may be encountered.

From the geochemical considerations and observations done during the survey, E&P samples should be analyzed for NOR's according the scheme presented in table 1. These types of analysis were requested during the interlaboratory test.

Table 1: NOR's encountered in E&P samples

E&P (by-)product stream	^{238}U progeny	^{232}Th progeny
Natural Gas - Natural Gas Liquids	^{222}Rn ^{222}Rn , ^{210}Pb , ^{210}Po	none none
Crude Oil	none	none
Production water - hard scale - medium hard to soft scale - scrapings - sludge	^{226}Ra , ^{210}Pb ^{226}Ra ^{226}Ra , ^{210}Pb ^{226}Ra , ^{210}Pb ^{226}Ra , ^{210}Pb	^{226}Ra , ^{224}Ra ^{226}Ra , ^{224}Ra ^{226}Ra , ^{224}Ra ^{226}Ra , ^{224}Ra ^{226}Ra , ^{224}Ra

INTERLABORATORY TEST PROGRAMME

The interlaboratory test programme, with participation of eleven Dutch institutes that are known to have expertise in NORM analysis, has been described in detail elsewhere⁵. The test programme consisting of four stages and was conducted over a period of three years. The actual oil and gas production samples varied from relatively simple single phase (e.g. methane gas, hydrocarbon condensate, production water and scale) to complex multi-phase samples (e.g. various sludge samples). After a pretreatment step to achieve homogeneity and a check via repeatability measurements, the samples were despatched to the participants. After 3 months results were collected and subjected to statistical evaluation and feedback was given to the institutes via a review meeting (i.e. discussion of the results, exchanging analytical expertise, agreeing on measures to be taken and on samples for the next stage).

Prior to the despatch, the homogeneity of each sample was determined at Shell Research Amsterdam by repeatability measurements using γ -spectrometry, except for (i) the condensate, where radiochemical ^{210}Po analysis (*vide infra*) was applied, and (ii) the methane gas where the ^{222}Rn analysis via the α -scintillation method (*vide infra*) was used. With reference to the estimated counting statistics error, the repeatability was less than 5% at 95% confidence level (2σ), while, the more laborious radiochemical technique (^{210}Po) did show a somewhat larger repeatability.

Except for the first stage of the test programme, in which results were compared to those obtained by Shell Research Amsterdam, the activity concentrations of γ -emitting NOR's in the samples were certified by the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig, Germany. However, PTB could not certify the α -activity concentration of ^{222}Rn in methane gas and that of ^{210}Po in the other samples. Therefore, to enable mutual comparison of the ^{210}Po results, an aliquot of an aqueous ^{210}Po standard has been analyzed by the institutes in stage four of the test programme.

ANALYSIS TECHNIQUES

The full radiometric analysis of E&P NORM samples generally requires at least three techniques for a complete determination of the activity concentrations of the various NOR's:

Lucas cell α -scintillation method for the determination of ^{222}Rn in natural gas

The inner surface of a cylindrical Lucas cell (± 250 mL) is coated with a high-efficiency α -scintillator material (e.g. zinc sulphide). Natural gas is introduced in the cell, which will be light-tight connected to a photomultiplier tube. After about two hours ^{222}Rn will be in secular equilibrium with its short-lived progeny, particularly its α -emitting daughters ^{218}Po and ^{214}Po (cf. Fig. 2). The ^{222}Rn concentration is computed from the number of detected scintillations (efficiency calibration required) due to α -decay. This measurement technique is fast, simple and easy to operate. Analytical results will be available at the same day.

γ -spectrometry for the determination of ^{226}Ra , ^{210}Pb , ^{228}Ra and ^{224}Ra in various types of E&P samples

A γ -spectrometer consists of a high efficiency, high energy resolution ($\Delta E/E < 0.2\%$), (non-)doped germanium detector, which is placed in a low background radiation area obtained by shielding with (old) lead bricks. In general E&P samples can be measured as received, but in some cases sample pre-treatment (e.g. for sludge the sample is separated into a liquid and solids part) is required. The sample is placed in a standard size, air-tight container on top of the detector and the current pulses in the biased semi-conducting germanium detector are registered. Spectrometer efficiency calibrations (including corrections for true coincidence) are performed using NORM reference materials. Both the quick analysis (results after about two days) and the accurate analysis (about one month) require expert knowledge and can not be carried out by a layman.

- The ^{226}Ra concentration can be computed accurately from the registered γ -emission intensity of its indirect daughter isotopes ^{214}Bi and ^{214}Pb . Therefore, an air-tight sample container has to be filled and stored in order to establish secular equilibrium between the isotopes, which is reached after about three to four weeks.
However, because of the absence of ^{235}U (a.o. γ -emission line at 185 keV), a quick and somewhat less accurate analysis can be performed in about one day by direct measurement of the 186 keV ^{226}Ra γ -emission line.
- The low γ -energy 46.5 keV line of ^{210}Pb can be determined only if a sensitive germanium detector is available. E&P NORM samples often have a relatively low activity concentration and a high density. As a result, γ -attenuation in the sample will occur, which has to be taken into account during calibration of the γ -spectrometer system. A correction for self-absorption of ^{210}Pb and/or other γ -photons of low energy has been made (e.g. via γ -transmission measurement¹¹ or via computed efficiencies from effective solid angles¹²). Correction factors between 1.5 and 2.5 are not exceptional, particularly when high Z elements (e.g. Ba, Pb) are present.
- After establishing secular equilibrium (in about one day) with its immediate daughter ^{228}Ac , the ^{226}Ra concentration can be computed from a selection of the most intense ^{228}Ac γ -lines.
- After establishing secular equilibrium (in about two days) with its indirect daughters ^{212}Pb , ^{212}Bi and/or ^{208}Tl , the ^{224}Ra concentration is computed from a selection of their most intense γ -lines present (cf. Appendix).

α-spectrometry for the determination of ²¹⁰Po in various kinds of E&P samples

An α-spectrometer is an evacuated (to enhance the mean free path of α-particles) chamber, in which a silver disc with the deposited α-emitter is placed opposite an α-energy discriminative detector. A fluid (water reduced by evaporation; *hydrocarbon condensate* acid extracted and subsequent reduction by evaporation) E&P sample is spiked with ²⁰⁸Po, while a solid E&P samples is acid digested and the obtained solution is spiked with ²⁰⁸Po and reduced by evaporation. Both ²¹⁰Po and ²⁰⁸Po (internal calibration of deposition efficiency) are deposited onto the silver disc. The method is very labour intensive and requires expert knowledge. It has a low detection limit, while results may be available after about one day.

Details on the methods applied, such as detection limits, accuracy, preparation and measuring time, are listed in Table 2:

Table 2: Characteristics of the NORM analysis methods applied.

NORM Analysis Method	Type of sample material	Detection limits for each NOR	Accuracy (2σ at 95% conf. level)	Preparation time (hours)	Measuring time (hours)
γ-spectrometry ²²⁶ Ra, ²¹⁰ Pb, ²²⁸ Ra, ²²⁴ Ra	Solid	0.02 Bq/g	10%	0.25 to 2.0	15
	Liquid	0.01 Bq/g	10%	0.25 to 0.5	15
α-spectrometry ²¹⁰ Po	Aqueous solutions	4×10 ⁻⁶ Bq/g	10%	3	15
	Solid	4×10 ⁻³ Bq/g	10%	5	15
	Condensates	0.5×10 ⁻³ Bq/g	10%	3	15
α-scintill. cell ²²² Rn	Natural gas	0.01 Bq/L (STP)	10%	0.25 to 0.3	3 to 5

RESULTS AND CONCLUSIONS OF THE TEST PROGRAMME

During the interlaboratory test programme, the need for standardization of E&P NORM analyses on samples from the gas and oil industry, which are highly specific (in terms of NOR's present, required (pre)treatment techniques, sample composition, etc.), was clearly demonstrated. After each stage, a steady improvement in the reproducibility and accuracy of the analytical results was shown which may partly be attributed to the open character of exchanging analytical expertise during the review in meetings. As such, the participation in the test programme on NORM analysis proved to be beneficial to all parties involved.

With few exceptions only, the NORM analyses methods used at the Dutch institutes were proven to be reliable and accurate. Next to locally standardized methods at several institutes, those used by Shell Research Amsterdam were proven to be reliable and accurate throughout the test, and were found to be a sound basis for broader standardization of E&P NORM analyses.

As described above only three distinct analytical methods are required to fully characterize E&P NORM samples, thereby yielding complete accurate and reliable results (Table 2).

- a) α-scintillation counting using a Lucas cell for the determination of ²²²Rn in natural gas.
- b) γ-spectrometry for determination of ²²⁶Ra, ²¹⁰Pb, ²²⁸Ra and ²²⁴Ra (γ-emitting NOR's) in the solid and liquid E&P NORM samples. Compared to radiochemical methods, this technique is fast, precise, accurate and yields a multi-nuclide analysis.
- c) acid digestion of the sample spiked with ²⁰⁸Po standard, followed by spontaneous deposition of polonium on silver, followed by α-spectrometry to measure the ²¹⁰Po concentration in the solid and liquid NORM samples.

The critical parameters in the application of these methods were:

- ad a) the use of methane gas for counting cell efficiency calibration, by purging methane gas through a standard ^{226}Ra solution in secular equilibrium with ^{222}Rn . It is essential that the density of the purging gas should correspond to that of the sample to be measured, as the counting efficiency of the cell increases with the decreasing gas density inside the chamber¹³. In case nitrogen is used as purging gas, the ^{222}Rn counting cell efficiency for natural gas will be too low by some 7%. Besides, ^{226}Ra calibration source available from manufacturers, from which ^{222}Rn is purged by air, is only suited for this purpose, if appropriate corrections for purge gas density are made.
- ad b) an accurate correction for self-absorption, in view of the low energy γ -emission of ^{210}Pb at 46.5 keV and the heavy matrix of the majority of the samples analyzed. A simple and adequate correction technique for self-absorption is based on γ -transmission measurements¹¹. This technique only requires the attenuated and unattenuated beam intensity of a ^{210}Pb point source to correct for self-absorption due the sample matrix at the 46.5 keV γ -line.
- a uniform NOR's γ -yield data set, based on recent insight/investigations in order to obtain reproducible/reliable results for the complex sludge samples. An inventory of the NOR ' γ -yields' applied by the institutes to calculate activity concentration, showed that amongst them values varying up to 30% were used. These γ -yields are often included with the commercially available data-acquisition and data-analysis software packages, whereas more recent literature values are also used. To exclude errors in results due to variation in ' γ -yields', it was agreed to use a uniform set of recently revised ' γ -yield' data^{14,15,16} for all γ -spectrometry results reported (cf. Appendix), after which a clear improvement in accuracy of the results has been observed.
- ad c) a well-calibrated ^{208}Po spike solution, for the ^{210}Po measurement. The accuracy of the ^{210}Po activity concentration analysis has been checked using a commercially available, aqueous ^{210}Po standard. Trends observed in the analytical results produced by the participants disappeared when corrected with the analytical results obtained for an aliquot of a commercially available ^{210}Po standard. Therefore, it could be concluded that the accuracy of the results is affected by that of the ^{208}Po standard rather than by sample digestion procedure and the α -spectrometry techniques.

The observed improvement in accuracy during the four stages of the test programme were shown to be independent of the applied instrumentation and data-analysis software packages. The improvement in the γ -spectrometric results averaged per stage, and normalized to PTB or Shell Research Amsterdam reference values is shown in Figure 3. With reference to a range set by the average PTB result $\pm 10\%$, a gradual improvement in the results may be observed in comparison with each preceding stage, which is noteworthy, especially for the complex sludge samples in Stage 3. For the ^{226}Ra results sludge 1 of Stage 4, it should be noted that a general trend has been observed, indicating that the PTB result might be inaccurate. For the complex sludge samples of the final Stage 4, all results reported are within $\pm 10\%$ of those reported by PTB, except for two institutes (04 and 09).

Due to lack of standardized and (internationally) approved on-site NORM sampling techniques for products, by-products and waste materials from gas and oil production and treatment industries, this item was not included in this study. It may, however, still be a major cause of discrepancies in the results from NORM analysis, and therefore is currently the subject of future research.

