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ENVIRONMENT
department

March 30, 1988

Mr. Michael Bell
U.S. Nuclear Regulatory Commission
Division of Waste Management
Mail Stop 697SS
Washington, DC, 20555

Dear Mr. Bell:

Enclosed are the two most recent reports issued by EEG. "A Critical
Assessment of Continuous Air Monitoring System at the Waste Isolation Pilot
Plant" by John C. Rodgers and Jim W. Kenney, EEG-38, and "Chemical and
Radiochemical Characteristics of Groundwater in the Culebra Dolomite,
Southeastern New Mexico" by Jenny B. Chapman, EEG-39.

EEG-38 is a detailed study of a critical component of the WIPP occupational
health protection and stack monitoring programs. The report focuses on
several likely deficiencies in the design of the alpha (L x-ray) and beta
continuous air monitors (CAMs) that have been installed at WIPP, and reaches
the conclusion that the present designs apparently fail to meet mandatory
performance requirements for such instruments prescribed by DOE Orders.

Specific topics covered include instrument calibration, background
interference correction, detector response, and particulate losses in the CAM
head. Much of the analysis is based on physical models of the fundamental
processes involved. In addition, the findings of a number of important
experiments are reported, which validate and substantiate many of the
conclusions.

Rodgers and Kenney recommend that a program of detailed performance testing of
the existing CAMs be started at once to confirm (or reject) the principal
findings of this report. In the meantime, it is strongly recommended that a
vigorous search be initiated to find alternative CAM systems and detection
schemes.

EEG-39 presents the interpretation of chemical data from groundwater samples
collected from the principal water-bearing unit in the WIPP area. The EEG
groundwater sampling program has three objectives: 1) major ion analyses help
determine groundwater flow paths and reactions that occur along the flow
paths, 2) radiochemical data establish a baseline of existing radionuclides in
area groundwater prior to the arrival of waste at WIPP, and 3) stable and
radioactive constituents help predict the behavior of the Culebra as a
radionuclide transport pathway. These topics are discussed in detail in the
report and all of the data are presented in the appendix.

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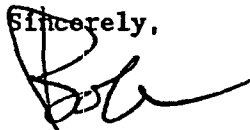NHPA 1,

Ms. Chapman concludes that major influx of freshwater recharge to the Culebra occurs south of the WIPP Site. There is also chemical evidence that the Culebra contains discrete flow paths (fractures or karst) interspersed with areas of slower water movement. Water chemistry variations correlate well with the amount of evaporite dissolution in the Rustler Formation. The concentrations of naturally occurring radionuclides indicate that some nuclides (Ra, U, and Th) are relatively soluble in the high-chloride, oxidizing Culebra environment. However, water in the southern, low-chloride area appears less favorable for transport of radium isotopes.

Recommendations include continued radionuclide analyses to improve the statistical data base and exploration of naturally occurring waste analogs. Additional sampling for major ion chemistry is needed in the southern area of suspected recharge and near well H-5 where conflicting interpretations suggest both very old and very young water.

Sincerely,

Robert H. Neill
Director

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Encls.

EEG-38

A CRITICAL ASSESSMENT OF CONTINUOUS AIR MONITORING SYSTEMS AT THE WASTE ISOLATION PILOT PLANT

John C. Rodgers
Jim W. Kenney

Environmental Evaluation Group
Reports

EEG-1   Goad, Donna, A Compilation of Site Selection Criteria
         Considerations and Concerns Appearing in the Literature on
         the Deep Disposal of Radioactive Wastes, June 1979.

EEG-2   Review Comments on Geological Characterization Report, Waste
         Isolation Pilot Plant (WIPP) Site, Southeastern New Mexico
         SAND 78-1596, Volumes I and II, December 1978.

EEG-3   Neill, Robert H., et al, (eds.) Radiological Health Review
         of the Draft Environmental Impact Statement (DOE/EIS-0026-D)
         Waste Isolation Pilot Plant, U. S. Department of Energy,
         August 1979.

EEG-4   Little, Marshall S., Review Comments on the Report of the
         Steering Committee on Waste Acceptance Criteria for the
         Waste Isolation Pilot Plant, February 1980.

EEG-5   Channell, James K., Calculated Radiation Doses From
         Deposition of Material Released in Hypothetical
         Transportation Accidents Involving WIPP-Related Radioactive
         Wastes, November 1980.

EEG-6   Geotechnical Considerations for Radiological Hazard
         Assessment of WIPP. A Report of a Meeting Held on January
         17-18, 1980, April 1980.

EEG-7   Chaturvedi, Lokesh, WIPP Site and Vicinity Geological Field
         Trip. A Report of a Field Trip to the Proposed Waste
         Isolation Pilot Plant Project in Southeastern New Mexico,
         June 16 to 18, 1980, November 1980.

EEG-8   Wofsy, Carla, The Significance of Certain Rustler Aquifer
         Parameters for Predicting Long-Term Radiation Doses from WIPP,
         September 1980.

EEG-9   Spiegler, Peter, An Approach to Calculating Upper Bounds on
         Maximum Individual Doses From the Use of Contaminated Well
         Water Following a WIPP Repository Breach, September 1981.

EEG-10   Radiological Health Review of the Final Environmental
         Impact Statement (DOE/EIS-0026) Waste Isolation Pilot
         Plant, U. S. Department of Energy, January 1981.

EEG-11   Channell, James K., Calculated Radiation Doses From
         Radionuclides Brought to the Surface if Future Drilling
         Intercepts the WIPP Repository and Pressurized Brine,
         January 1982.

A CRITICAL ASSESSMENT OF CONTINUOUS AIR MONITORING
SYSTEMS AT THE WASTE ISOLATION PILOT PLANT

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March 1988

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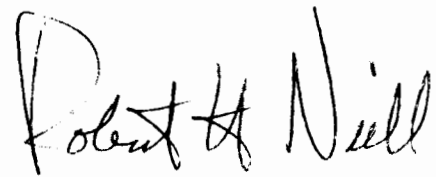Division, Health Physics Group, P. O. Box 1663, Los Alamos, NM 87545.

FOREWORD

The purpose of the Environmental Evaluation Group (EEG) is to conduct an independent technical evaluation of the potential radiation exposure to people from the proposed Federal Radioactive Waste Isolation Pilot Plant (WIPP) near Carlsbad, in order to protect the public health and safety and ensure that there is minimal environmental degradation. The EEG is part of the Environmental Improvement Division, a component of the New Mexico Health and Environment Department -- the agency charged with the primary responsibility for protecting the health of the citizens of New Mexico.

The Group is neither a proponent nor an opponent of WIPP.

Analyses are conducted of available data concerning the proposed site, the design of the repository, its planned operation, and its long-term stability. These analyses include assessments of reports issued by the U. S. Department of Energy (DOE) and its contractors, other Federal agencies and organizations, as they relate to the potential health, safety and environmental impacts from WIPP.

The project is funded entirely by the U. S. Department of Energy through Contract DE-AC04-79AL10752 with the New Mexico Health and Environment Department.

Robert H. Neill
Director

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EXECUTIVE SUMMARY

The State of New Mexico Environmental Evaluation Group has undertaken an extensive review of the expected performance of the Waste Isolation Pilot Plant (WIPP) Continuous Air Monitor (CAM) instrumentation for monitoring of airborne transuranic (TRU) radionuclides. The CAM design chosen and installed at WIPP represents a significant departure from the original bid specifications for this system, and appears to have failed to meet the minimum performance requirements for such devices.

A particularly significant finding of the assessment is that this instrument is likely to fail to properly correct for the background interference caused by the collection of radon and thoron progeny collected on the filter during long-term continuous sampling. Although the analysis was limited by the unavailability of actual performance data, detailed computer simulations were used to identify specific aspects of the expected instrument response which contribute to its anticipated shortcomings.

Among the specific problems identified in addition to background subtraction were potential loss of sample in nozzles and transport lines, and sample loss or distortion in the CAM head itself due to impaction or asymmetrical deposition during sampling.

The several concerns raised by this assessment warrant a full and careful review by the WIPP Project Office. If these findings are confirmed by much needed laboratory and field tests of the instrument, a new approach to continuous plutonium monitoring will have to be identified and demonstrated before the project becomes operational given the crucial role these instruments have in the safe operation of the plant.

SUMMARY OF RECOMMENDATIONS

Based on the detailed analysis of the plutonium continuous monitoring instrumentation, the following recommendations are made:

1) The Nuclear Research Corporation (NRC) L x-ray and beta CAMs must be subjected to thorough laboratory performance testing to demonstrate that they are capable of meeting the requirements of DOE Orders under realistic WIPP conditions.

2) On-site confirmatory testing under worse-case conditions expected at various locations is recommended as well.

3) Alternative approaches to the L x-ray detection of plutonium need to be considered in light of the apparent deficiencies in the present instrument design.

4) The WIPP Project Office should make provisions for including EEG in the needed CAM design review, peer reviews, and test plan development to ensure full and prompt review of plans to develop a sound alternative.

ACKNOWLEDGEMENTS

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1.1 Overview of Continuous Air Monitoring Design Issues

The safe operation of the Waste Isolation Pilot Plant (WIPP) site requires that air in the work place above and below ground, and the exhaust air, be continuously monitored for the presence of airborne transuranic (TRU) radionuclides (esp. Pu-238, Pu-239, and Am-241), and certain fission products as well (esp. Cs-137 and Sr-90).

It should be understood at the outset that a complete set of alpha and beta Continuous Air Monitors (CAMs) sufficient to meet the needs of monitoring above ground in the Waste Handling Building and the Exhaust Filter Building complex (including the stack monitors), and below ground in the drifts and storage/disposal rooms has been designed, built, and installed at WIPP. All of this has been accomplished with a minimum of review, discussion, and consultation with the Environmental Evaluation Group (EEG). The only outside review known to have occurred was an apparently limited and specific investigation by the Inhalation Toxicology Research Institute (ITRI) to determine whether the prototype instrument satisfactorily met the bid specifications. The instrumentation in place was designed and constructed by the Nuclear Research Corporation (NRC). Early indications of EEG concerns about these instruments were orally communicated to WIPP staff as early as November 1986, when measurements of the radon daughter background was conducted on-site. Furthermore, detailed preliminary findings were communicated to the Site Manager by letter in September of 1987.[1] The Site Manager was urged to conduct appropriate testing of the performance of these devices so that changes could be implemented in a timely fashion. To date no proposed experimental tests have been identified to EEG, nor have the results of such testing, if conducted, been received. The EEG assessment that follows, therefore, suffers from the

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inherent limitations of a study done without verification by in-place testing. However, care has been taken to explore expected performance with realistic models of the physical processes affecting instrument performance, and thus the predicted flaws are based on more than mere hunches. But once again, we would urge that detailed experimental testing be undertaken to validate the performance of these installed devices before they are put into service.

The three major monitoring devices in the WIPP environmental monitoring system (EMS) are: 1) the area radiation monitor (beta-gamma field), 2) alpha (L x-ray) air monitor, and 3) beta-gamma air monitor. The first of these is a fixed beta-gamma radiation field detector designed to passively monitor the external exposure potential in the vicinity of the monitoring station. It is not isotope specific, is not subject to interference by normal background radiation (since it is designed to simply detect the ambient field whether background or not), and its design and operations appear appropriate to its intended use. Hence it will not be discussed further. The other two systems, in contrast, must be designed to appropriately compensate for background and interference.