CONCLUSIONS

The oil/gas industry should be well aware that through the build-up of unsupported NOR's, contamination of the decay products of ^{238}U and ^{232}Th may occur in its streams, (by)products and (part of) its installations;

- E&P samples do generally not include the 'parents' ^{238}U and ^{232}Th at levels constituting a radiological risk.
- Radium is the first 'daughter' element encountered, ^{226}Ra and ^{228}Ra respectively.
- E&P samples may contain unsupported NOR's, ^{226}Ra , ^{222}Rn , ^{210}Pb and ^{210}Po (^{238}U -series) and ^{228}Ra and ^{224}Ra (^{232}Th -series).

Three distinct NOR's analysis techniques are required to perform a full radiometric analysis of E&P NORM samples:

- for natural gas samples: Lucas cell α -scintillation method (^{222}Rn).
- for solid and fluid samples: γ -spectrometry (^{226}Ra , ^{210}Pb , ^{228}Ra and ^{224}Ra) & α -spectrometry (^{210}Po).

From the interlaboratory test programme, the following items may introduce large errors in E&P NORM analysis results:

- Lucas cell α -scintillation method: the choice of the purge gas for the efficiency calibration of the cell.
- γ -spectrometry: the correction technique used for self-absorption correction in case of ^{210}Pb , and the selection of ' γ -yield' data for conversion to other NOR specific activity concentrations;
- α -spectrometry: the accuracy of the ^{209}Po 'spike' used for ^{210}Po analysis, and

In view of the importance of the findings to all institutes as a first step to standardisation, the results of the interlaboratory test programme on NORM analysis have been published in a joint multi-institute co-authorship⁵. As such, these NORM analysis methods will not be prescribed in a strict manner to the operating companies, but the various attention points therein which may lead to erroneous results have clearly been indicated.

Finally, also the standardisation of methods for E&P NORM sampling is, as far as possible, is a critical need. A generic standardisation for E&P sampling is currently the subject of a currently ongoing research program at Shell Research Amsterdam.

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Series	Sub-series	In secular equilibrium with	γ -emitting isotope	γ -photon energy (keV)	γ -photon yield (%)
^{232}Th	^{228}Ra	^{228}Ac	^{228}Ac	338.4	11.50
				911.1	25.30
				969.1	15.40
	^{228}Th (^{224}Ra)	$^{224}\text{Ra}, ^{220}\text{Rn}, ^{216}\text{Po},$ $^{212}\text{Pb}, ^{212}\text{Bi},$ $^{212}\text{Po}/^{208}\text{Tl}$	^{212}Pb	238.6	43.10
				300.0	3.34
			^{212}Bi	727.3	6.93
			^{208}Tl	583.1	29.40
^{238}U	^{234}U	$^{234}\text{Th}, ^{234\text{m}}\text{Pa}, ^{234}\text{U},$ ^{230}Th	^{234}Th	63.3	3.81
				92.8	5.57
			$^{234\text{m}}\text{Pa}$	1001.0	0.79
	^{226}Ra	$^{222}\text{Rn}, ^{218}\text{Po}, ^{214}\text{Pb},$ $^{214}\text{Bi}, ^{214}\text{Po}$	^{230}Th	67.7	0.37
			^{226}Ra	186.1	3.51
				258.8	0.55
				295.2	19.30
				351.9	37.60
				480.4	0.32
			^{214}Bi	389.1	0.42
				609.3	46.09
				665.5	1.56
				719.9	0.38
				768.4	4.94
				806.2	1.22
				934.1	3.03
				1120.3	15.10
				1155.2	1.63
				1238.1	5.79
				1281.0	1.43
				1377.8	4.00
				1385.3	0.78
				1401.5	1.27
				1408.0	2.15
				1509.2	2.11
				1661.3	1.15
				1729.6	2.92
				1764.5	15.40
				1847.5	2.12
				2118.5	1.21
				2204.0	4.99
	^{210}Pb	^{210}Bi (^{210}Po)	^{210}Pb	46.5	4.00

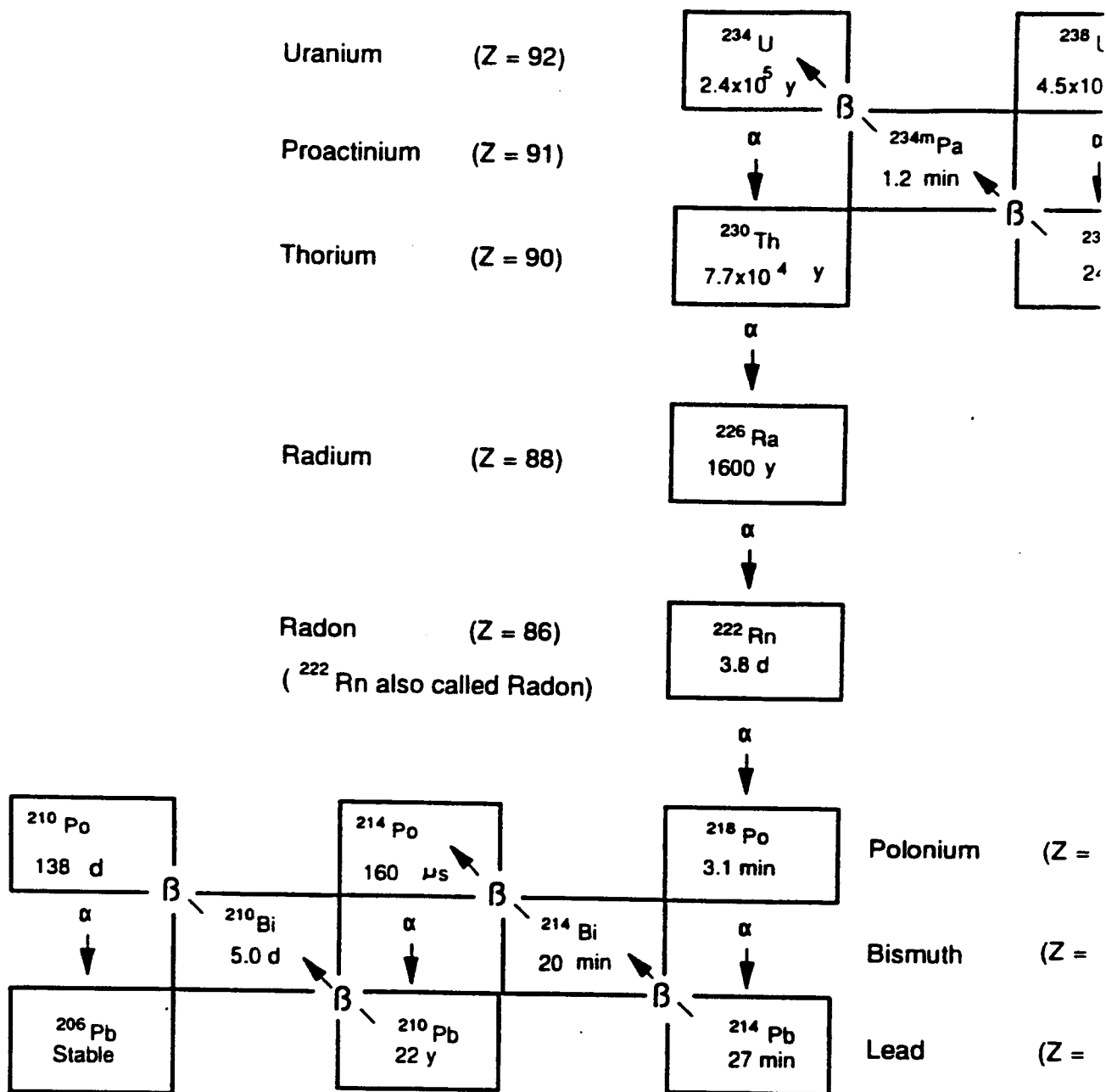


Figure 1
Principal decay scheme of ^{238}U . The chemical symbol (or atomic number Z) reflects the chemical properties, the 'superscript' isotopic number (or atomic mass) reflects the radiation properties.
 α -decay lowers the atomic number with 2 and atomic mass with 4.
 β -decay increases the atomic number with 1, while the atomic mass remains the same.

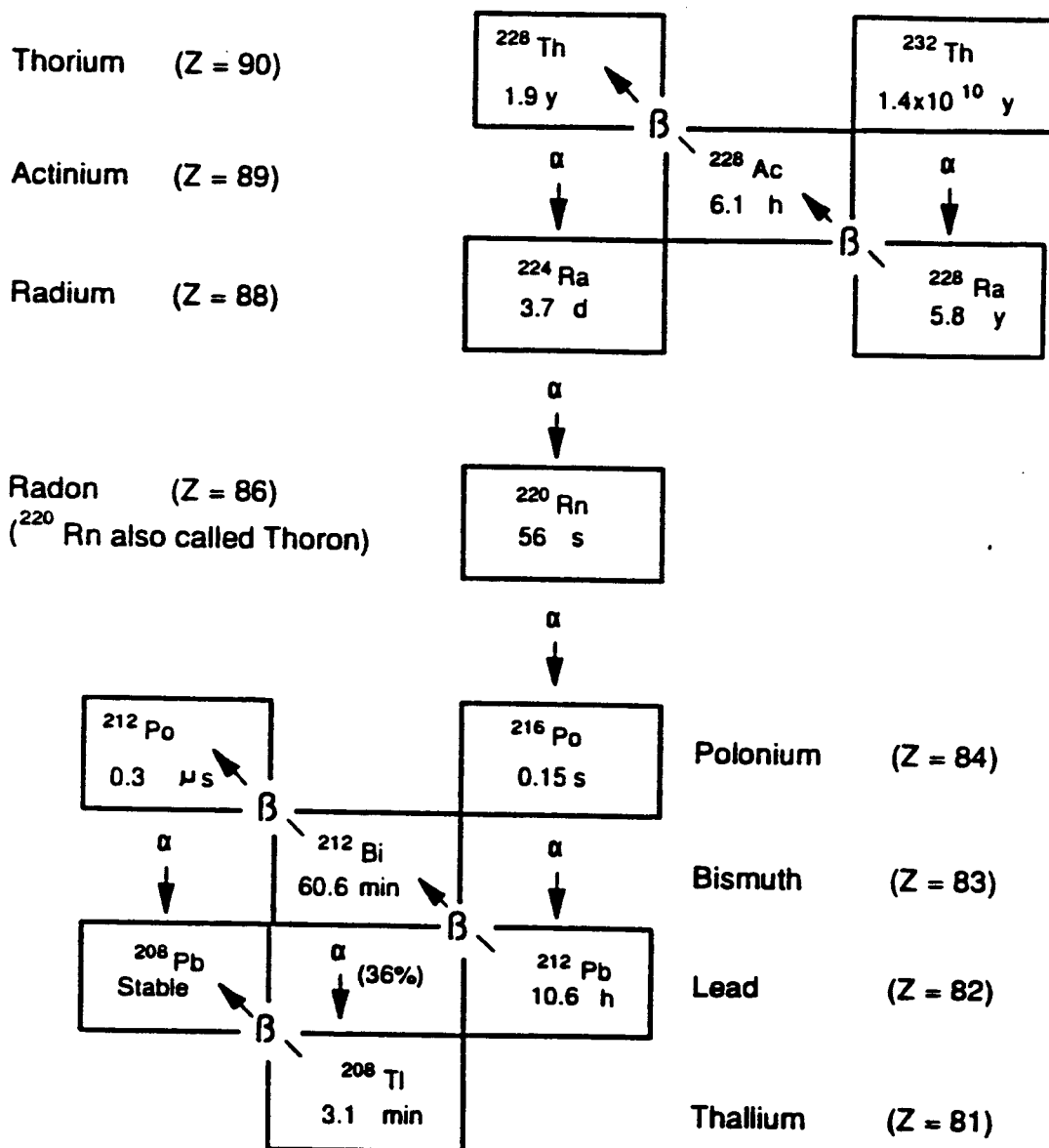
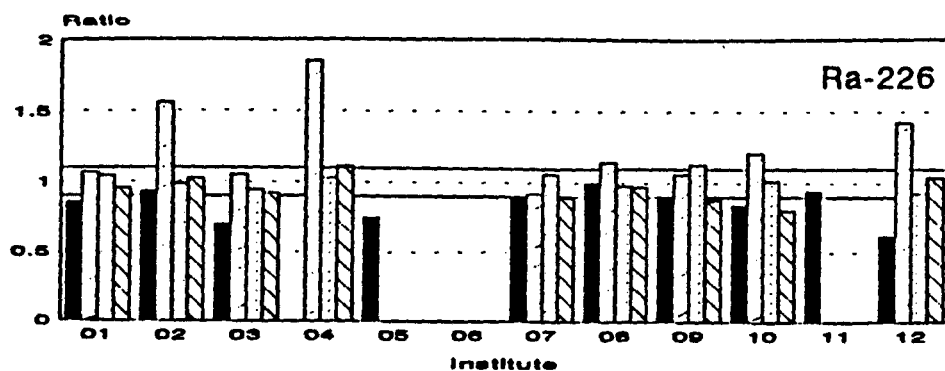


Figure 2

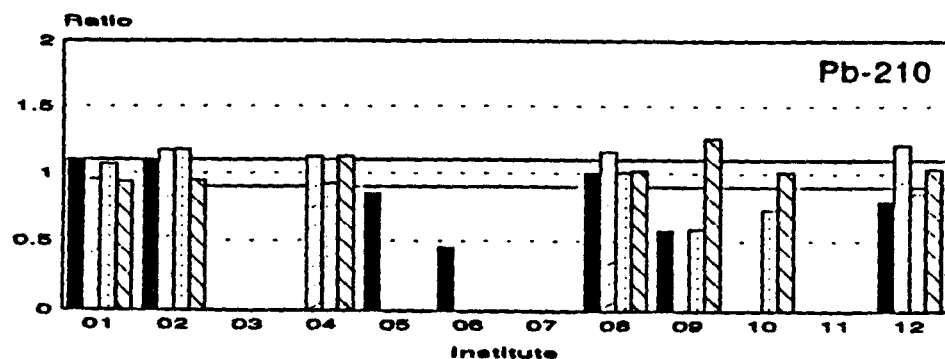
Principal decay scheme of ^{232}Th . The chemical symbol (or atomic number Z) reflects the chemical properties, the 'superscript' isotopic number (or atomic mass) reflects the radiation properties.

α -decay lowers the atomic number with 2 and atomic mass with 4.

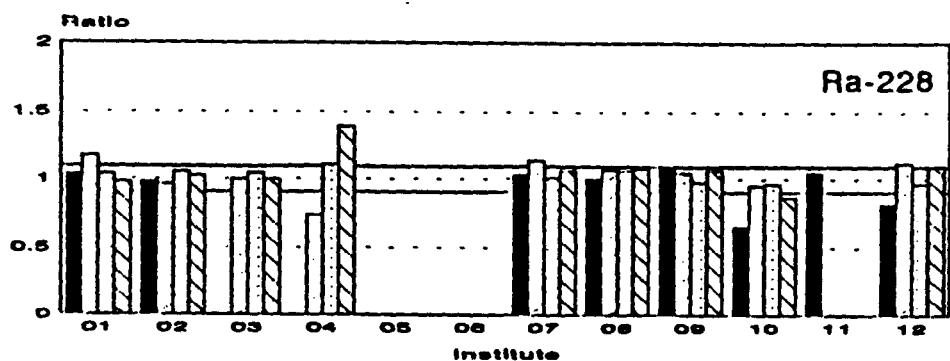
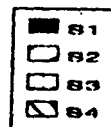
β -decay increases the atomic number with 1, while the atomic mass remains the same.



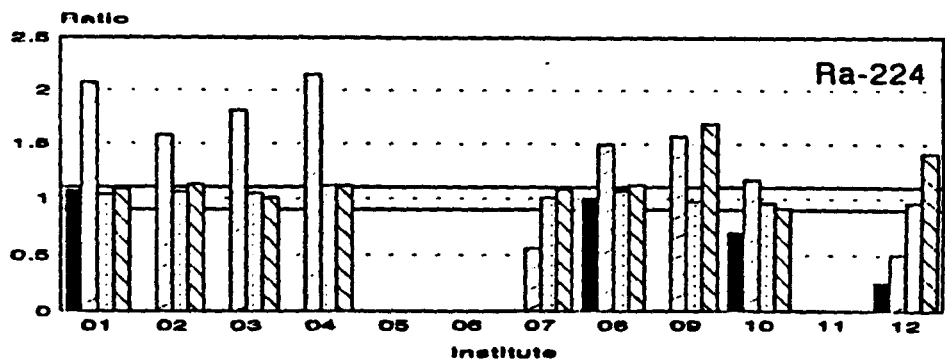
(a)



(b)



(c)



(d)

Figure 3

Accuracy improvement trends of averaged NOR γ -spectrometric results for each stage (S1 to S4) of the interlaboratory test programme. Results as well as a 10% error range are shown, relative to the results of the reference institute (i.e. PTB).

DETECTING NORM CONTAMINATED TUBING DURING PULLING OPERATIONS

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ABSTRACT

A system has been developed and field tested that can detect NORM contaminated tubing as it is pulled from a well. Early, automatic identification of contaminated tubing can reduce the manpower required to manually survey all tubing for contamination. Early detection will also allow field personnel to separate the contaminated tubing and handle it as needed to prevent the spread of contamination.

The system is composed of a sodium iodide detector and an alarming rate meter that can be mounted remotely. The alarm level is adjustable so it can be set to the desired level for different tube sizes, installation geometries, and background levels.

INTRODUCTION

Oil well tubing contaminated with NORM (Naturally Occurring Radioactive Material) is found in many producing fields. To reduce the manpower requirements needed to identify contaminated tubing, we have developed and tested a system to identify contaminated tubing as it is pulled from a well.

To reduce the cost of development and reduce the final cost to users, we tested components of several commercial instruments. We laboratory tested several systems and selected two of them for a field test. One of the systems selected for a field test was mounted in an explosion proof box such as may be required in hazardous locations.

Field tests have been held in west Texas and near offshore Louisiana. The field tested systems consisted of a 3×3 inch sodium iodide crystal detector and an alarming ratemeter. The systems were able to detect the contaminated tubing above 50 μ R/hr.

Systems such as the ones tested can be packaged for use in oil fields and mounted on workover rigs with the alarm positioned near the rig operator. The systems could be combined with an automatic marking system that would mark any tube section that caused the system to alarm.

DISCUSSION

Several manufacturers have developed instruments to detect various types of radiation under many conditions. For the needs of this project, we decided a sodium iodide crystal detector was the best detector because of its high detection efficiency and relatively low cost. Among the items tested were a *Gate Frisker* and an *Alarming Ratemeter* coupled with three different sodium iodide (Nal) detectors.

The systems originally tested were not designed for use in hazardous locations or for use in bad weather. Based on successful field tests of two of those systems, a system designed for use in Class I, Division 1, Group D hazardous locations was tested offshore Louisiana.

The system field tested in Louisiana consisted of an enclosure that contained a shielded, 3×3 inch, sodium iodide crystal detector. This enclosure was connected to an alarming rate meter by a fifty foot long cable. All signals in the cable were "intrinsically safe" so the cable

DETECTING NORM CONTAMINATED TUBING DURING PULLING OPERATIONS



Figure 1 Detector in Explosion Proof Enclosure

could be run in hazardous locations. The alarming rate meter was placed in a weather tight enclosure that was not rated for hazardous locations. With minor changes, the entire system could be designed for operation in hazardous locations.

The rate meter counts the gamma rays detected by the detector and displays the count rate on an analog meter. A 4 position switch can be adjusted to change the full scale value. For the second field test, switches were mounted on the outside of the enclosure to allow the:

1. Alarm level to be displayed,
2. Alarm level to be adjusted,
3. Alarm to be reset, and
4. System to be turned on/off.

These systems tested were designed to operate on internal batteries or on standard 120 volt power. The batteries can power the system for over 24 hours.

Laboratory Tests

Laboratory tests were performed at Texaco EPTD's Bellaire, Texas facility using several different instruments to determine if it is feasible to use commercially available instruments to detect NORM contaminated tubing as it is pulled from a well. The tests indicated it is feasible, however some changes will be required to optimize them for oil field applications.

We tested several different sodium iodide crystal

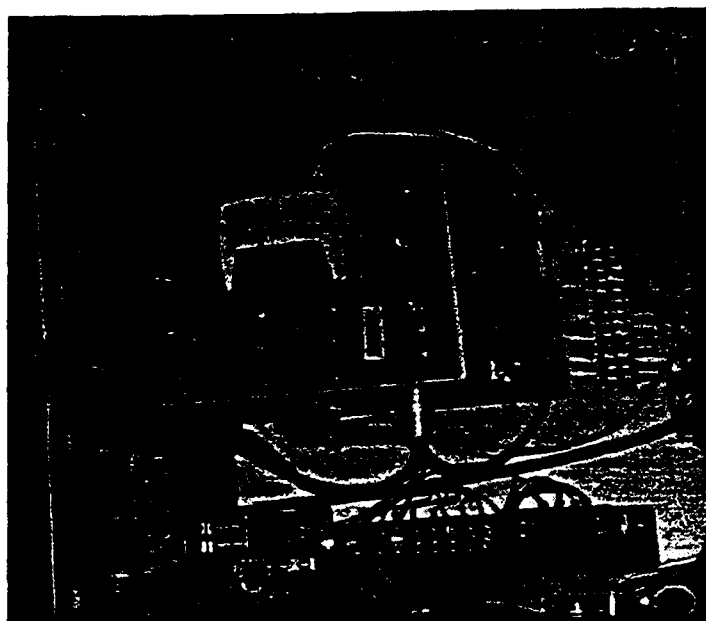


Figure 2 Ratemeter and Enclosure

detectors and an alarming ratemeter. The alarming ratemeter used had a pulse output that was connected to a computer controlled multichannel scaler. Thus the count rate as a function of time could be recorded for later analysis.

For the tests, a small radium calibration source was placed inside a three inch, schedule 40, steel pipe. The source caused a maximum dose rate of 25 μ R/hr at the surface of the pipe. A physically small and relatively weak source was picked as a "worst case." Distributed sources will cause a stronger detector response.

The detector of the instrument under test was placed a measured distance from the center line of the pipe. The pipe was lowered and then raised passed the detector under test. The counts in the detector were recorded. The distance from the detector to the pipe was changed and the test repeated.

Tests to simulate different pulling speeds were also performed. Pulling speed of 1000 to 5000 ft/hr were simulated in 1000 ft/hr increments.

Tests were performed using 2x2 inch and 3x3 inch sodium iodide detectors. One of the 3x3 inch detectors was shielded to reduce background count rate. The other detectors were unshielded. Tests were also performed using a one inch thick plate of aluminum to simulate the presence of an explosion proof housing. The aluminum plate reduced the counts in the detector, but the 3x3

inch detectors were able to detect the presence of the radioactive source.

The source was kept in the pipe by a cap that was clamped onto the pipe. On some data runs, the 2×2 detector could not detect the radioactive source. The instances of non detection were probably caused by the pipe rotating into a position that placed the clamp hinge between the source and the detector. This additional thickness of steel would significantly reduce the detectable counts from the source. Based on these tests, it was determined that a 2×2 detector could not reliably detect low levels of NORM contamination under typical field conditions.

Figure 3 shows the results from a 3×3 inch detector for three different distances between the source and detector at 5000 ft/hr. The counts are in one second intervals. The data from the pass when the source is nearest the detector shows the most counts. With this detector at any of these three source to detector spacings, the system would alarm if the alarm level was set to $1\frac{1}{3}$ the average background count rate.

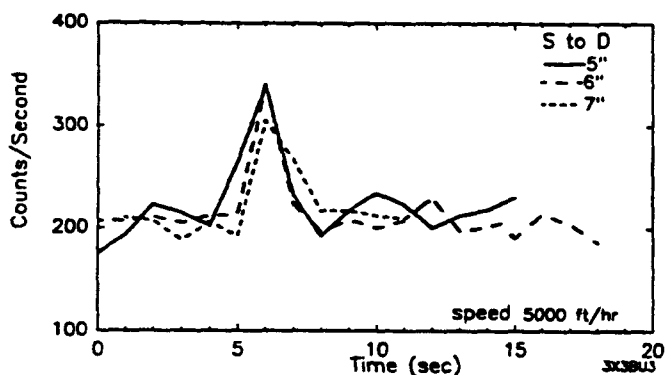


Figure 3 3×3 Nal Detector at 5000 ft/hr.

Different pulling speeds also effect the detector response. This effect can be seen in Figure 4. The slower speeds cause a higher response and a wider peak. This effect will probably not be as pronounced with a distributed source such as will be usually seen in the field because with contamination spread over several feet of tubing, the contaminated area would be detectable for several seconds.