The second of these, the alpha (or L x-ray) detector, is designed to continuously monitor for TRU alpha emitting radionuclides in air, whether it is in the work place, the discharged from the exhaust shaft, or the ventilation system of the Waste Handling Building. The L x-ray scintillation detector system functions by monitoring the x-radiation emitted by radionuclides collected on a filter in line with a controlled, continuous air pump or central vacuum system. In order to be an effective detector of TRU radionuclides, the system must both be sensitive to x-radiation from TRU radionuclides, and at the same time be designed to reject or correct for x-radiation from background radionuclides collected on the filter. Since many, but not all alpha-emitters and beta-emitting radionuclides also decay with the emission of L x-rays, the potential exists for interference with the scintillation detection system from normal background radon and thoron decay products. Background correction is attempted in this instrument by monitoring two energy calibrated channels in the detector signal-processing unit simultaneously, one a background

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channel at energies somewhat higher than the L x-rays of Pu and Am, the other set to detect the L x-rays from Am and Pu. The concept of background subtraction in such a system apparently is that the interference signal consists of a continuum of photon energies through the entire energy region of interest. Detection of this continuum level in an energy window at somewhat higher energy than the Pu/Am window is relied upon to obtain a background count which can be used to remove background interference by subtraction. However, if one or more of the naturally occurring radionuclides always present on the filter emits x-rays that have a spectrum of energies, which are nearly coincident with those of Am and Pu, then a serious interference condition could exist. That is, if a flux of low-energy photons (8-26 keV) from background radionuclides collected on the sample filter is being emitted continuously and is increasing as sampling occurs, and is not exactly balanced by an increase in the flux of photons in the background subtraction window (29-51 keV), or at least a constant ratio of counts develops, then this scheme for control of interference will fail. As will be discussed below, the present NRC system appears to be susceptible to this failure mode.

An additional complication arises from the fact that x-ray emission from alpha-emitters occurs typically only a fraction of the time and is variable from one radionuclide to another. For Pu-239 the yield of photons is only about 4% of all decays. For Am-241, the yield is about 3 times higher, but still a low yield. The combination of low and variable yield, low photon energy, and the presence of background L x-ray emitters presents some serious design challenges for this system. These will be discussed in detail in the following sections.

The third of these detectors, the beta-gamma detector, is designed like the alpha detector to monitor airborne radionuclides in the work place and exhaust air by active sampling of air. Again, the sensitive volume of this detector is positioned to receive radiation from radionuclides deposited on a filter. But because the detector pulse output is not proportional to energy (it is a Geiger-Mueller [GM] tube), background subtraction by two channel counting is not feasible. Background subtraction is attempted by use of a second detector monitoring the ambient beta-gamma flux. The

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difficulty with this approach is that the two detectors are sensing two fundamentally different radiation fields. The apparent presumption is that the principal sources of gamma background are high-energy photons which would affect both the ambient detector and the shielded filter detector equally, or that the two fields are highly correlated even if they are different. However, once again, the beta-gamma activity from radon and thoron daughters collected on the filter could be substantial after a few hours, and cause an appreciable count rate above the anticipated ambient GM tube count rate, and thus would not be corrected. Overall, therefore, both the L x-ray and beta CAM concepts appear to be seriously flawed and will not function to detect a release of transuranic radionuclides with required sensitivity, response time, and insensitivity to fluctuating background.

1.2 Continuous Air Monitor Bid Specifications

The bid specifications for the alpha and beta CAMs provide a useful point of departure for further review of these systems since they directly, or by reference, define a set of quantitative criteria by which the function and performance of the proposed WIPP CAM system can be judged.

The applicable codes, specifications, and standards which form a part of the WIPP Division 18 Effluent Monitoring System bid specification are: ANSI N13.1-69,[2] ANSI N42.18-79,[3] and DOE Order 5480.1A.[4] The DOE Order itself contains certain Radiation Protection Prescribed (as opposed to Recommended) Standards, among them being the "A Guide to Reducing Radiation Exposure to As Low As Reasonably Achievable (ALARA)".[5] Prescribed Standards must be met by all DOE contractors unless an exemption is obtained.

The DOE ALARA Guide contains (in Section 6.2) Monitoring Instrument Requirements, which include requirements for both alpha and beta CAMs. Among the specifications for the alpha CAMs are the requirements for:

a) electronics containing adjustable pulse height analysis circuitry for identification of discrete alpha emitters and effective rejection of radon alphas,

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b) a system counting efficiency of at least 10% of total source emission (4π), and

c) an ability to detect 1 MPC (Maximum Permissible Concentration) of the alpha emitting nuclide within a 8-hour period (for Pu-239 this value is stated to be $2\text{x}10^{-12} \mu\text{Ci/cm}^3$).

For the beta CAMs the ALARA requirements are that:

a) where highly radiotoxic beta emitters are present, features must be present that provide for rejection of counts due to radon daughters such that MPC levels can be detected (e.g., two-channel counting or coincidence counting),

b) there be a system counting efficiency of at least 15% of total source emission (4π) for beta particles of $\geq$ 500 keV energy, and

c) the system has the ability to detect MPC levels within a 2-hour period. (For Sr-Y-90 this is equal to $1\text{x}10^{-9} \mu\text{Ci/cm}^3$).

It should be noted that these ALARA requirements are not fully consistent with the MPC requirements of ANSI N42.18, in that the MPC stated is much less restrictive than those of ANSI for beta emitters: for Cs-137, ANSI requires $5\text{x}10^{-12} \mu\text{Ci/cc}$; for Sr-90, $4\text{x}10^{-12} \mu\text{Ci/cc}$. The stated MPC for plutonium agrees ($2\text{x}10^{-12} \mu\text{Ci/cc}$).

In the bid specifications, the required sensitivity is stated in two different ways. First, the bid specification for the detectors is that they must be able to detect the particular radioactivity of concern with a 4 MPC-hour sensitivity "as set forth in the DOE radiation protection standard" (Presumably 5480.1A). For soluble Pu-239 the MPC is $6\text{x}10^{-14} \mu\text{Ci/cc}$ in uncontrolled areas, and $2\text{x}10^{-12} \mu\text{Ci/cc}$ in controlled areas; for Sr-Y-90 it is $3\text{x}10^{-11} \mu\text{Ci/cc}$ in uncontrolled areas, $1\text{x}10^{-9} \mu\text{Ci/cc}$ in controlled areas; for Cs-137 it is $5\text{x}10^{-10} \mu\text{Ci/cc}$ in uncontrolled areas, and $1\text{x}10^{-8} \mu\text{Ci/cc}$ in controlled areas. However, DOE 5480.1A also requires use

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of exposure levels for on-site personnel, one-fifth of the DOE guides for design purposes. It is not explicitly stated that the "one-fifth" rule applies to concentration guides as well as whole-body external exposure. We presume that it does inasmuch as dose is the primary limit. The 5480.1A implementation guidance makes reference not only to design factors such as occupancy time and shielding, but also "source terms". Moreover, the exposure dose equivalent limits apply to combined internal and external exposure, and hence, to derived limits such as air concentration limits.

It should also be noted that 5480.1A requires following the ALARA Guide as well. The bid specification data sheet summarizes the minimum detectability and response time for the detectors as:

Alpha Particulate: $3x10^{-11} \mu Ci/cc$ in 2 hours
Beta Particulate: $1x10^{-11} \mu Ci/cc$ in 2 hours

Hence, in the bid specifications, the chosen performance specification is apparently the insoluble limit for Pu-238 in air rather than the soluble limit for Pu-239, coupled with a response time of two hours rather than the ALARA time of 8 hours. The resultant bid specification required integral response is then $6x10^{-11} \mu Ci$-hrs/ml versus the Order 5480.1 required $1.6x10^{-11} \mu Ci$-hrs/ml. What emerges from these conflicting standards is the appearance that while the beta emitter detectability requirement is in reasonable agreement with DOE ALARA guidance for planning purposes, the alpha emitter requirement is far less restrictive than it should be. This apparent failure to comply with DOE guidance and orders will be discussed further below.

Sensitivity is also specified in the bid specifications in terms of the response of the respective detectors to activity deposited on filters, which corresponds to system efficiency:

Sr-90 $2.8x10^{5}$ cpm/μCi
Cs-137 $1.4x10^{5}$ cpm/μCi
Pu-239 $4x10^{5}$ cpm/μCi

For the Cs-137 beta emitter the implicit system efficiency is only 6%, or half of the ALARA guidance. More on this later. The Sr-90 efficiency is acceptable. For plutonium, the implicit required efficiency is 18%, which readily meets ALARA efficiency requirements. Of course, the concern for sensitivity is a separate issue. The third ALARA consideration, besides sensitivity and efficiency, is the requirement for dealing with the radon progeny interference and contributions to background.

Although ALARA guidance requires equal concern for radon progeny interference in alpha and beta systems, the bid specifications for the WIPP system treat them very differently. For the alpha system the requirement is that the instrument shall provide a means of subtracting alpha radiation background counts from radon and thoron daughters that interfere with the detection of Pu-239 alpha energies. A two-channel approach is described. The contractor must demonstrate that over-subtraction is not a problem (or provide a failure alarm). These provisions are in agreement with the ALARA requirements. For the beta system, two detectors are required. One is used as the sample detector and the other is to measure "general area background count rate". It is stated explicitly that this second detector will "ignore any count rate generated by the sample itself". This two-detector scheme is completely at variance with the ALARA requirements, which clearly call for rejection of counts due to radon progeny such that MPC levels can be detected. It is not at all clear that the proposed beta background subtraction scheme could function as needed unless the activity on the filter is always a simple and constant proportion of ambient background, (which it is not), nor is it clear why the concern for background subtraction from natural radionuclides collected on the filter should be dropped for this system. A more detailed quantitative evaluation of radon interference will be provided below.

What accounts for these apparent violations of different portions of the DOE Orders in the bid specifications themselves? There are no discussions of the rationale for the selected requirements or the need for waiving the mandatory of ALARA requirements, if such a rationale exists. The DOE Orders do provide for an exemption to the Orders in section 5480.4, which states: "Heads of Field Organizations shall submit to the appropriate

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program Secretarial Offices, a request for permanent exemption from the mandatory standards of this Attachment".[6] To our knowledge, no application for exemption was made prior to issuance of these bid specifications or purchase of the systems. Hence, bid specifications appear to be in violation of DOE Order 5480.4.

1.3 Specifications of the Constructed CAM Instrumentation

Inasmuch as instrument systems have already been constructed (possibly according to modified bid specifications without exemption from mandatory ES4H Standards), it is of interest to determine whether in fact the alpha and beta monitoring systems themselves are in violation of DOE Orders, which is a more serious concern than improper bid specifications. The following system specifications were taken from the contractor's manual (Nuclear Research Corp.) provided with the instrumentation now on-site at WIPP, Chapters V (Detector X-Ray/Alpha MD-70), VI (Detector Beta/Gamma MD-80), and VIII (Ratemeter DRM-200).[7]

1.3.1 X-Ray/Alpha CAM, MD-70 Specifications

The detector for the L x-ray system consists of a calcium fluoride scintillation crystal 1½" in diameter, 1/8" thick coupled to a photomultiplier. The window of the detector is aluminized mylar (2 mil). The specified sensitivity is:

Pu-239: 1.6×10^4 cpm/μCi on filter
Am-241: 1.3×10^5 cpm/μCi on filter

(10 mV per keV of incident radiation when HV is properly set).

It is immediately clear that the implicit detector efficiency (CPM/DPM, 0.7% for Pu-239, is an order of magnitude below both the bid specification and the DOE Order efficiency requirements. This is a serious discrepancy, leaving little room for adjustments to compensate for other deficiencies in the system to be described. There is no specific sensitivity claim made, so direct comparison with the sensitivity standard is not possible.