For the shielded detector, the response is similar. The average background count rate is only about $\frac{1}{4}$ of the background count rate of the unshielded 3×3 inch detector and about $\frac{1}{2}$ the unshielded 2×2 inch detector. The lower background makes it easier to detect low levels

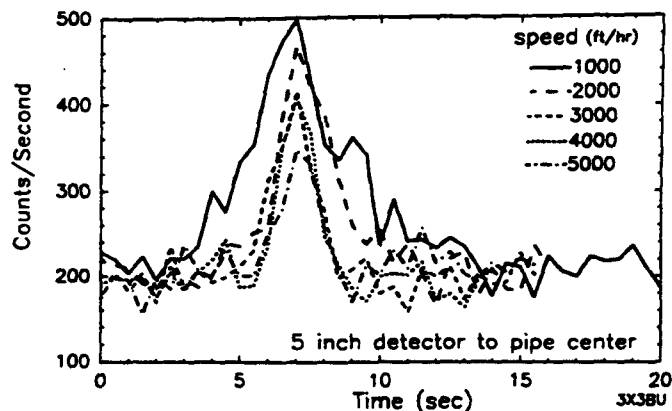


Figure 5 3×3 Inch Unshielded Detector at Various Pulling Speeds

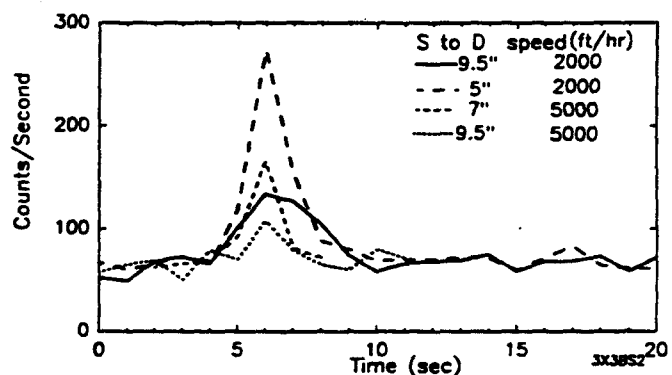


Figure 4 3×3 Nal Shielded detector results.

of NORM contamination. The results of some measurements with a shielded detector are shown in Figure 5. This plot shows different spacings and different pulling speeds. The closer the detector is to the source and the slower the pulling rate, the higher the response in the detector.

As a result of the laboratory tests, the 3×3 inch sodium iodide detector was selected as the detector to be used in the field test.

Field Tests

Two successful field tests of the NORM pipe detector have been performed. The first one was in west Texas and the second offshore Louisiana. For the west Texas test, contaminated 2-1/2 inch tubing was brought in from another well.

For the field test, a 16 inch long spool piece of 6 inch casing was installed between the BOP's and the pipe slips. Openings were cut in the sides of the spool pieces

DETECTING NORM CONTAMINATED TUBING DURING PULLING OPERATIONS

to improve detection efficiency. The enclosures containing the detectors were clamped onto the spool piece flange. An improved method of mounting in locations that do not require equipment to rated for hazardous atmospheres may be to weld a "mounting tube" onto the side of the spool piece or to mount it in the branch of a tee. The mounting tube should have an ID sufficient to accept the detector with some padding to reduce detector vibration. The end of the mounting tube should be made of 1/4 inch or thinner steel to keep source attenuation to a minimum.

For the first field test, one shielded, 3×3 inch, sodium iodide crystal detector was mounted in a NEMA 12 enclosure and connected to an alarming ratemeter. Output from the ratemeter was connected to a multichannel scaler data acquisition card in a portable computer. The detector was mounted about 5 inches from the center of the tubing.

A second shielded 3×3 inch sodium iodide crystal detector was mounted in an explosion proof box such as may be required in hazardous areas. The detector in this box was mounted about 7-1/2 inches from the center of the tubing.

The contaminated tubing had various levels of contamination along its length. One spot near the bottom of the tubing measured 200 $\mu\text{R/hr}$. Other sections of the pipe had less contamination. Figure 6 is a plot of some field test data. The first peak shows the detector counts as the contaminated tube was slowly pulled from the well.

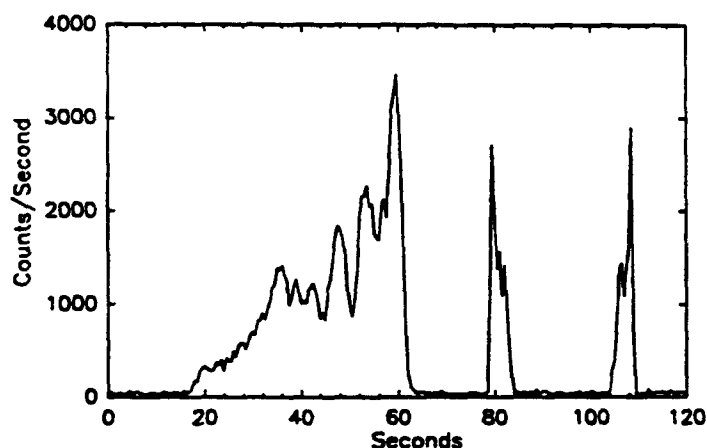


Figure 6 Field Test Detector Response

The slow pulling rate ($\approx 4,000$ ft/hr) allows a profile of the contamination to be easily viewed. The shape of the

peak shows that the contamination about 5 feet from the top of the tube is about 1/3 what it is near the bottom. If we assume that 200 $\mu\text{R/hr}$ causes 3000 counts/sec in the detector and the response is linear then a response of 750 counts/sec will be caused by a field of 50 $\mu\text{R/hr}$. That count rate is well above the background count rate of this detector. Thus, the alarm level could be adjusted to detect contamination levels ≤ 50 $\mu\text{R/hr}$ with little chance of false alarm.

The second and third peaks show the tube being rapidly lowered into and then pulled from the well. These peaks have the expected shapes when compared to the first peak. The faster pulling rate ($\approx 19,000$ ft/hr) data indicates that even though the maximum count rate was reduced, the count rate is still significantly above background.

A field test of a system rated for use in hazardous atmospheres was performed in near offshore Louisiana in August 1995.

The system was assembled, tested, and the background count rate recorded. A comparison of the background count rates on the workover barge and in the laboratory is shown in Figure 7. The average laboratory background rate is about 85 counts/second. The background rate on the barge averaged about 18 counts/second. This reduction in count rate is expected since most background counts are due to small amounts of radioactive material in the soil.

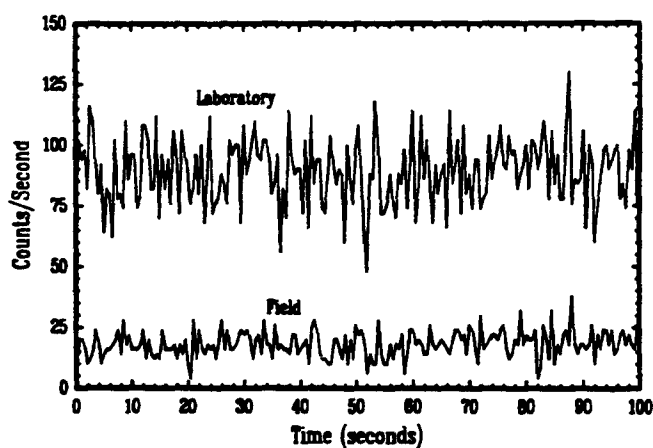


Figure 7 Background Count Rate

For the field test, the explosion proof enclosure containing the detector was installed on the bell nipple above the BOP's. Figure 8 shows the installation of the detector system. The enclosure was installed as close to the

nipple as possible. This placed the detector about 8 inches from the center of the tubing being pulled.

The closer the detector is placed to the tubing and the less material there is between the detector and the tubing being pulled, the more sensitive the system will be to low levels of radioactive contamination.

In this installation, the bell nipple was filled with brine. The detector would have been more sensitive if the detector was installed on the bell nipple above the brine.



Figure 8 Response from Gas Lift Valve

This well was selected for the tests because:

1. This field has a history of NORM problems and
2. This well had produced a large amount of water since it was last worked over.

Thus, the tubing in this well was expected to be contaminated with NORM.

During pulling operations, the packer refused to release from the bottom of the tubing string. Thus, the pulling rate was limited to 2400 ft/hr to prevent significant reduction of the hydrostatic head in the well and possibly cause gas incursion into the well.

No NORM contamination of the tubing was detected. However, NORM contamination was detected in the bottom four gas lift valves. The contamination in each valve caused a reading of less than 30 $\mu\text{R/hr}$. This level was detectable with this system even though it is less than 20 $\mu\text{R/hr}$ above background.

The contaminated gas lift valves were detected because I was watching the computer display during pulling

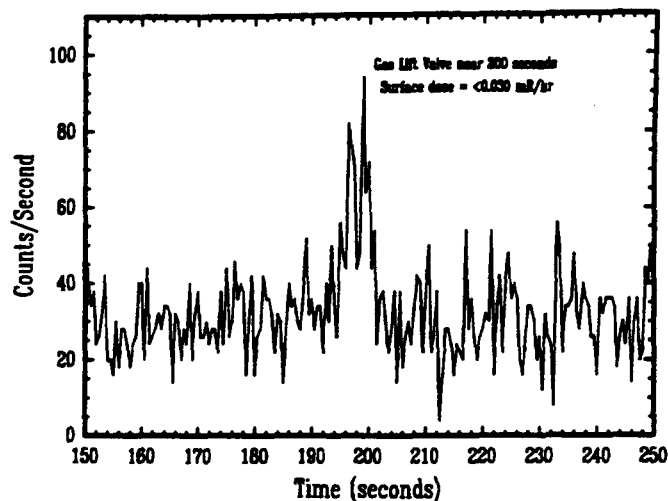


Figure 9 Response from Gas Lift Valve

operations. I am not sure this low level of contamination would have been detected if the pulling rate had been greater than 10000 ft/hr since an alarm level low enough to detect this level of contamination would result in several false alarms.

In most applications, if the detector can be positioned no more than 8 inches from the tubing and there is less than 1/2 inch of steel between the detector and the tubing, contamination levels of 50 $\mu\text{R/hr}$ can be detected with a properly operating/adjusted system similar to the one tested.

CONCLUSIONS

- Sodium iodide crystals of 3 x 3 inch and larger can be used with an alarming ratemeter to detect NORM contaminated tubing. Smaller crystal detectors may be able to detect contaminated tubing if they are positioned within 5 inches of the center of the tubing.
- The detector can be placed inside an aluminum explosion proof enclosure and still detect contaminated tubing.
- To increase sensitivity, the detector should be positioned so the face of the detector is as close as possible to the center of the borehole. Tests have shown that distances of less than 8 inches are acceptable if there is little steel between the tubing and the detector.

DETECTING NORM CONTAMINATED TUBING DURING PULLING OPERATIONS

- The amount of material between the detector and the tubing being pulled from the well should be kept to a minimum to improve system sensitivity.
- The response time of the ratemeter alarm circuit should be about one second. Faster response times will increase error rates and response times of 3 seconds or longer may not allow low levels of contamination to be detected at high pulling rates.
- Shielding of the detector will decrease the background count rate and thus improve the signal to noise ratio. That will improve the ability of a system to detect low levels of contamination.

For installations where:

- 1) the pulling rates are high,
- 2) the detector must be placed over 8 inches from the center of the tubing,
- 3) there is more than a 1/2 inch of steel between the tubing and the detector, or
- 4) the tubing is large or casing is being pulled,

tests should be performed to determine if this system can detect down to the regulatory limit or if a system with a larger detector or multiple detectors would be required to detect low levels of NORM contamination.

Tests will need to be performed in each field to determine the proper alarm level setting for detecting contaminated tubing. Since background counting rates, tube pulling rates, and installation configuration will vary from field to field and rig to rig, the proper settings for each installation must be determined. A suggested starting alarm level is 1-1/2 times background count rate for onshore installations and 2 times background for offshore installations..

ACKNOWLEDGEMENTS

I wish to thank Mr. Ralph Harding for requesting this device and for his support during the first field test of these systems. I also wish to thank Mr. Charles Brooks for arranging for the second field test and the assistance of the workover crews during the field tests. The field test would not have been a success without their help.