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A study of the sensitivity of the NRC system when operated in a certain level of salt dust has been made by the Inhalation Toxicology Research Institute (ITRI) over a year ago, and a published report was prepared in October 1987. Unfortunately, this report has not been made available to EEG for review in spite of repeated requests. Based on personal communication with the principal investigator of this project, the NRC instrument was able to satisfactorily detect the modified bid specification required plutonium activity (presumably $3 \times 10^{-11} \mu Ci/ml$ sampled at 1 CFM for 2 hours, or $1 \times 10^{-4} \mu Ci$) deposited on a filter and covered with a layer of salt dust equivalent to sampling 24 hours at $2mg/m^3$. However, by depositing both the plutonium and the salt aerosols in a very brief period of time rather than the normal 24-hour sampling period, and not monitoring for the presence of radon/thoron daughters, the continued ability to perform in the presence of background activity on the filter remains untested and unknown as far as can be ascertained at this time. Later in the report we calculate expected performance based on assumed concentrations of radionuclides and pump rates.

The third requirement, background subtraction, is briefly described in the manual. In principle, two channels of the processed detector signal are monitored simultaneously, an x-ray channel centered at 17 keV with a 50% window, and a background channel centered at 40 keV with a 28% window. Two minute counts are made in these two energy channels, individually averaged and stored. The difference between these averaged counts is calculated, stored and output as the current net count-rate. The stored output is used in a microprocessor controlled curve-fitting routine to calculate rate of rise or rate of decrease.

No specific mention by the manufacturer is made in the user manual chapter on background subtraction of the possibility that radon progeny interference could be a problem or would be corrected by the above described system. Until the system is tested in the laboratory under controlled radon/thoron progeny concentration conditions, and also field tested, the expected response to radon interference must be evaluated by simulation.

1.3.2 Beta/Gamma CAM, MD-80

The detectors for this system consist of 2 GM tubes, one oriented toward the sampling filter paper sensing both background radiation and radiation from the accumulated particulate matter. The second GM tube is oriented outwards, sensing only ambient radiation, thus this design does not implement the bid specified design which was critiqued above. The specified sensitivity for Cs-137 is 3×10^5 cpm/μCi, implying an efficiency of 13.5%. This efficiency readily meets the bid specification. Once again, no detection sensitivity is specified, so a calculation will have to be made (below). The ratemeter circuitry and background subtraction logic are said to be the same as described for the x-ray detector.

As stated in the overview, there is no reason to believe that the proposed background subtraction scheme will function to effectively subtract the background signal generated by radon daughters accumulated on the filter. It is designed to subtract a signal generated by ambient radiation, but this is not equivalent to subtraction of the signal from beta/gamma flux produced by the background radionuclides collected on the filter itself as required by the DOE Orders.

1.3.3 Calibration

An additional component of background subtraction system performance is calibration and setup. In Chapter 10 of the NRC manual[7] the following claim is made:

> "The alpha (x-ray) channel uses an Am-241 gamma source of nominally 1 μCi which emits L x-rays of nearly identical energy and relative yield to those of most transuranic nuclides, including Pu-239". (Section 10.2)

If true, this claim would fulfill the requirement of ANSI N13.10 that a calibration radionuclide shall permit calibrating the range of energy and rate capabilities intended for the system. However, the claim is not supported by reference to data. According to current compilations,[8] the

following data describe the energy and yields of important transuranics expected in typical drum wastes:

Table 1. Transuranic Radionuclide X-Ray Emissions

Radionuclide	Typical Waste Ci/Drum	X-Ray, Gamma Energy(keV)	Yield(%)
U-233	1.6×10^{-3}	13	3.9
Pu-238	3.5	13.6	11.6
Pu-239	0.4	13.6	4.4
		17.2	- - -
Pu-240	8.8×10^{-2}	13.6	11.0
Am-241	1.3	13.9	43.0
		17.2	- - -
		26.3	2.4
		59.5	35.9
Cm-244	9.8×10^{-2}	14.3	10.3

It would appear, therefore, that while the energy of Am-241 is appropriate, the yield is substantially higher than any of the others, about 10 times that of Pu-239. So in fact, the proposed calibration source is at variance with the requirements of DOE Orders. At the same time, this large discrepancy creates a condition whereby a signal from a deposit of pure Pu-239 on a filter could be interpreted to be 1/10 its actual value (i.e., the calibration assumes the deposit is pure Am-241).

The calibration of the x-ray detector and analyzers requires that the L x-rays of Am-241 be detected as efficiently as possible, but at the same time that the 59.5 keV gamma emissions are not efficiently detected in the background channel. Chapter 13 calibration procedures in the NRC[7] manual call for adjusting the x-ray detector high voltage, and thus its gain, to center the primary (x-ray) channel at 17 keV and place the background channel between the x-ray and the 59.5 keV gamma emissions. The calibration checkpoint is that the count rate in the x-ray channel is within 10% of the factory calibration data sheet value for the given detector, i.e., nominally 130,000 cpm if the calibration source is 1 μCi and the efficiency is as high as claimed. However, the instructions suggest the peak count rate should be about 40,000 cpm, with no apparent reason given for it being lower.

It is evidently critical that an Am-241 source be used for this setup since a maladjustment of gain could cause the system to fail to detect Am-241 by cancellation of counts in the two channels (i.e., 59.5 keV in the upper channel and 13.9 keV plus 17.2 keV in the lower channel with about the same yield). At the same time, it is clear that small drifts in gain or peak broadening in the CaF_2 detector could have large effects on performance. It should be noted, for later reference, that the factory calibration and field setup and calibration makes no mention of whether peak broadening associated with the detection of 59.5 keV photons does contribute substantially to the background channel. If it did, the performance of the system as a whole would be compromised. As it is, the reported sensitivity of the detector when expressed on a per-photon basis suggests a difference between Am-241 and Pu-239, which indicates an interference does exist. Further details of the analysis of the detector are provided in Chapter 2. These details are of importance in the discussion below of radon daughter interference, where it is critical to judge whether background subtraction at the energies involved in the two channels will work.

1.3.4 Background Correction

A final note on calibration and setup concerns the adjustment of the background channel. The manufacturer's instrument manual,[7] Chapter 13 (13.1.1.4) claims that the manufacturer-provided check source (Ba-133) "produces the same spectrum as external gamma background." In fact,[8] the spectrum is as follows:

Table 2. Barium-133 X-Ray Spectrum

Energy (keV)	Yield (%)
4.29	17.0
30.63	34.2
30.9	63.4
35.0	22.8
53.0	2.1
79.0	2.6
80.9	33.0

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During calibration of background subtraction, this source is inserted next to the x-ray detector, and the background channel window is adjusted such that the x-ray channel cpm and the background channel cpm are identical to within 5%. The question raised by use of this source and procedure is: How can a flux of photons with energies in the background channel (30-35 keV) with a yield of about 100% be made to "agree" with the flux of photons from this source detected in the x-ray channel (8-26 keV) when apparently there are no photons matching the Pu/Am L x-rays energies in the spectrum and the yield of the only low-energy x-ray is 17%? One possibility is that the x-ray channel is receiving photons from extraneous sources (especially electronic noise). Another possibility is that a portion of the next lower energy photons of this set (~31 keV) are being detected in the x-ray channel, while the higher energy ones are being counted in the background channel. This seems to be the most likely explanation due to the expected peak broadening of this type of detector. In any case, it does not appear that the prescribed procedure is a simple calibration using a source which directly contributes photons of proper energy to both the x-ray channel and the background channel, as implied in the manual. This is a serious flaw in the instrument setup since it indicates that background subtraction relies on a fixed ratio subtraction based on Ba-133 emission energies.

One thing certainly true about the prescribed background channel adjustment is that it does nothing to directly address background introduced by radon and thoron decay products collected on the filter paper. In particular, as will be shown below, the spectrum of x-rays from these decay products does not at all match the Ba-133 spectrum. Hence, any fixed correction of the expected direct contribution of L x-rays from background emitters on the filter to the x-ray channel would be fortuitious, if it occurred at all.[1] Here, clearly, is a violation of the intent of both the bid specifications and the DOE ALARA requirements, in those sections previously mentioned in which specific correction of the interference by radon progeny is required.

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[1] The actual L x-ray background signal will be a dynamically changing one due to the accumulation of radon and thoron progeny at a rate proportional to the ambient concentrations and sampling rate. This process will be described in detail below in Chapter 2.

The absence of specifications of procedures for dealing with radon/thoron L x-ray background effects in the NRC manual (and in the Bechtel bid specifications) suggests ignorance of the fact that certain radon and thoron progeny emit L x-rays.

Background correction in the beta CAM is similarly crude, if not more so. In this case, the background detecting GM tube is designed to be oriented such that it receives input only from the ambient beta-gamma field, and not from the filter itself. After signal averaging, the background and filter detector outputs are subtracted to yield a net count rate for each count interval. There is no separate radon daughter detection channel, and moreover, none is possible in principle because a GM tube generates a fixed pulse height with energy. Thus, the beta CAM background subtraction process cannot track the dynamic changes in background produced by the beta-emitting decay products of radon and thoron as required by DOE Orders.

1.4 Discussion of CAM Design

In sum, both the alpha CAM and the beta CAM design concepts appear to incorporate features and performance characteristics which represent serious departures from regulatory requirements and good monitoring practice. The approaches to background subtraction in both instruments are particularly questionable. The beta CAM background suppression technique is most unsatisfactory since it is incapable of discrimination by energy, nor it is sensitive to background accumulation on the sample itself. This instrument will not be analyzed further.

Although the alpha CAM does possess energy discrimination capabilities and uses a two-channel background suppression approach, it too has a number of undesirable characteristics, as has been suggested. These characteristics and the problem of correction of radon and thoron progeny collected on the sample will be discussed below. Additional issues related to the potential distortion of an aerosol sample during collection in the CAM head will be addressed as well.

2.0 L X-RAY DECTION BY THE WIPP ALPHA CAM

Although the DOE specifications for the CAM systems at WIPP clearly require
counting alpha emissions of the radionuclides of concern, the actual system
constructed and installed at WIPP utilizes the detection of L x-ray
emissions. In the following discussion the implementation of this
detection scheme will be carefully evaluated and discussed.

2.1 Detector Design Considerations

The detector built for the alpha CAM is a thin scintillator type consisting
of a 1/8" thick CaF_2(Eu) crystal coupled to a photomultiplier tube. The
use of a thin crystal is a common approach to the problem of detecting low-
energy gammas and x-rays in the presence of higher energy photons, called a
"Fidler" detector. High-energy photons pass through the crystal,
depositing relatively little energy, while the low-energy photons are
completely absorbed. This characteristic can be inferred from the linear
attenuation coefficient of CaF_2 as a function of energy. Only at energies
above 100 keV, is absorption dominated by Compton scattering events. The
Compton absorption coefficient is relatively small compared with
photoelectric absorption coefficients at lower energies. At the L x-ray
energy of the transuranic radionuclides (13-20 keV) the photoelectric
absorption coefficients are large, so that any L x-ray photon interactions
with very thin crystals will result in complete photoelectric absorption,
but Compton events will be rare.

While it is evident that the detector thickness is quite appropriate, the
choice of scintillator material is not so clearly appropriate. The light
output of CaF_2 is only one-half that of NaI scintillator material. Also,
the decay constant of the light pulses from CaF_2 is over four times longer
than it is for NaI. Both of these characteristics are a disadvantage for
the proposed system. The principal advantages of CaF_2, that is, in non-
hygroscopic, and rugged, are not serious concerns in the proposed WIPP
application since the scintillator can be readily sealed with a thin
beryllium window to admit low-energy photons and would not be subject to
severe mechanical shocks in normal use.

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The indications are that apparently a scintillator used for other applications where mechanical ruggedness and non-hygroscopic characteristics offset the relative disadvantages of this crystal was put to use in the WIPP application without full review. Experts contacted by EEG[9] could not identify any advantages the use of CaF_2 would have for the WIPP application, and in fact, worried that the above mentioned light emission characteristics would lead to degraded instrument performance attributable to peak broadening effects and to the necessity to amplify small pulse output from the photomultiplier which would, in the process, amplify electronic noise as well. The consensus of several expert opinions was that $CaF_2(Eu)$ was a poor choice of scintillator.

2.2 Monte Carlo Simulation of Detector Response

In order to further explore the characteristics of the proposed detector and sample configuration in the WIPP CAM head, a Monte Carlo simulation model was developed and applied to what is currently known about the WIPP CAM. Two basic response characteristics have been explored with the model. First, due to the fact that the WIPP CAMs must sometimes function in very dusty environments, the possibility of instrument response degradation by buildup of salt aerosols on the filter was considered. Second, the characteristic energy dependence of response of a thin CaF_2 detector was investigated in the process of evaluating the overall response characteristics of this instrument, particularly with regard to background interference. The results of energy dependence modeling were then combined with a model of the process of accumulation of the decay products of radon and thoron on a filter to predict overall instrument performance.

2.2.1 Photon-Detector Interaction Model

The basic geometry of the sampling head from the perspective of photon transport from sample to detectors is shown in Fig. 1. A sample, of thickness, D, is assumed to have been deposited on a filter. An air space of height H separates a detector of radius R from the sample surface. The sample surface is considered to be the origin of an X, Y, Z coordinate

DETECTOR GEOMETRY FOR X-RAY TRANSPORT

L X-ray

Detector

xp,yp,zp

Inlet

H

D

xs,ys,zs

x1,y1,z1

Sample

Filter
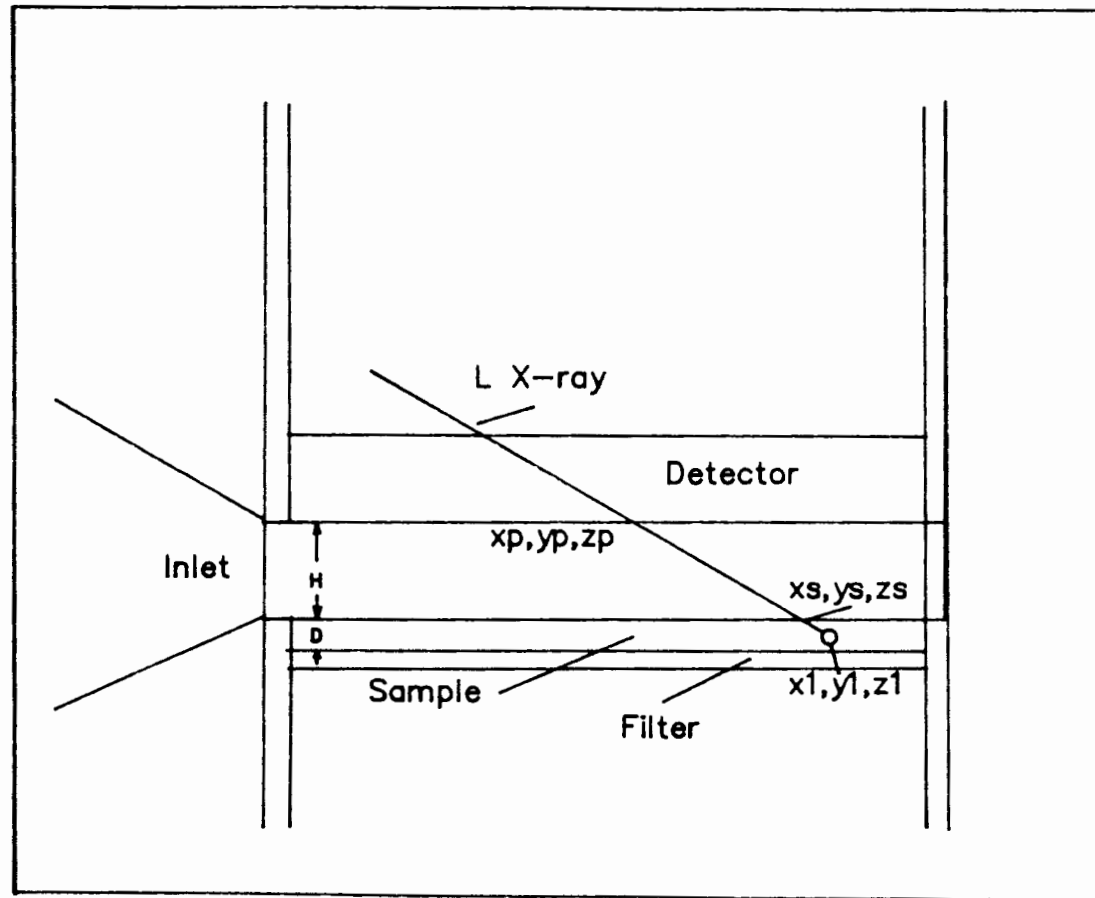
Figure 1

system for purposes of tracking photons emitted in the sample volume up to the detector. The Monte Carlo simulation is simplified to include only the total absorption probability of the entire trajectory and not multiple scatter events. For low-energy photons this is a reasonable approximation to the actual process. Thus, the simulation basically samples the trajectory length from which the expectation value of total absorption can be calculated.

In outline, the computation proceeds as follows for each photon history once the geometry is defined:

1) Randomly assign values to the coordinates of a point within the sample volume from which a photon is released, $P_1(X_1,Y_1,Z_1)$, where $0 \leq X_1 \leq R$, $0 \leq Y_1 \leq R$, $D \leq Z_1 \leq 0$.

2) Randomly assign direction cosines to the photon trajectory: dx, dy, dz.

3) Consider a line L through $P_1(X_1, Y_1, Z_1)$ in the direction dx, dy, dz: all other points $P(X, Y, Z)$, if on this line, are such that the distance in the direction P_1 P is $X - X_1$, $Y_1 - Y$, $Z - Z_1$, proportional to dx:dy:dz; hence,

$$X - X_1 = t\ dx,\ Y - Y_1 = tdy,\ Z - Z_1 = tdz$$

where t is a proportionality factor such that the distance P_1 P is proportional to t. For the condition that $Z = H$ (at the detector):

$$H - Z_1 = tdz,\ \text{hence } t = (H - Z_1)/dz$$

4) Then at $Z = H$, the test of whether a trajectory line passes through the detector, is whether or not the X, Y coordinates of that point are such that

$$X_p \leq \sqrt{R^2 - Y_p^{\ 2}}$$
$$Y_p \leq \sqrt{R^2 - X_p^{\ 2}}$$

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5) Similarly, at the air interface ($Z = 0$) in the direction P_1 P, for a trajectory point at the sample surface with coordinates X_s, Y_s:

$$X_s - X_1 = t'dx, \ Y_s - Y_1 = t'dy, \ Z_s - Z_1 = t'dz \quad ,$$

where

$$Z_s = 0 \quad ,$$

hence,

$$t' = Z_1/dz \quad .$$

6) Then generally, the pathlength through the sample, S_1 is:

$$S_1 = \sqrt{(X_s - X_1)^2 + (Y_s - Y_1)^2 + (Z_s - Z_1)^2} \quad ,$$

and the pathlength through the entire distance to the detector S_2 is:

$$S_2 = \sqrt{(X_p - X_1)^2 + (Y_p - Y_1)^2 + (Z_p - Z_1)^2} \quad ,$$

hence, the pathlength through the air alone is

$$D_{air} = S_2 - S_1 \quad .$$

A similar pathlength through the detector itself is also calculated.

7) These distances are then converted to density thickness (g/cm^2), and the probability for absorption calculated for that trajectory using energy dependent linear absorption coefficients for the sample (salt), air, and the detector (CaF_2).

8) The relative number of photons arriving at the back side of the detector represent photons not absorbed and hence not detected. Hence the

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energy and geometry dependent efficiency of the crystal can be estimated. This again is only an approximation since it does not take account of Compton scattered photons that escape, but it is a good approximation for low-energy photons.

9) Finally, for each photon trajectory through the sample and through the air, a fractional expected count can be calculated and corrected by detector efficiency. The summed count then represents the expected response leading to an overall system efficiency (counts/100,000 photon histories).

Simulations with the above described code have produced several results of interest. A buildup of salt on the CAM filter is not likely to produce serious degradation of detector response in those CAMs incorporated into the stack monitoring system (operating at 1 CFM and a dust loading of $2mg/m^3$), provided that the salt is deposited in a uniform layer over the filter. Impaction of larger size particles, as described elsewhere in this report, could become a serious problem. Also, non-uniform deposition may occur which could affect performance. This finding is consistent with informal communication of the results of the ITRI study of salt buildup interference in the WIPP CAM, which was that only a slight reduction in sensitivity occurs even if the TRU activity is completely buried under a layer of salt dust at the maximum dust loading expected in the exhaust duct.

This result, however, has to be interpreted with some caution. For the case of the underground CAMs, dust loadings could be much more severe, based on the early ITRI investigations.[10] To our knowledge, no recent detailed studies of underground dust loadings at a variety of potential CAM locations have been made. Accordingly, it is not possible to state with complete assurance that the even deposition of salt dust on the CAM filters at underground locations would not be expected to interfere with detector response. We urge that appropriate measurements be made with the CAM to check the effects of higher dust loading expected at a variety of underground locations on CAM performance.

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The possible effects of non-uniform deposition on detection efficiency were explored with the L x-ray model. The results, to be discussed in Chapter 4 below, indicate that, as expected, the efficiency for detection of photons drops dramatically as the source area is further and further from the center of the filter. For that portion of the sample (the larger-size particle fraction) that is non-uniformly deposited near the far wall of the CAM head by inertial processes, detection will be far less efficient than for the smaller size fraction deposited near the center. Hence, the detector efficiency is expected to be particle size dependent. Certainly, the published efficiency or field determined efficiency obtained by use of a uniform plated source inserted in the detector is not a suitable representation of the true detector response for the same radionuclide attached to a wide range of particle sizes.

The modeled energy dependent response of the detector indicates that indeed there is some loss of efficiency in the transuranic L x-ray energy range (10-20 keV) due to absorption, revealed as a slight decrease in efficiency in the very low energy range compared with peak response. The effect seems to be small (see Fig. 2). However, the efficiency falls off in the energy range of the background window even more, and is considerably reduced at the 60 keV energy of the Am-241 gamma emission. All higher energy photons would not be expected to contribute appreciably to the gross count due to very low absorption probability.

The results of the simulation shown in Fig. 2 provide a means for checking the overall accuracy of the simulation through intercomparison with the reported nominal sensitivity of the NRC detector. The reported value of sensitivity for Pu-239 is 1.6×10^4 cpm/μCi. When corrected for the 0.04 L x-ray yield, this corresponds to an efficiency of 18%, which compares very well with the simulated 17.6%-21.5% efficiency values in the energy range 10-20 keV. The approximate 2% higher simulation estimate very likely reflects the effect of background subtraction which would tend to slightly lower the net count per disintegration. The L x-ray simulation model appears, therefore, to be quite satisfactorily validated against the reported nominal Pu-239 sensitivity.