My thanks also go to Dr. I. R. Supernaw for his suggestions during the selection and laboratory testing of these devices.

HIGH DENSITY RADIOLOGICAL SITE CHARACTERIZATION SURVEYS FOR NORM SITES

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Abstract

A continuous (1 sample/sec) walkover radiological survey was performed over 13.4 hectares of open and wooded, NORM-contaminated terrain. Data from a near-surface NaI detector, and either a second NaI detector or a dose rate meter at one meter above the surface, were recorded at each sampling location along with the position coordinates. The position coordinates were automatically determined each second to an accuracy of 15 cm using an ultrasonic time-of-flight technique.

Introduction

The radiological surveys were conducted on three sites. Site A contained active and inactive oil wells, piping runs and pipe storage areas. Site B was the location of a pipe descaling operation, two oil storage tanks and an unregulated public dumping area. Site C contained three standing storage tanks, one destroyed tank, fields in which the descaled piping was used for fencing, farm buildings and ponds. The topography of these sites was generally flat with some hills and slopes.

Survey System

The radiological survey was conducted using Chemrad's USRADS® System interfaced to two scintillation detectors and a dose ratemeter. The scintillation detectors were Ludlum Model 44-2 Sodium Iodide (NaI) probes (2.54 cm x 2.54 cm) connected to Ludlum Model 3 ratemeters. The dose ratemeter was the Bicron MicroRem meter.

The USRADS System consisted of a Data Pack, Stationary Receivers, a Master Controller and a portable personal computer. The Master Controller and the computer system

were mounted in a utility vehicle which was positioned near the survey area. The Data Pack and the detectors were carried by the surveyor. The Data Pack contained the interface circuitry for the radiological survey instruments, the ultrasonic transmitter, and radio frequency (RF) equipment to establish two-way communication with the Master Controller. A handheld terminal was also connected to the Data Pack to allow the surveyor to remotely interact with the computer. The surveyor also carried a two-way radio for voice communications with the computer operator.

Independent of the USRADS Survey, an attempt was made to use a differential global positioning system (GPS) to survey the same areas. This system proved to be satisfactory for surveying open areas, but incapable of operating suitably for walkover surveys in woods or around structures where the view of the sky was obstructed. Unfortunately, the contaminated locations were most often in these obstructed areas, so the use of the GPS system was discontinued.

System Operation

Each second the Data Pack transmitted the radiological survey data from all three detectors to the computer via the RF link. At the beginning of each one second interval, the ultrasonic crystal mounted on the surveyor's backpack was pulsed by the Data Pack. The Stationary Receivers measured the time taken for this pulse to reach each of them. Each Stationary Receiver relayed a stop signal to the Master Controller via an RF transmission upon receiving the ultrasonic signal.

Through this process, the distance of the surveyor from each of the Stationary Receivers was determined using the measured speed of sound. The computer then calculated the location of the surveyor relative to the Stationary Receiver locations and stored the radiological survey data with that location. The location of the surveyor and the radiological survey data were plotted on a grid map that was shown on the computer's display in real time. The computer operator performed an on-line quality assurance inspection that the survey was being conducted according to the prescribed protocol, that the detectors were performing properly, and that the findings were reasonable. Any deviations from these conditions were immediately investigated and corrected. Once proper survey coverage has been accomplished, the operator has the choice of several software routines to assist in the identification of anomalies or other items of interest.

The number of Stationary Receivers deployed depended on the size of the area to be surveyed, the topography of the site, and the presence of woods or structures. Up to thirteen Stationary Receivers were used on sites as large as 100 m x 100 m. The time taken to setup the USRADS System and complete the survey of these larger sites would require as much as three hours, particularly if the site contained steep terrain or woods.

Survey Protocol

The standard survey protocol called for one NaI detector to be swung back and forth in

front of the surveyor while being held approximately 15 cm above the ground. The second NaI detector and the dose ratemeter were carried approximately 1 meter above the surface.

To the extent physically possible, the surveyor walked over the site along parallel lanes separated by 1 to 1.5 meters at a pace of approximately .75 meters/sec. This typically provided over 8,000 sample locations for each detector within a 100m x 100m area.

Areas in which elevated activity was detected were further surveyed by repeating the above protocol in a cross hatched pattern.

In addition to the walkover survey, one minute stationary biased samples were collected at designated locations. These were the locations of highest readings within an area, or positions at which manual readings had previously been taken using a Pressurized Ion Chamber (PIC) at 1 meter above the surface to determine the radiation dose rate. If the USRADS Survey was performed prior to the PIC readings being made, then the USRADS biased locations were marked with flags so that PIC readings could subsequently be made at the same locations.

Results

The designated areas were successfully surveyed according to the prescribed protocol, including wooded areas and around buildings and structures. Preliminary data presentations for each day's work were used to perform QA/QC on the data, to review the survey's progress, to consider the significance of any findings, and to direct the subsequent day's activities. Daily deliverables included color track maps showing each detector's position and readout by color levels each second.

Hot spots found during the survey were later successfully relocated using the maps produced. Some of the surveys showed contaminated areas outside of areas previously remediated. The USRADS System produced maps defining the areas that were above release levels.

Final deliverables included a consolidated track map showing surveyor coverage (consolidated for an entire site) and consolidated color contour maps showing the radiation findings as isopleths of constant intensity for each detector. The final data presentations, including the final version of the daily deliverables, were delivered within two weeks following the completion of the survey.

Examples of the deliverables are shown in Figures 1-8. Figures 1 and 2 show the color track maps for the NaI detectors at 15 cm and 1 meter above the surface, respectively, for the survey of OW1 Site. Figures 3 and 4 show the respective color contour maps for this area. Note that the contamination is located on a service road adjacent to the well where it can easily be transferred onto the wheels of vehicles.

Figures 5 and 6 show the corresponding track maps at the OW52 Site, while the color

contour maps are shown in Figures 7 and 8. Of significance in these maps is the finding of a hot area in the northern portion of the area surveyed. This area was on the edge of a washout, suggesting that part of the contaminated soil may have eroded downhill. Also noteworthy was the finding of a small hot spot near the middle of the surveyed area which was an isolated concentration of activity in an otherwise clean area.

Conclusions

The USRADS Radiological Survey was highly successful in accurately and cost effectively locating radioactive hot spots and measuring the radiological dose rate in all terrains, including woods and around structures. Clean areas were also thoroughly documented.

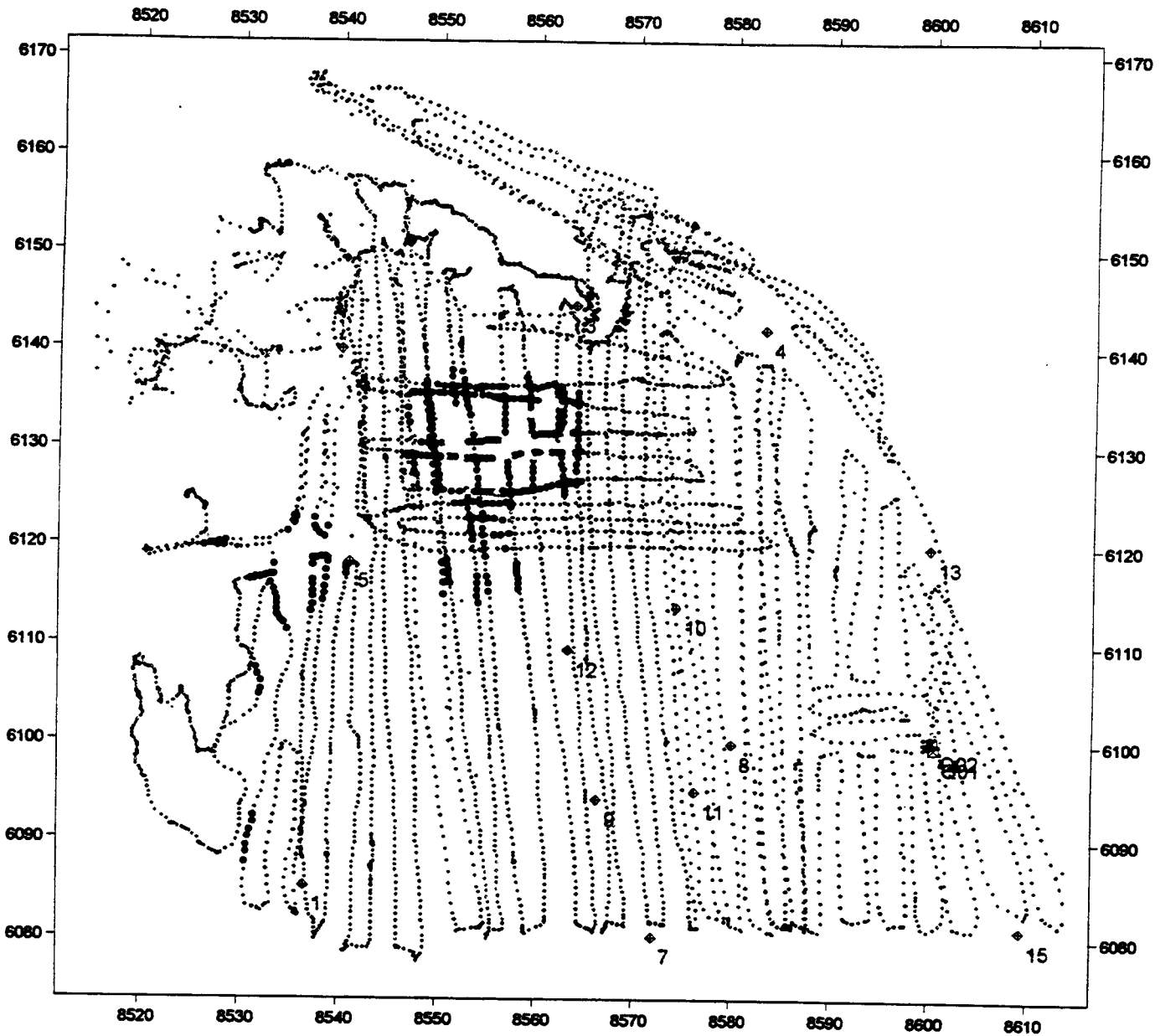
The GPS positioning technology was shown to work satisfactorily, but with somewhat poorer positional resolution, in open areas, but did not work in wooded areas or around structures.

Site: OW1 (A)

Signal: NaI (CPM)

Time: 13:00:59 05/24/95

Threshold:
> 15161 •



9405.5478

Figure 1.

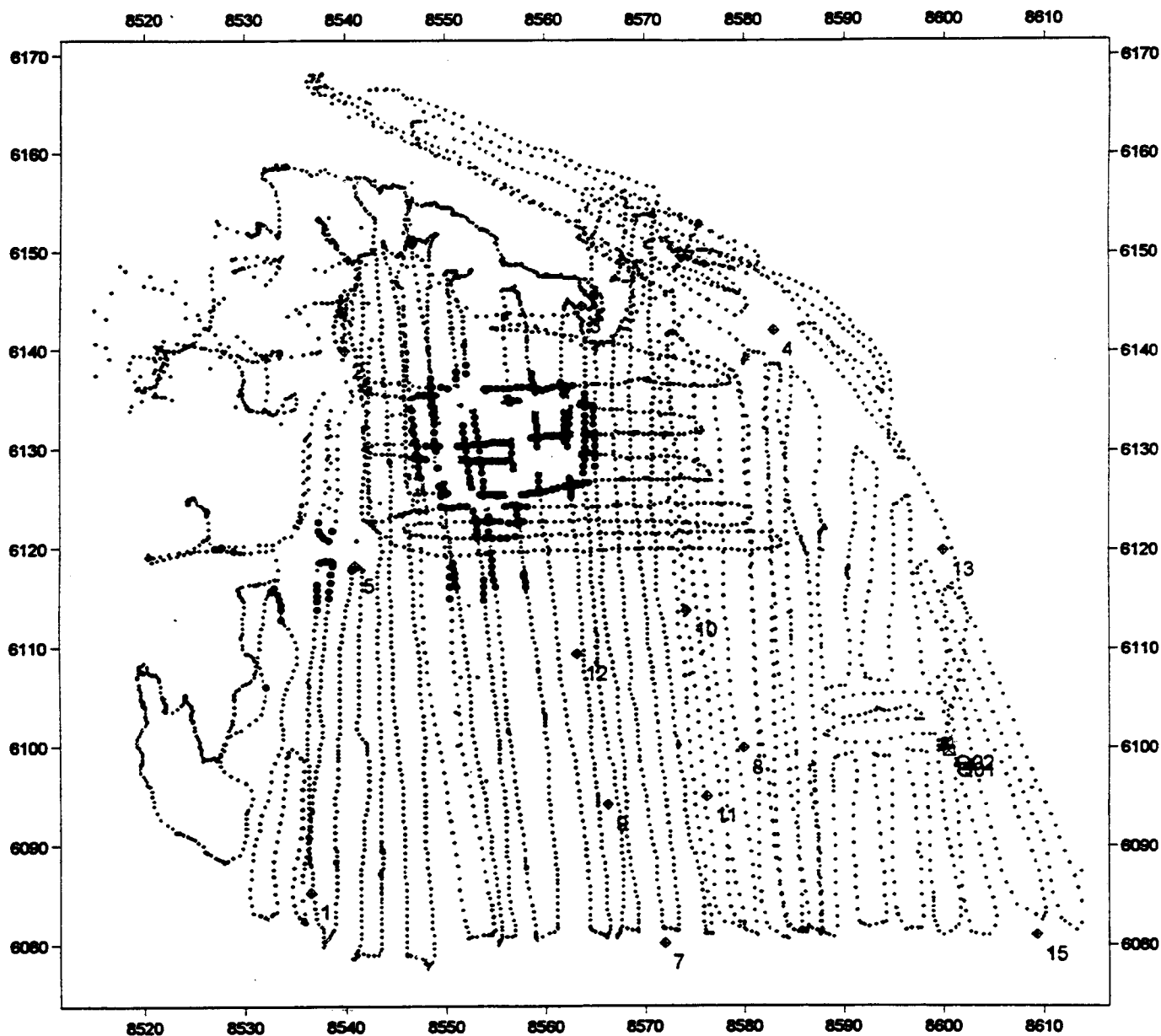
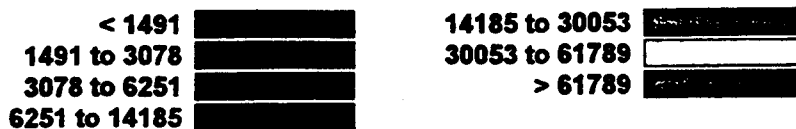
1 of 1

Site: OW1 (A)

Signal: 1MNaI (CPM)

Time: 13:00:59 05/24/95

Threshold:
> 14920 •



9405.8060

Figure 2.

1 of 1

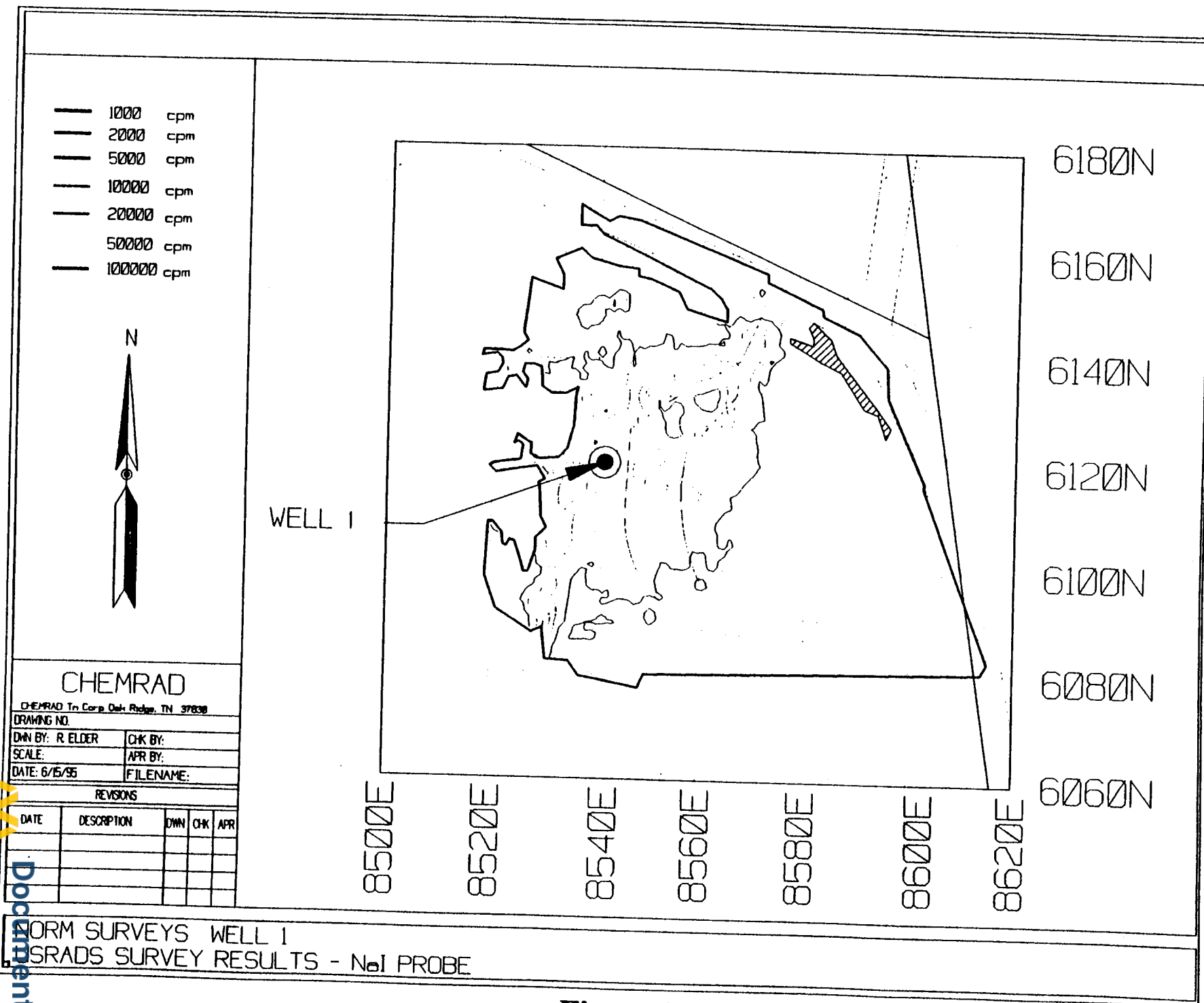


Figure 3.

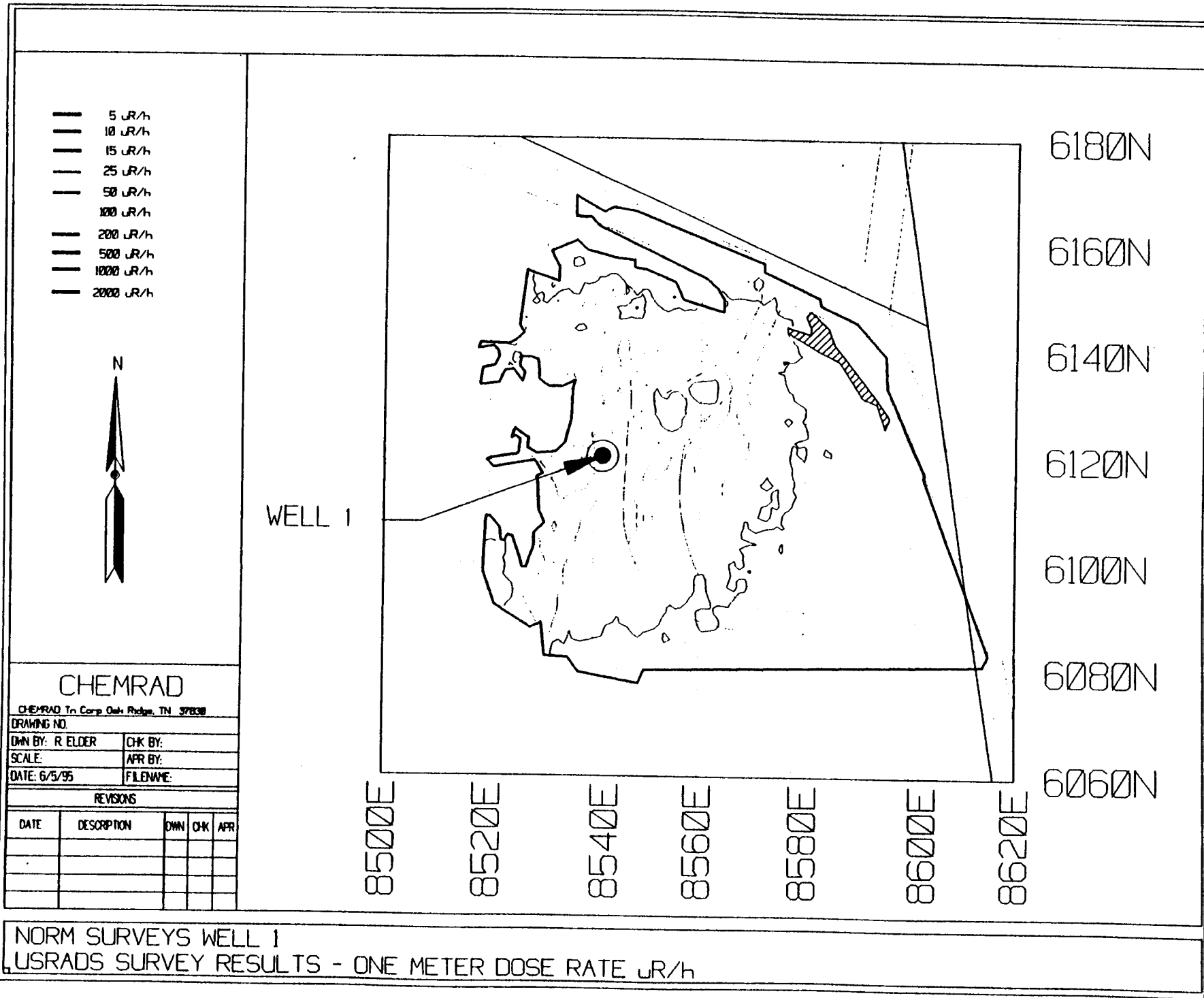


Figure 4

Site: OW52 (A)

Signal: Nal (CPM)

Time: 09:58:29 05/24/95

Threshold:
> 13121 •

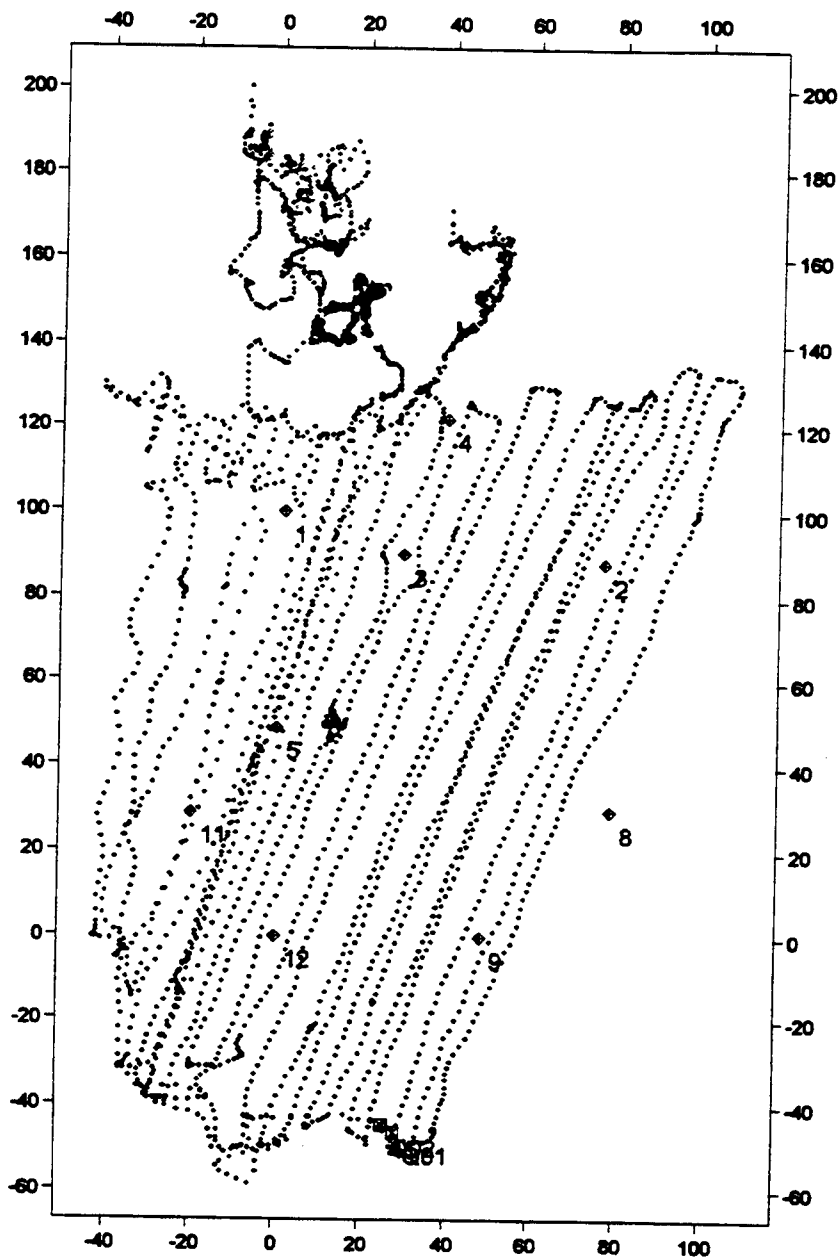
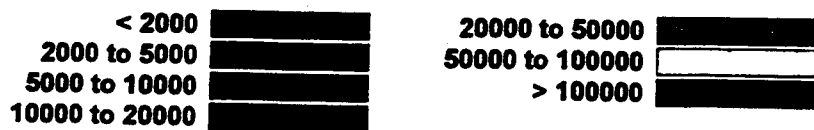


Figure 5.