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MONTE–CARLO SIMULATION OF L X–RAY DETECTOR
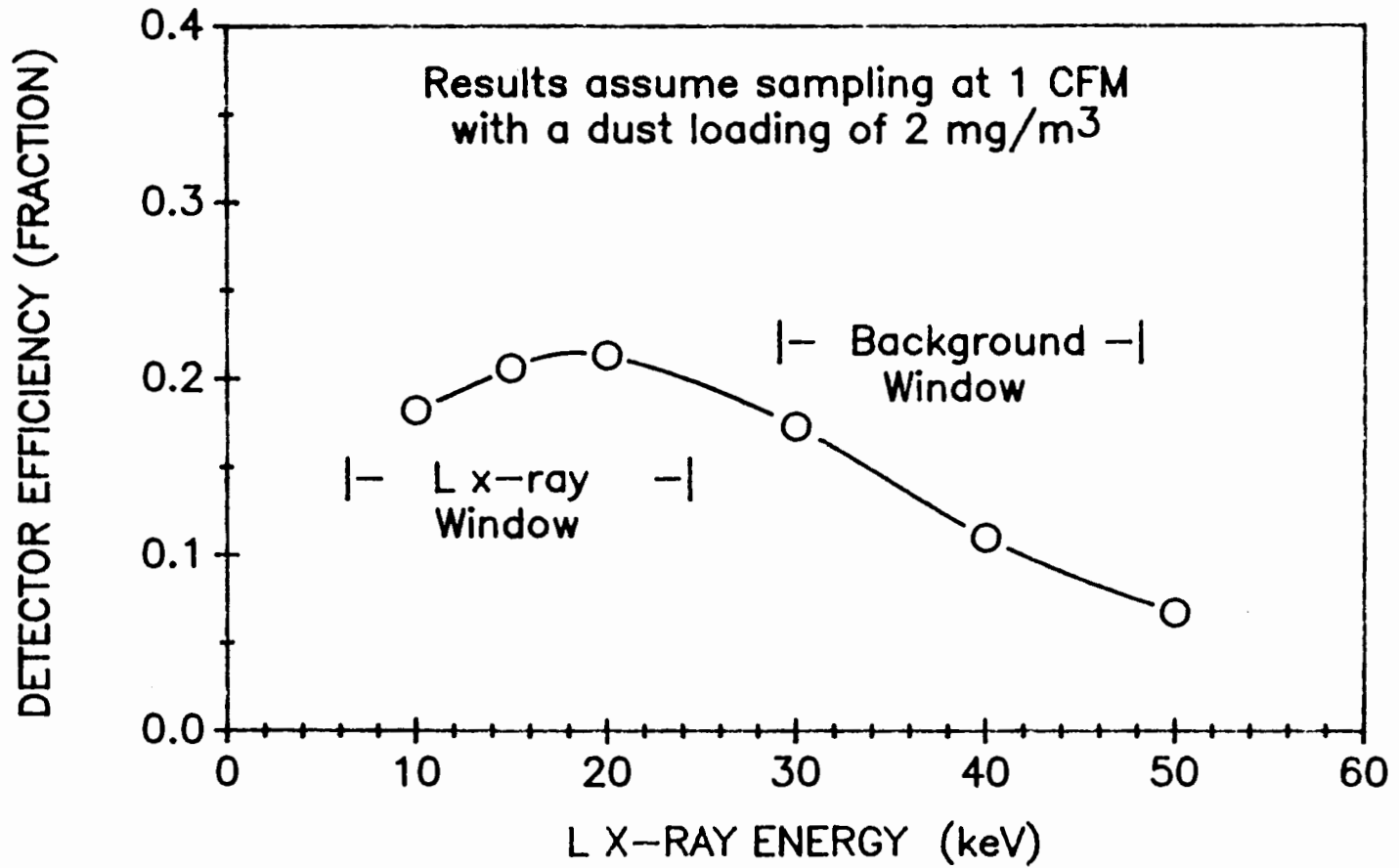EFFICIENCY AS A FUNCTION OF ENERGY

Results assume sampling at 1 CFM
with a dust loading of 2 mg/m^3

Figure 2

However, when the reported nominal sensitivity of Am-241 (1.3 x 10^5 cpm/μCi) is corrected for yield (0.43 to 0.64), the resultant efficiency for detection by L x-ray emissions is between 9.2% and 13.6%, depending on the value of yield used. Inasmuch as the L x-ray energies of Am-241 are nearly identical to Pu-239, this difference in efficiency requires further discussion.

One likely explanation for the reduction in efficiency in the detection of Am-241 L x-ray photons (identical in energy to the L x-ray photons of Pu-239) is that peak broadening in the detection of the 59.5 keV gamma (yield of 35.9%) from Am-241 decay causes some of these pulses to spill over into the background subtraction channel (28% window centered at 40 keV). Thus, an additional background baseline subtraction occurs with Am-241 resulting in lower efficiency. The resolution of the 59.5 keV peak is approximately 42%. For the 17 keV peak it is approximately 70%. Peak resolution is progressively worse for lower energy photopeaks. These quite poor resolutions are approximately 1.4 times larger than comparable NaI(Tl) photopeak resolutions at the same energy, and are attributable to the 50% less light output of CaF_2(Eu) relative to NaI(Tl). The upper energy of the background channel corresponds roughly to the lower energy edge of the expected 59.5 keV peak at half-maximum. Ten to twelve percent of the counts in the photo peak could be counted as background. The significance of this discussion of peak broadening effects lies in the implication of such poor resolution for background subtraction, not the apparent reduction in Am-241 detection efficiency, which is not large.

In sum, the L x-ray detector simulation model has been validated against the manufacturer's specification for Pu-239. It predicts serious non-linear response for non-uniform deposition of activity across the filter surface. The detector model also predicts a lower detection efficiency for higher energy photons (above about 60 keV) and peak broadening effects in the CaF_2 scintillator medium which lead to poor peak resolution, and the need for very wide energy window settings to capture a substantial portion of the photo peak at low energy. Thus the potential for enhanced difficulties with background subtraction are traceable directly to a poor choice of scintillator material.

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2.2.2 Model of X-ray Activity on a Filter During Continuous Sampling

In order to investigate the dynamics of radon and thoron daughter accumulation on a filter during continuous sampling, a model of the process was developed based on the solution of the coupled differential equations describing the rate at which radon and thoron progeny atoms accumulate on a filter during sampling.

$$dN_i(t)/dt = q_i V + \lambda_{i-1} N_{i-1}(t) - \lambda_i N_i(t)$$

where

$$i = \begin{array}{l} 1 \text{ for Po-218} \\ 2 \text{ for Pb-214} \\ 3 \text{ for Bi-214} \end{array}$$

N_i = number of ith type atoms on filter

λ_i = decay constant of ith type atom (min^{-1})

q_i = air concentrations of ith type (atoms/liter)

V = sampling rate (liters/min)

The model is generalized to an extent to allow for the possibility that the radon/thoron background source terms change during sampling. The strategy of the code is as follows:

1) Concentrations of the radon progeny (RaA, RaB, and RaC) thoron progeny (TnA, TnB, and TnC), and transuranic radionuclides (Pu-238, Pu-239, Am-241) are entered into the code.

2) If the sources are time varying over the interval of continuous sampling a slope can be specified.

3) By successive calls to a subroutine for computing activity of each progeny on the filter as a function of time, the activity of each is continuously tracked, including the effects of branching.

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4) Using appropriate yields of alpha emission and photon emission and appropriate detector efficiences, alpha activity (CPM) and L x-ray activity (CPM) due to each source are computed and output as a function of time.

An experimental verification of the model was performed by EEG at the WIPP site in December 1987. The experimental protocol called for: a) collecting a high volume (4 CFM) sample for the determination of radon daughter concentrations by the modified Tsivoglou Method at the beginning and end of the experiment, b) collection of a continuous high volume (4 CFM) sample interrupted at approximately 3 minutes, 10 minutes, 30 minutes, 1 hour, 2-1/2 hours, and 4 hours for 3-minute gross alpha counts, and c) a gross alpha count taken 29 counts after the last sampling interval to permit decay of radon daughters and determination of thoron daughter activity.

The results of these measurements are shown below.

Table 3. Continuous Monitoring Simulation Experiment Results

Measured Radon Daughter Concentrations

Time	RaA(pCi/l)	RaB(pCi/l)	RaC(Ci/l)
12/9 0835	0.185+0.056	0.179+0.026	0.117+0.023
12/9 1400	0.057+0.044	0.099+0.021	0.102+0.019

- -

Measured Gross Alpha Activity
on Continuous Sample Filter

Time from Start to End of Sample (min.)	3-Minute Gross Alpha Counts
3	101
10	243
30	688
60	1105
150	1564
240	1373

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The results of the model simulation of gross alpha counts as a function of time, given the conditions of the experiment, are shown in Fig. 3 along with the measured data points. Note that a declining radon daughter source term was included based on a simple linear fit to the observed rate of decline over the four-hour sampling interval. As would be expected, the observed counts during the early phase of ingrowth are somewhat lower than the model predicts due to the frequent interruption for counting which caused some loss of activity by decay. Overall, the agreement between model predictions and the experiment is quite good. A correlation plot (Fig. 4) between measured and modeled counts shows the expected offset, but a slope of 1, and $R^2 - 0.96$.

Given a validated model of the process of accumulation of background activity on a continuous air sample, the results of the predicted efficiency of the L x-ray detector as a function of energy can be combined with this model to produce estimates of expected x-ray count rates from the CAM as a function of time under normal conditions (i.e., normal background concentration of radon/thoron progeny), and under conditions when TRU airborne activity is present and accumulating on the filter as well.

2.3 Radon and Thoron Daughter Background Interference

The apparent reason why the alpha instrumentation specified in the original WIPP bid specification for the CAM was not built was due to the difficult problem of meeting the requirements for dealing with the alpha interference from radon and thoron daughters collected on the filter from ambient sources. The presence of a large amount of salt dust in most of the CAM applications assures that a degraded spectrum of alpha energies overlapping the TRU alpha energies will result.

As was mentioned in the introduction, the apparent operating assumption in the switch to the L x-ray detection mode was that the radon and thoron daughters do not emit L x-rays in their decay processes. In fact there are a number of radon and thoron progeny which emit L x-rays of sufficient energy and intensity to present a serious interference problem.

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MODELED AND MEASURED GROSS ALPHA COUNTS
FROM CONTINUOUS AIR SAMPLING

Modeled —

Measured

Sampling conditions:
Thoron progeny = .0029
RaA,RaB,RaC = .i85: .179: .1174
Flow rate= 4 CFM

3-minute counts at each point

Declining radon daughter source slope:
    RaA = -3.94e-04
    RaB = -2.42e-04
    RaC = -4.89e-05
(units: pCi/l/min from start of sample)

3-MINUTE COUNTS

TIME FROM START OF SAMPLING (min)
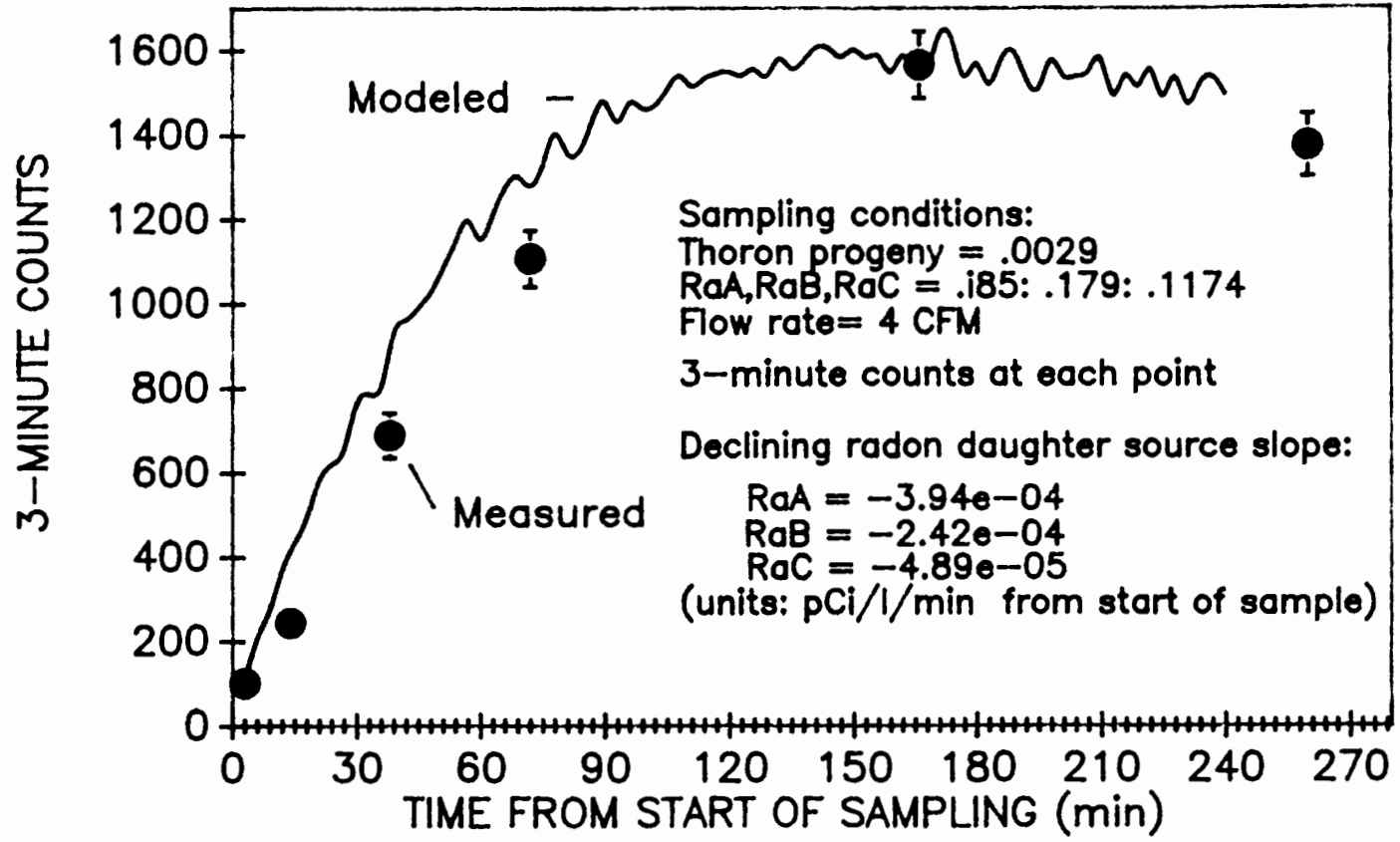
Figure 3

PREDICTED VS. MEASURED GROSS ALPHA COUNTS

Regression (R ^{2}= 0.96)

3-MINUTE COUNTS (MEASURED)

3-MINUTE COUNTS (PREDICTED)
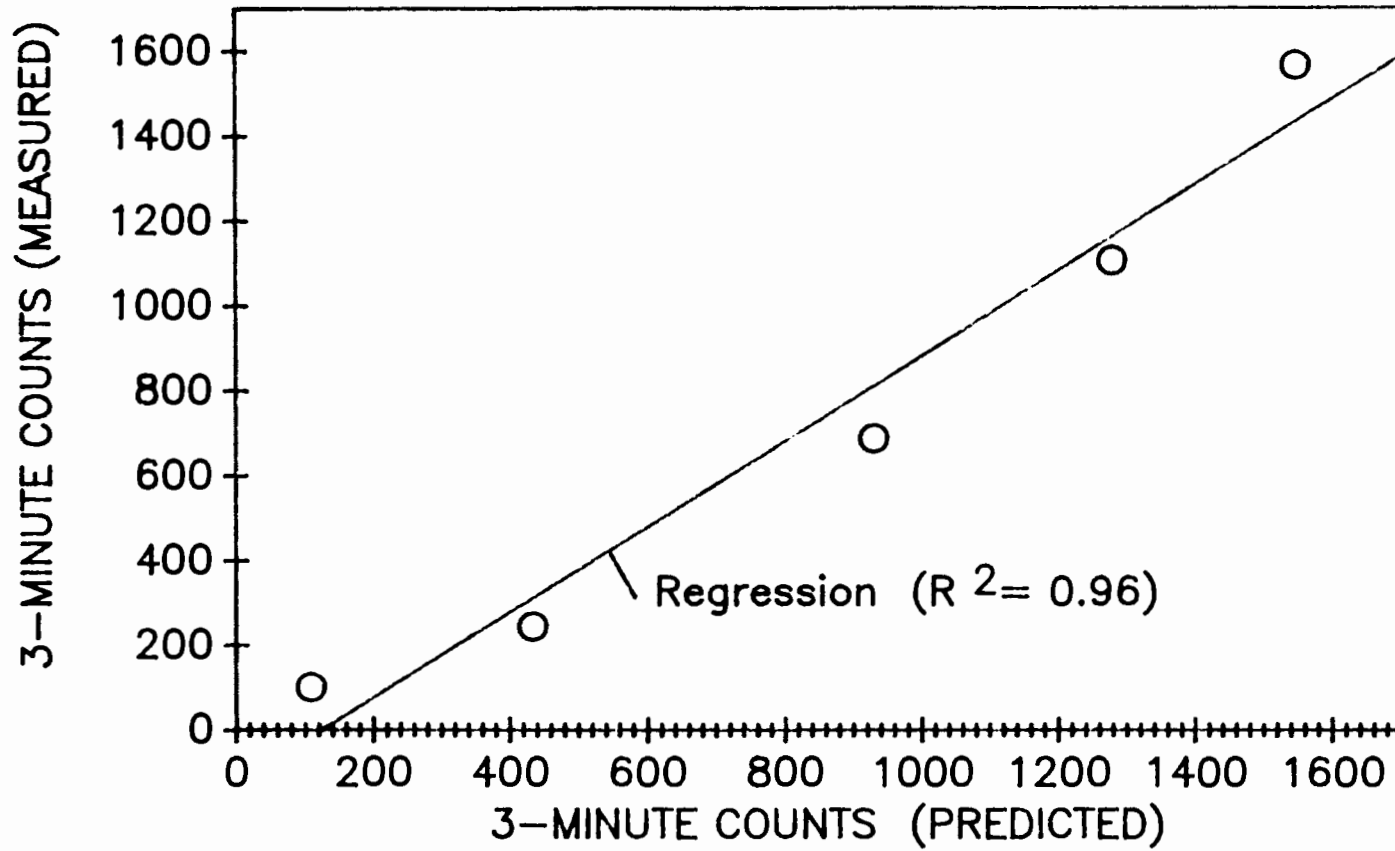
Figure 4

Table 4. Radon and Thoron Decay Product L X-Ray Emissions

Radionuclide	L X-Ray Energy (keV)	Yield (%)
Tl-208	10.6	2.9
Bi-212	10.3	7.7
	39.86	1.02
Pb-212	10.8	15.5
	74.81	10.7
Pb-214	10.8	13.5
	53.22	1.1
Bi-214	11.0	0.52

At first glance, it might be thought that since the lower energy photons of these radon/thoron daughters are all near 11 keV, which is several kilo-electron volts below the lowest Pu/Am L x-ray energies (13.6-26.3 keV), they would not be detected given the specified energy window of the detector. However, peak broadening on the order of 70% is sufficient to more than adequately compensate for the few kilo-electron volt difference in energy between plutonium and radon/thoron progeny L x-rays. The best resolution of the question of sensitivity to radon daughter L x-rays is an experimental test.

Although the WIPP instrument was not available for EEG study, an NRC plutonium survey instrument which uses an apparently identical CaF_2 detector was tested to determine whether such a detector with an analyzer set to detect plutonium x-rays would also detect radon daughter x-rays. The experiment was conducted as follows:

a) The detector was fitted with about 2-1/2 inches of lead shielding to reduce the background count rate to about 6 cpm.

b) A five-minute sample of ambient aerosols was taken to determine the concentrations of the three radon daughters (Po-218, Pb-214, and Bi-214) using the modified Tsivoglou Method. The results were typical background levels (0.196 $\pm$ 0.0475 pCi/l, 0.212 $\pm$ 0.022 pCi/l, and 0.162 $\pm$ 0.20 pCi/l, respectively).

c)   Then a 20-minute sample was taken at 6 CFM (169.9 1/min) to provide an equilibrium sample for analysis. The sample was placed close to the CaF_2(Eu) detector and counted for one hour to provide a characteristic decay curve. A plot of the data is shown in Fig. 5. The resultant half-time for decay of 37-minutes is a definite indication of the detection of radon daughters by their x-ray emissions.

d)   From sampling theory, the activity A on a filter after sampling for t minutes in an atmosphere with concentration C, at a rate Q, is given by:

$$A = CQ \ (1 - \exp(-\lambda t))/\lambda$$

where λ is the disintegration constant of the radionuclide. Given the measured concentrations of RaB (Pb-214) and RaC (Bi-214), the second and third radon daughter activities on the filter shortly after counting, based on the above equation, would have been approximately

$$A_B = 1164 \text{ pCi}$$

$$A_C = 719 \text{ pCi} \quad .$$

Converting to L x-ray activity using the photon yield per disintegration:

$$LX_B = (0.135)(1164)(2.22) - 349 \text{ photons/min.}$$

$$LX_C = (0.0052)(719)(2.22) - 8 \text{ photons/min.}$$

for a total of 357 photon/min. of x-ray activity. The measured activity on the filter was 82.2 cpm averaged over the first 5 minutes. The implied efficiency for radon daughter L x-ray detection is therefore

$$EFF_R = 82.2/357 - 0.23 \text{ cpm/dpm} \quad .$$

e)   A plutonium calibration source of 4.42×10^6 DPM was placed in front of the detector and counted several minutes yielding a count rate of 9681 cpm. The plutonium counting efficiency is then determined to be

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L X—RAY COUNTS FROM RADON DAUGHTERS
CaF$_2$ DETECTOR