9402.9224

1 of 1

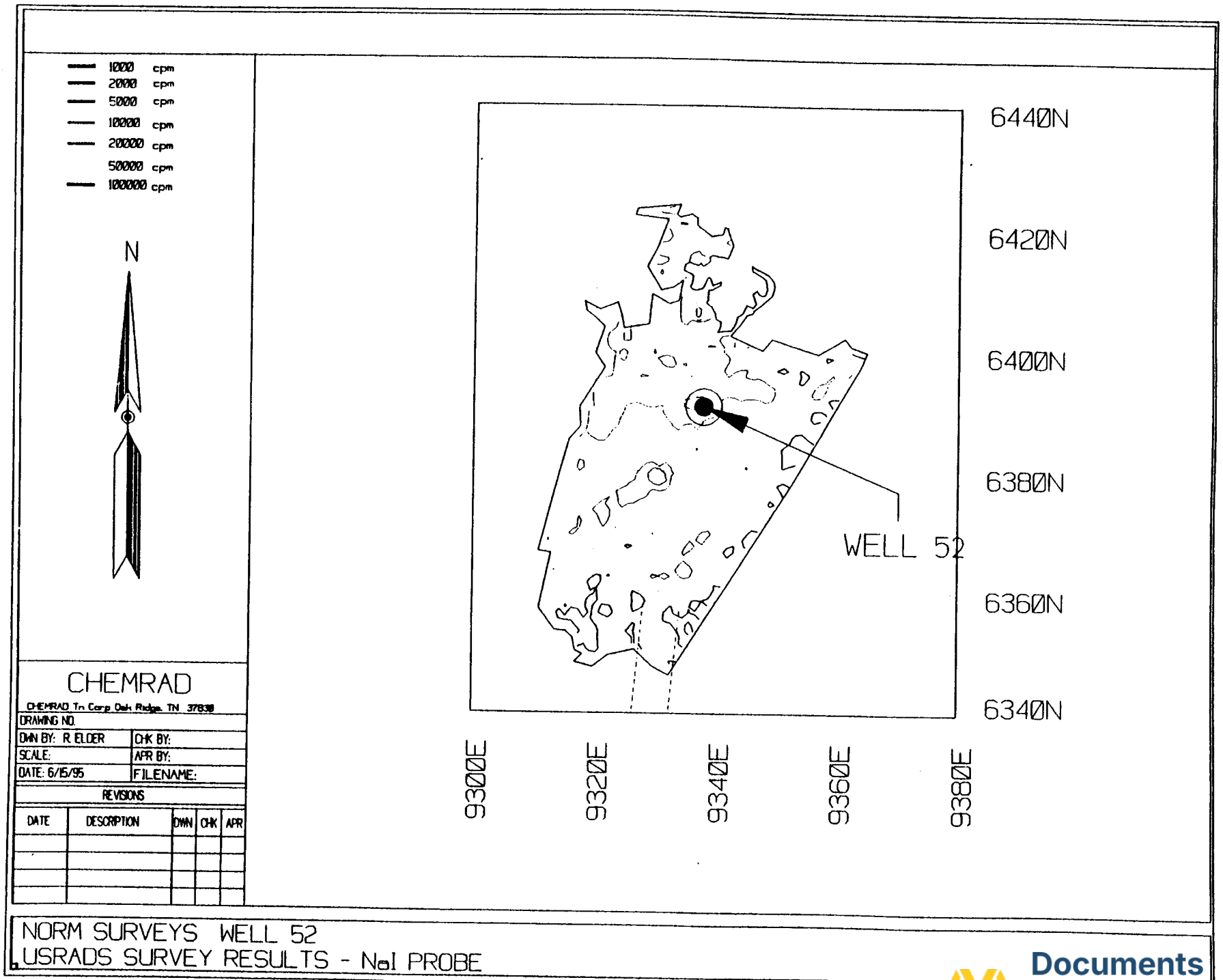


Figure 7.

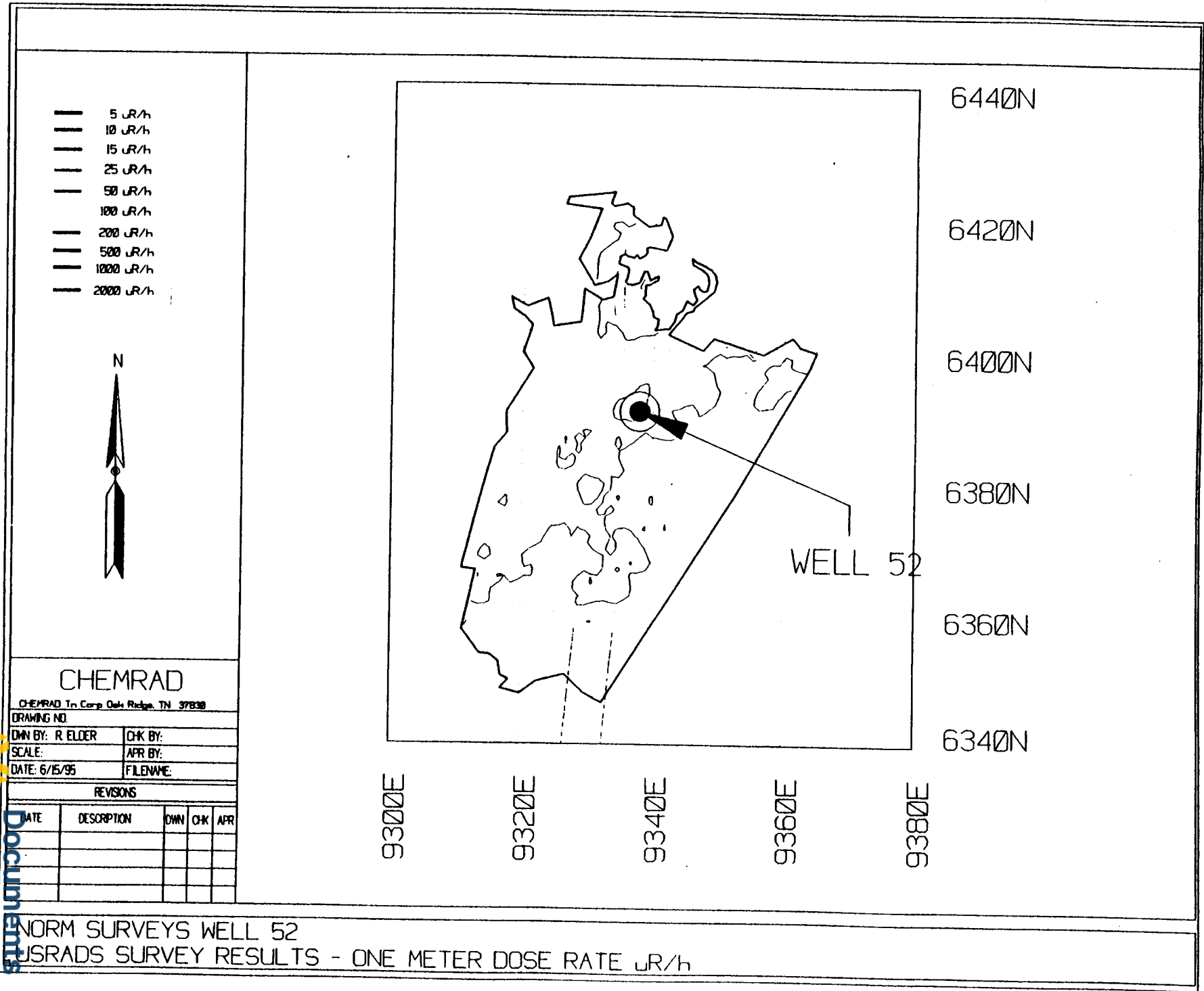


Figure 8

Methods of Estimating Dose to Individuals from NORM

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Abstract

Dose from NORM for occupational situations does not require methods and techniques different from other ionizing radiation situations. In some case additional care is necessary since the radiations cannot be distinguished from background, except for intensity.

The regulations concerning occupational dose from NORM are 10 CFR 20 and 29 CFR 1910.96. In order to show that no radiation protection program is required under either of these regulations, careful measurements with compensated instrument readings may be necessary, as well as, a good definition of the local background. Dosimetry devices may be required for a radiation protection program and could be very useful in demonstrating no need for a radiation protection program.

The regulations governing occupational dose also require controlling public dose. Public dose must be considered in a radiation protection program.

Risk calculations for occupational situations may be required under RCRA and CERCLA, although there is no clear regulatory basis for the risk calculations. Such calculations require attention to detail if risk limits are approached. Knowledge of the background is essential and measurements should be made at actual work locations. Dosimetry devices may be useful, but measurements must be well controlled.

Introduction

Ionizing radiation dose to individuals in occupational situations is covered by 10 CFR 20 or 29 CFR 1910.96. Both these regulations limit dose to individuals to 5000 mrem/y (50 mSv/y) and require a radiation protection program when it is expected that any individual may obtain an annual dose in excess of 500 mrem (5 mSv). These annual doses are summed from all methods of exposure at the work place, external and internal. In general, members of the public should not receive in excess of 100 mrem/y (1 mSv/y). Few cases of occupational or public dose from NORM are explicitly covered by 10 CFR 20. The language of 29 CFR 1910.96 incorporates, by reference, 10 CFR 20 for all industrial situations not applicable under 10 CFR 20. Therefore, industrial uses of NORM are regulated with respect to dose to individuals under 29 CFR 1910.96. This is hardly an undisputed interpretation.

Additional and specific regulation of some forms of NORM can be found in NESHAPs. Based on the classification in NESHAPs of NORM as a hazardous substance, further regulation of NORM is possible under language in RCRA and CERCLA. This interpretation is also a source of dispute. Regulation under NESHAPs, RCRA, or

CERCLA may depart from dose as a limit and use life-time risk as a limit. Depending upon the exposure period, a life-time risk of 10^{-4} may result in an annual dose far less than 100 mrem (1 mSv).

The dose range of interest, up to 5000 mrem/y, less than 500 mrem/y, less than 100 mrem/y, or less than a life-time risk of 10^{-4} , present differing problems in individual dose measurement. Not surprisingly, the difficulties increase with decreasing dose.

We discuss methods and problems of external dose measurement based on experience in several types of facilities where NORM was encountered. These cases include metal and rare earth extraction, water treatment, phosphorus and phosphate extraction, lithium extraction and processing, and a plant disposing of NORM contaminated materials, where NORM was used in construction of the facilities. Internal dose was also a concern at most of these facilities, but internal dose will not be covered.