RaA = 0.196 pCi/l
RaB = 0.212 pCi/l
RaC = 0.162 pCi/l

70 minute sample at 6 CFM

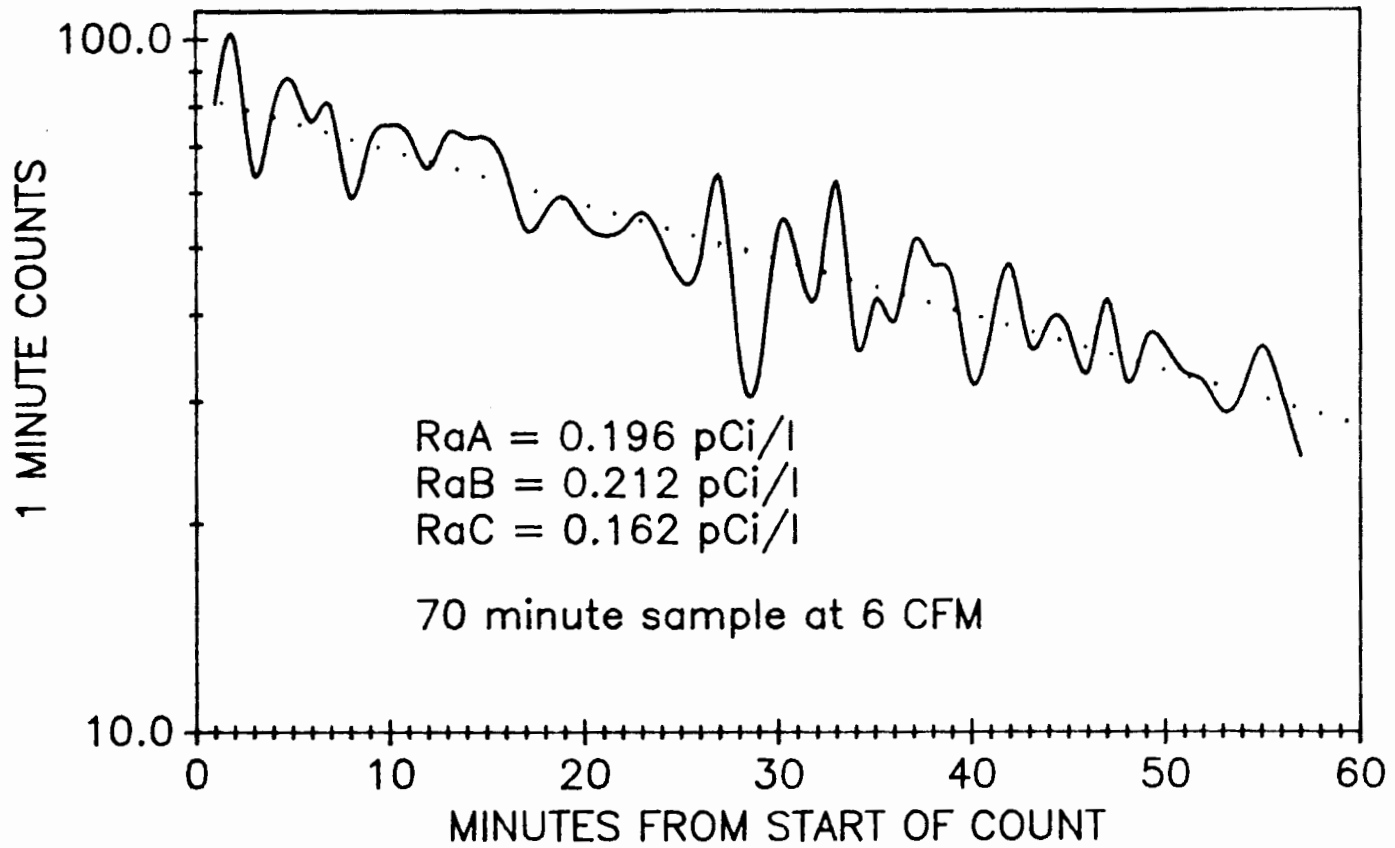Figure 5

$$EFF_P = 9684/4.42 \times 10^6 \times 0.04 - 0.05 \text{ cpm/dpm} \quad .$$

Therefore, for this detector, the efficiency for detection of radon daughters was not the same as the detection of plutonium L x-rays, but may have been actually several times better, depending on whether or not the geometry was the same.

The conclusion of this experiment is that the CaF_2 detector in the WIPP CAM will be quite capable of detecting the L x-ray flux from radon daughters collected on the CAM filter and perhaps even more efficiently than L x-ray flux from TRU radionuclides.

It might be argued that the WIPP instrument is more sophisticated than a survey instrument due to the use of an independent background counting channel. However, as the tabulation above shows, only Bi-212 has an x-ray with energy close to the background channel window (39.85 keV). But the yield is only 1.2% which is an order of magnitude lower yield than several of the other x-ray emissions with energies falling within the plutonium energy window. Another possibility is that due to peak broadening, the 74.8 keV photons from Pb-212, and 53.22 keV photons from Pb-214 may also contribute to background subtraction. However, due to the lower detection efficiency of higher energy photons, this effect is likely to be small. Hence, it is improbable that the detection of the L x-rays from radon/thoron daughter activity on the CAM filter will be self-correcting.

In summary, the energies and yields of x-rays of the radon and thoron daughters indicate that their L x-ray emissions could very easily be detected in the plutonium energy window of the WIPP CAMs. Experimentation with a similar instrument with the same detector used for plutonium survey purposes indicates that indeed efficient radon daughter x-ray detection capability is present. And finally, the absence of a high yield of x-rays in the energy range of the background channel means the interference will not be self-correcting. Remaining questions are whether or not the interfering background activities are present in significant concentrations in ambient and underground air, and what level of interference is likely to be experienced in the dynamic process of collecting airborne activity on a filter.

2.4 Predicted Consequences of Background Interference on CAM Performance

The foregoing assessment of the L x-ray CAM instrumentation indicates that very significant interference from naturally occurring background radionuclides will occur. In an effort to predict how severe this interference could be, a series of simulations were conducted using the output of the CAM detector model to predict the energy dependent detection efficiencies in the x-ray window (and background window), and the sample collection model to predict accumulated activity. The results were combined to predict 3-minute x-ray counts during a continuous sampling interval of 24 hours (1440 minutes).

The sampling scenarios were chosen to illustrate how the L x-ray count rate from a TRU airborne release would compare with the count rate from background radionuclides collected on a filter once sampling had been underway for several hours. Two cases were chosen for illustrative purposes.

2.4.1 Case 1: X-Ray Counts from 1 MPC of Pu-238

If it is assumed that a release event occurs such that the air concentration of Pu-238 is at 1 MPC (i.e., 2×10^{-12} μCi/ml), and that the CAM had been in operation for 12 hours before the release, then a pattern of counts shown in Fig. 6 might occur. Several hours must pass before even a few counts from Pu-238 are registered. Neither the absolute value of the counts nor the slope of the counts contains sufficient information to trigger an alarm. If the release involved Pu-239 instead of Pu-238, the situation would be worse since the yield of L x-ray from Pu-239 is about one-third that of Pu-238. In the case of Am-241, the potential for detection is enhanced since the L x-ray yield is increased by a factor of 4. Still, this would correspond to roughly 8-12 counts at 960 minutes with

L X—RAY COUNTS FROM Rn/Tn DAUGHTERS AND 1 MPC OF PLUTONIUM

Sampling rate = 1 CFM
Rn daughters = .18 pCi/l
Pu—238 = .002 pCi/l
Release at 720 min.

—— Pu—238
— - Rn/Tn
· · · Total

4 MPC hrs.

3—MINUTE COUNTS (y-axis: 0, 20, 40, 60, 80, 100, 120, 140)

TIME FROM START OF SAMPLING (Min) (x-axis: 0, 240, 480, 720, 960, 1200, 1440)

Figure 6

a corresponding background at that time of 50-60 counts, hardly a favorable
condition for detection. The 2-sigma statistic for the background L x-ray
count at that time is approximately 16 counts, which is an indication of
the difficulty to be expected in making a reliable detection of TRU under
these circumstances.

The CAM instrumentation does more than just collect gross counts in a given
interval, but also computes averages of the gross count and background
counts, and from these computes a net count and also a rate of increase or
decrease in count. Due to the expected very low contribution to the
background channel from radon daughters (modeled to be less than 1 count
per 3-minute intervals at 960 minutes) from the 39.86 keV photon of Bi-
212), the background subtraction process may not change the outcome.
Furthermore, as Fig. 6 suggests, the slope of the total L x-ray count
apparently does not diverge sufficiently relative to the stochastic change
in slope to trigger an alarm. Only carefully planned and executed studies
of the response characteristics of this instrument can reveal whether the
predicted poor performance of averaging and detection of rate of rise in
net count is correct.

2.4.2 $\underline{\text{Case 2}}$: X-Ray Counts from 1 MPC of TRU Waste Mixture

Since the typical TRU waste will contain a mixture of the principal TRU
radionuclides including those having high as well a low L x-ray yields,
this case is of particular interest. Based on average drum contents (3.5
Ci of Pu-238, 0.49 Ci of Pu-239, and 1.3 Ci of Am-241) a 1 MPC_a mixture of
these three radionuclides based on the relationships:

$$\frac{C_{P38}}{MPC_{P38}} + \frac{C_{P39}}{MPC_{P39}} + \frac{C_{A41}}{MPC_{A41}} \leq 1$$

$$C_{P38} = 7.1 \, C_{P39}$$

$$C_{A41} = 2.65 \, C_{P39}$$

where

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C_{P38} = concentration of Pu-238 (pCi/l)

C_{P39} = concentration of Pu-239 (pCi/l)

C_{A41} = concentration of Am-241 (pCi/l)

MPC_{P38} = 2 x 10^{-12} μCi/ml

MPC_{P39} = 2 x 10^{-12} μCi/ml

MPC_{A41} = 6 x 10^{-12} μCi/ml

and the ratios of concentration are determined from the waste concentration ratios above. The concentrations that result are:

C_{P38} = 1.58 x 10^{-3} pCi/l

C_{P39} = 2.2 x 10^{-4} pCi/l

C_{A41} = 5.9 x 10^{-4} pCi/l

Figure 7 shows the outcome of simulation with the mixture as inputs, as would be expected, the presence of Am-241 in this 1 MPC mixture leads to a higher count than the simulation of 1 MPC concentration of Pu-238 at corresponding times. But once again, neither absolute value, net count, nor rate of change of count could be counted upon to produce detection at 4 MPC-hours or sooner. The fact that the natural background contribution to the L x-ray count flattens out in the model simulation reflects the simplifying assumption of stable radon daughter concentration. This tends to exaggerate the divergence of the TRU x-ray signal from background, since a stable background for 12 to 24 hours is an exceptional condition. In fact, the radon/thoron background usually exhibits strong diurnal fluctuations in addition to more sudden fluctuations induced by storm fronts. There is, in other words, the potential for rather sharp increases and decreases in background x-ray counts that could easily exceed the changes in slope illustrated by the above simulations of the releases of MPC concentrations of TRU radionuclides.

Only a systematic investigation of the effects of typical natural fluctuations in background on L x-ray variability can provide the basis for predicting the long-term background characteristics the CAM instrument in the variety of settings at WIPP.

L X–RAY COUNTS FROM Rn/Tn DAUGHTERS
AND 1 MPC OF TYPICAL MIXTURE OF TRU RADIONUCLIDES

Sampling rate = 1 CFM
Rn daughters = .18 pCi/l
    Pu–238   = .00158 pCi/l
    Pu–239   = .00022 pCi/l
    Am–241   = .00059 pCi/l
Release at 720 min.