Dose Estimation Methods

Occupational Dose

The occupational category under 10 CFR 20 and 29 CFR 1910.96 require measurements to estimate doses from 0 to 5000 mrem/y or measurements to ensure that doses will be less than 500 mrem/y. These dose ranges are those of the nuclear and medical communities and have a long history of measurement. There are many standard techniques for dose rate and time estimates of dose or direct dose measurement using dosimetry devices. Doses can also be modeled based on concentrations of radionuclides and shape of the source. There are few problems with accuracy with any of the available techniques until limits are reached, either 5000 or 500 mrem/y or some other administrative limit.

The accuracy problems are associated with defining background and non-linear response of devices and instruments in the measurement of background and NORM. The problems occur with over-response rather than under-response. The response problems occur because calibrations are usually made at only a single energy rather than a spectrum of energies typical of NORM and the devices usually have an energy dependent response, which may preclude correct calibration under any circumstance.

At typical sites of the nuclear and medical communities, only a few locations account for the dose to workers. Dose rate measurements in these cases range upward from a few to tens of mrem/h. Standard dose rate instruments easily measure these dose rates. If the radiation field is not localized, but general, exposing the worker at all or nearly all time, then a dose rate of 0.25 mrem/h for 2000 h would constitute the annual dose limit. A dose rate this small or smaller strains the reliability of standard instruments designed to read in mrem/h. More sensitive instruments, designed to read in μ rem/h are required.

Dosimetry devices are usually required to measure dose when a radiation protection program is required. Standard dosimetry devices should have few limitations in the region

of dose from 500 to 5000 mrem/y. Two important variables must be considered, time of exposure and storage background for the device, although non-linear energy response may be critical in some cases. Nevertheless, most programs are straight-forward, unless dose limits are approached. If there is a possibility of exceeding dose limits, a program for controlling dose should be used before refining the dosimetry process.

Public Dose

When there is a consideration of occupational dose, 10 CFR 20 and 29 CFR 1910.96 require consideration of public dose. The public category of dose for a limit of 100 mrem/y is, almost always, calculated. The calculation is usually conservative, based on a Reasonable Maximum Individual. Dose can be measured using dosimetry devices and sensitive recording instruments at typical public locations. These measurements are usually performed in conjunction with a calculation of dose. Measurements are acceptable as the only method of dosimetry.

Some type of measurement is necessary for calculating dose, whether the measurement is of dose, dose rate, or concentration of radionuclides. The Reasonable Maximum Individual is determined at the 95 percentile of the propagated uncertainty of measurements and time estimates. It is often necessary to reduce the uncertainties by increasing the number of measurement locations and the measurement accuracy. Standard techniques are usually available for making environmental measurements for dose calculations.

Risk Calculations

Dose measurements to support RCRA or CERCLA do not apply to occupational situations according to 29 CFR 1910.96, but risk assessments for outdoor occupations on sites where there is RCRA or CERCLA activity have been made. Workers assessed were not involved in the RCRA or CERCLA activity. To date no consequences have resulted from such risk assessments of which the authors have knowledge, since the risks were found to be 10^{-4} or less. Considerable effort was required to obtain an accurate risk assessment. Accuracy was achieved by basing the calculations on appropriate measurements at appropriate locations.

Dosimetry

Dosimetry is an attractive method of measuring individual dose since it is specific to an individual. There is no need to perform calculations or dose rate measurements. If additional information is necessary, such as the location where the dose was received, additional measurements are necessary since the dose is specific to the individual, not the location. Transit and storage dose must be subtracted from the total dose to obtain individual dose. Storage dose is dose received by the dosimeter for times not worn by the individual. Storage time usually exceeds wear time by a factor of 2 or more. Transit dose is the dose received between the storage location and the dosimeter processing location. Transit dose for remote processing facilities can equal the storage dose. Storage and transit dose can usually be obtained from control dosimeters. These 2 subtracted doses

become more important as the individual dose decreases and the time to measure the dose increases.

The time to measure the dose is dependent upon the accuracy and sensitivity required for the dose estimate. Doses that are clearly different from background can be measured in a short time with acceptable accuracy. Doses down to 100 mrem/y can be measured in a three month exposure. Smaller doses require considerable care for the transit and storage doses and up to a year of exposure. Figure 1 shows measurements made with Al_2O_3 dosimeters in an attempt to measure as low as 10 mrem without particular control of the storage dose.

The figure shows measurements made at 2 dose rates, 20 and 40 $\mu\text{rem/h}$. The intercept of the 2 lines is 12.5 mrem and is the sum of the storage and transit doses. The horizontal line at 22 mrem is the detection limit based on 5 dosimeters at the storage location. The results show that at the lower dose rate of 20 $\mu\text{rem/h}$ over 400 h or 10 weeks of exposure are necessary for a detectable difference from background. The higher dose rate of 40 $\mu\text{rem/h}$ was detectable in less than 200 h or 5 weeks. Based on this analysis, at least, 3 months would be required to obtain acceptable accuracy at the higher dose rate (Currie 1978).

In a test where the dosimeters were worn by individuals, the control on the wear time and storage dose was sufficient for a detection limit of 30 mrem or 3 times the static test. Further tests were performed for better storage control in a low background area. A detection limit of 15 mrem was found to be achievable.

Dose Rate

Dose rate is the most popular method of estimating dose, because it is immediate and identifies the source. The major drawback is that good estimates of the time of exposure and the position of exposure are necessary. For occupational doses reportable under 10 CFR 20 and 29 CFR 1910.96, dose by a dosimetry device is usually required. Dose rate measurement and a conservative calculation is sufficient to show that a radiation protection program is not required because doses are less than 500 mrem/y.

The accuracy of dose rate measurements may be critical when limits are approached. It may be necessary to reduce conservatism. Conservatism can be reduced by converting over-responding instruments using an equation that converts the calibration conditions to the NORM conditions. Additional removal of conservatism may be gained by converting to equivalent dose based on the geometry of the NORM source (EPA 1993, ICRP 1987, Zankl 1992).

Conversion of instrument response to NORM conditions requires knowledge of the NORM dose rate at various locations for comparison to the instrument reading at the same locations. Pressurized ionization chambers (PIC) are energy independent over the range of background and NORM energies (Beck 1971). It is accepted as the standard for

dose in air for human dose measurements. Other types of ion chambers and compensated GM detectors may also be adequate but should be compared to a PIC under the conditions to be measured. Tissue equivalent plastic scintillation detectors can be very close to NORM dose rate, requiring no conversion.

The conversion of the dose rate is usually more important at the lowest dose rates. Measurements of low dose rates are more likely to require conversion, not because of the energy mixture at low dose rates, but because most low dose rate instruments are chosen for their sensitivity. In other words, a high response at low dose rates usually means over response. Detection of radiation in some cases may be more important than correctly measuring it. A popular instrument for low dose rate measurements is the sodium iodide scintillation detector. The usual calibration of this instrument results in over response for low energies and under response for high energies. For NORM, this means over response. Attempting to adjust the response for certain conditions is usually not advisable, unless it is known that use will be restricted to only those conditions.

The sodium iodide readings can be converted to PIC equivalents by comparing to a PIC over a range of dose rates, then regressing the sodium iodide readings against the PIC readings. The result is

$$\text{PIC} = a(\text{NaI}) + b,$$

PIC is the PIC equivalent, NaI is the sodium iodide reading, a is a constant less than 1 and b is a constant greater than 1. The authors have found that a is usually near 0.6 for all locations and b is usually equal to the cosmic dose rate. The constant b is, therefore, altitude dependent. The generality of this equation allows for easy conversion without extensive measurements, but should always be confirmed in critical situations.

Geometric compensation for equivalent dose may also be necessary to eliminate conservatism. The PIC is designed to measure dose in air, which can usually be converted to dose in tissue at 1 cm depth by multiplying by 0.96 (Beck 1971). This conversion is not sufficient for conversion to whole body dose for all energies since equivalent dose must be calculated for internal organs (EPA 1993, ICRP 1987, Zankl 1992). The geometry of the source must also be considered since the organs are differently irradiated from different directions. Figure 2 shows the energy dependent conversion for a body standing on an infinite planar source. For the usual energy spectrum for NORM, the PIC conversion factor for the planar source is 0.65.

Measure or Model

For annual doses down to 500 mrem/y it may be required that doses be measured using dosimetry devices, but that dose rates be measured at locations for purposes of posting and designing a radiation protection program. From 500 to below 100 mrem/y a combination of measurement and calculation may be possible, depending upon the accuracy or need to demonstrate protection is adequate by conservative estimate. As

doses become more critical, a more exacting measurement program may be necessary. If occupational doses become limited by RCRA or CERCLA risk estimates, careful measurement and exacting calculations may be necessary.

Measurement for the lower dose regions may be limited by the choice of instrument or dosimetry device. The lower the dose, the more sensitive the instrument or device, inviting over response. Important to low dose measurements is correctly measuring the background and the range of background. It may be important to know the local background for a given measurement rather than the general background for the area. Figure 3 shows a probability plot for background in a region where NORM measurements were made. Knowing the local background in this case can be very important, especially if dose rates on the order of 10 μ rem/h are important. Observe from the figure that 2 straight lines can be fitted to the data, indicating 2 different background areas. The area comprised both a volcanic flow and alluvium.

Modeling in this case must also consider this differing background. Models may only allow an average background, considering all dose rates above the average to be from the NORM source. Common models also may use the highest source dose rate for a location rather than the average for the location. Models may also discount shielding and maximize the time of exposure. The inherent conservatism of some models is acceptable, as long as the doses calculated are below the imposed limits. When limits are exceeded, then a more careful evaluation of the dose is necessary.

When limits are exceeded in a modeled dose, conservatism must be eliminated. Conservatism is inherent in most models, but often, the model can be improved by improving the measurements and by performing measurements representative of the work situation. In a case involving the authors, risk was modeled for occupational doses at CERCLA sites. The first risk calculation was based on dose rates calculated from the NORM concentration at selected locations. It was assumed that these concentrations were representative of other locations.

When the calculated risk easily exceeded life-time risks of 10^{-4} , dose rate measurements were requested for the same locations where dose rate was calculated from the concentrations. The dose rate measurements were performed with a GM detector without compensation for the over-response. The dose rates measured were found to be equal to or higher than the calculations. Since the locations used for the calculations and measurements were not typical of occupational locations, measurements from other locations were used to obtain average dose rates for areas of the sites. These additional measurements were previously performed under NESHAPs and/or RCRA. These measurements were made with an NaI scintillator, were also not made at occupational locations, and did not appreciably alter the risk assessment.

A series of measurements were made in occupational locations and in occupational situations (shielding) using a PIC and a tissue equivalent plastic scintillator. The majority of the occupational time was spent in vehicles, heavy earth moving equipment, or in

equipment control rooms. Figure 4 shows the results of some of these measurements. At each position measured, the total unshielded, total shielded, and net (above background) dose rates are shown. The net dose rate is the quantity necessary to calculate occupational risk. Once actual, net dose rates are known, the life-time risk can be calculated. The calculation for each occupation is performed as follows:

$$\text{Risk} = \text{RF} \times \text{DA} \times \text{WY},$$

where

RF = risk factor

DA = annual dose

WY = years of work.

Annual dose is

$$\text{DA} = \sum (\text{NDR}_i \times \text{ET}_i \times \text{EF}_i),$$

where

NDR = net dose rate (average for the location)

ET = exposure time in hours per day

EF = exposure frequency in days per year.

At this point in the calculation, the uncertainties can be propagated for determination of a Reasonable Maximum Individual. Often, in the name of conservatism, a Reasonable Maximum Value is calculated for each term, which greatly inflates the calculated risk. In the CERCLA risk calculations for these sites Reasonable Maximum Values were used, but the use of net dose resulted in life-time risks of less than 10^{-4} . Calculation of equivalent dose, which was not done in these cases, would have further reduced the calculated risk.

Conclusions

Compliance with 10 CFR 20 and 29 CFR 1910.96 can be accomplished with standard equipment and methods for radiation protection programs. Care must be taken as limits are approached that instrument readings are properly compensated. When it can be shown that no radiation protection program is required under either of these regulations, careful measurements with compensated instrument readings may be necessary, as well as, a good definition of the local background. Dosimetry devices may be required for a radiation protection program and could be very useful in demonstrating no need for a radiation protection program. It may be necessary to convert doses if the dosimetry devices over-respond to NORM.

Risk calculations for occupational situations under RCRA and CERCLA require attention to detail if limits are approached. Knowledge of the background is essential and measurements should be made at actual work locations. Dosimetry devices may be useful, but storage and transit doses must be well controlled.