—— TRU x–rays
—·– Rn/Tn
···· Total

3–MINUTE COUNTS

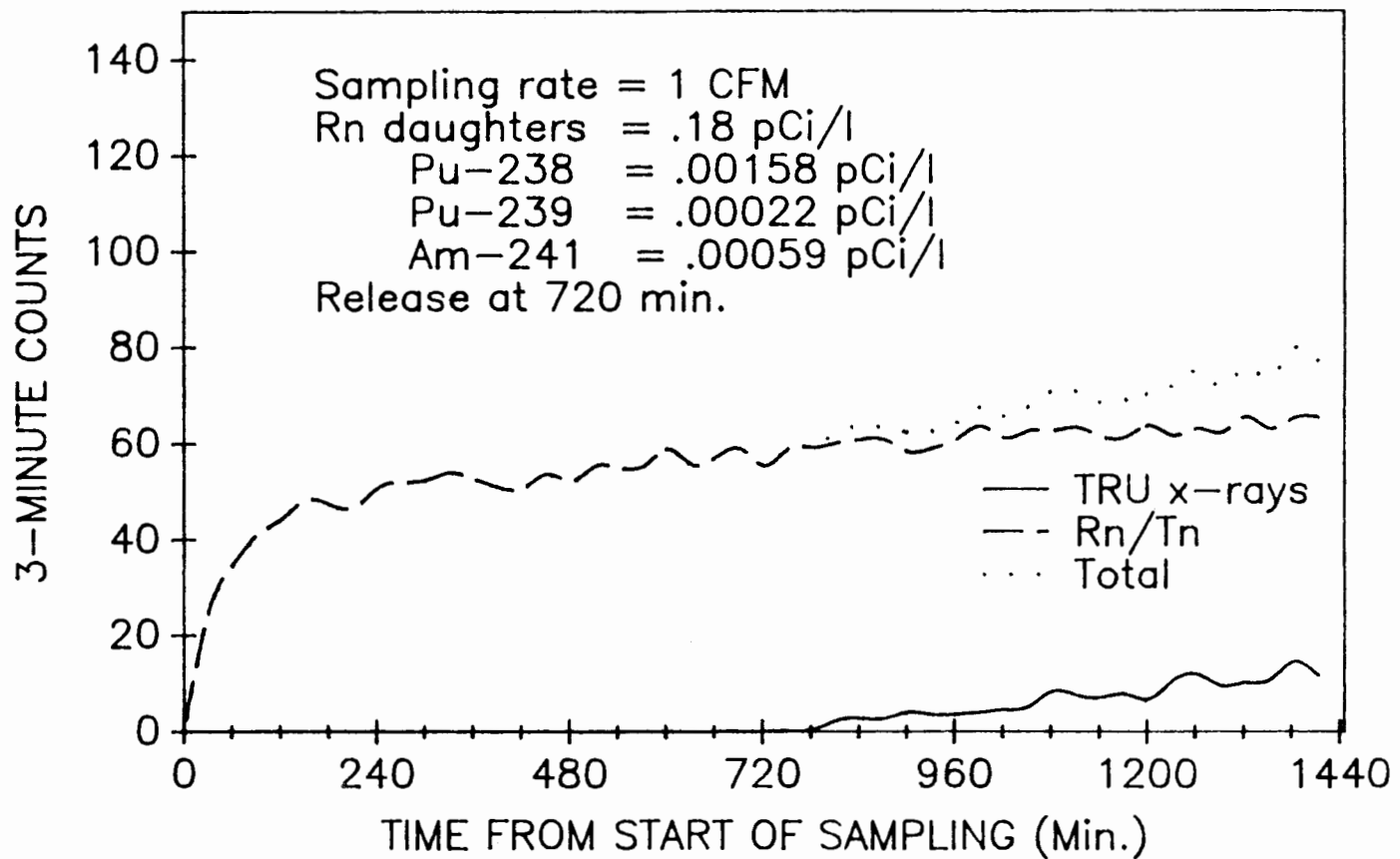TIME FROM START OF SAMPLING (Min.)

Figure 7

2.4.3 Ambient Radon and Thoron Daughter Concentrations at WIPP

In an effort to document the presence and the magnitude of radon and thoron daughter concentrations, a large number of ambient high volume air samples have been taken and analyzed. In one series of measurements made on-site, samples were taken of the surface ambient air and discharge air from the underground portions of the WIPP facility (Fig. 8). The results indicate that a) the ambient levels of the radionuclides of concern are sufficient to produce significant background signals, b) there are large fluctuations in concentrations likely to cause strong variations in sample activity during 24-hours continuous sampling, and c) although there are differences in concentrations between underground and exhaust air, certainly the mine does not act as a filter of incoming air in the sense of reducing background activity to inconsequential levels.

2.5 Discussions of L X-Ray Detection of TRU in the Presence of Radon/Thoron Interference

The foregoing analysis has shown that the present design for airborne TRU detection using a two-channel L X-ray detection scheme is subject to severe interference problems created by the buildup of x-ray emitting daughters of radon and thoron. It has been seen that the thin CaF_2 detector is quite sensitive to these naturally occurring x-ray emitters. By virtue of relatively poor peak resolution, a narrowing of energy discrimination windows is apt not to be a suitable fix. Concentrations of these background radionuclides have been measured on-site at WIPP and found to be present in sufficient concentration to be of serious concern. A simulation of detector response under potential accident conditions suggests that an adequate response (average net count or rate of change of count) is unlikely to occur in the presence of anticipated background on the filter.

Changing the size of the CaF_2 detector or even changing the detector type back to alpha detection are unlikely to resolve these problems with the present CAM if the basic two channel background suppression approach is retained. A careful redesign of the entire approach is indicated.
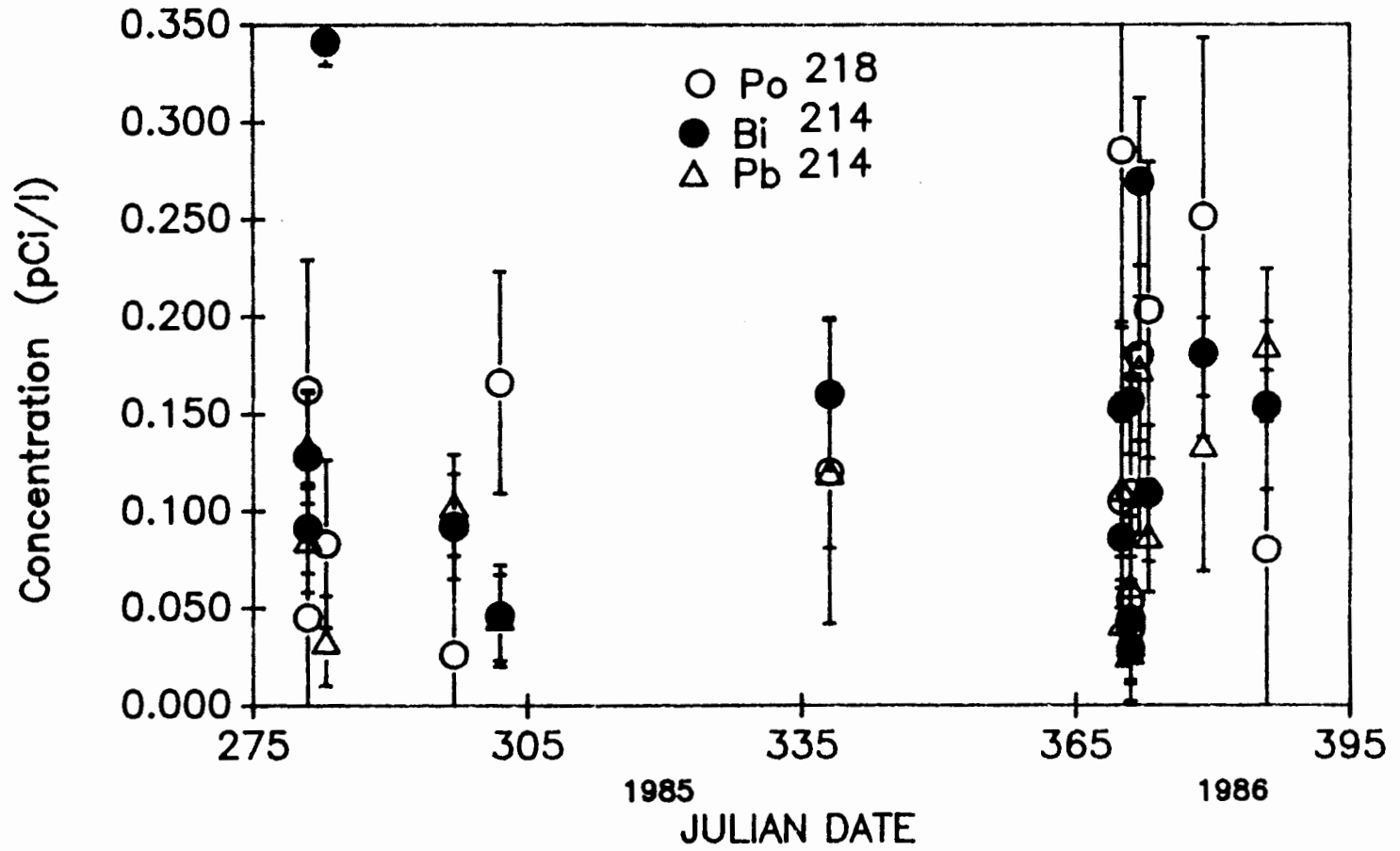
RADON DAUGHTER CONCENTRATIONS
AT THE WIPP SITE

Figure 8

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3.0 CAM PROBE AND TRANSPORT LINE DESIGN ISSUES

CAM applications at the WIPP site include monitoring of work place
environments in the Waste Handling Building, monitoring of work place,
haulage and ventilation drifts and waste storage areas underground, and
monitoring in the exhaust system. Another concern is the ability of the
sampling probe and transport line to deliver a representative sample to the
detector chamber in the CAM head. In the case of CAM applications in the
exhaust duct at the surface, this issue has been addressed in detail in the
EEG report on stack monitoring issues.[11] For other CAM applications at
WIPP somewhat different sampling conditions exist and must be considered
separately.

3.1 Work Place Monitoring Concerns

The working environments at WIPP in which transuranic wastes will be
routinely handled, such as in the Waste Handling Building where TRUPACTs
are unloaded and the waste-filled drums and boxes are handled and prepared
for transfer underground for disposal will be continuously monitored using
skirt mounted CAM units. Although inlet geometries and sampling rates have
not been identified by EEG for this application, it is assumed based on
observed CAM applications underground that the inlet will simply consist of
a 3/4" ID stainless steel pipe mating to the inlet line just above the CAM
head. Required sampling rates have been estimated to be at least 1 CFM to
meet the necessary sensitivity (4 MPC-hr. response). The concern is
whether this nominal sampling rate introduce distortions in the sample.

An assumption is often made that the sampling condition in the work place
is calm air sampling. But at some level of air movement, calm air sampling
should be replaced by isokinetic sampling representative of the conditions
at the location of the CAM station. The maximum wind velocity for which a
calm air sampling assumption is justified has been suggested to be:[12]

$$W \leq 0.002 \ (D^2 U/d_a^{\ 4})^{1/3}$$

where:

W = wind speed (cm/s);

D = probe diameter (cm);

U = entry velocity (cm/s);

d_a = aerodynamic diameter (cm).

For the assumed WIPP CAM configuration, the equation indicates that wind speed should be less than 42 cm/s to permit the assumption of 90% aspirations efficiency for particles as large as 28 μm in calm air. But wind speeds in the work environment are generally between 10 and 100 cm/s (Buchan et al.),[12] so calm conditions cannot always be assumed, and some aspiriation bias against larger-size aerosols must be expected.

Since the inlet in work-room applications may simply be a horizontal or vertically oriented tube in the breathing zone, it is reasonable to assume that under many (if not most) conditions, the airflow crosses the inlet opening at 90° to the direction of inlet flows. Such a sampling geometry leads to sample distortion with respect to larger-size particles.

A number of work-place CAM probe geometries have been examined to determine the potential distortion in sample delivered to the CAM head as a result of inlet configuration, inlet orientation, sampling rate, and room airflow velocity. The two basic CAM configurations assumed are 1) a low airflow room monitoring configuration consisting of a short 3/4" diameter probe with a 3-ft. overall transport line length up to the CAM head, and 2) an underground drift monitoring configuration consisting of a 3/4" diameter probe inlet feeding a 15-ft. horizontal transport line (from the center of the drift to the rib) and an 8-ft. vertical line from the back down to the CAM head.

3.2 Low Airflow Conditions

When the CAM units are used as room air monitors under near-calm conditions and with relatively short transport lines, degradation of sample aspiration performance might be expected to occur (based on modeling) for particles in the 15-μm AED size range or larger (see Fig. 9). This analysis assumes a

SAMPLING EFFICIENCY UNDER LOW
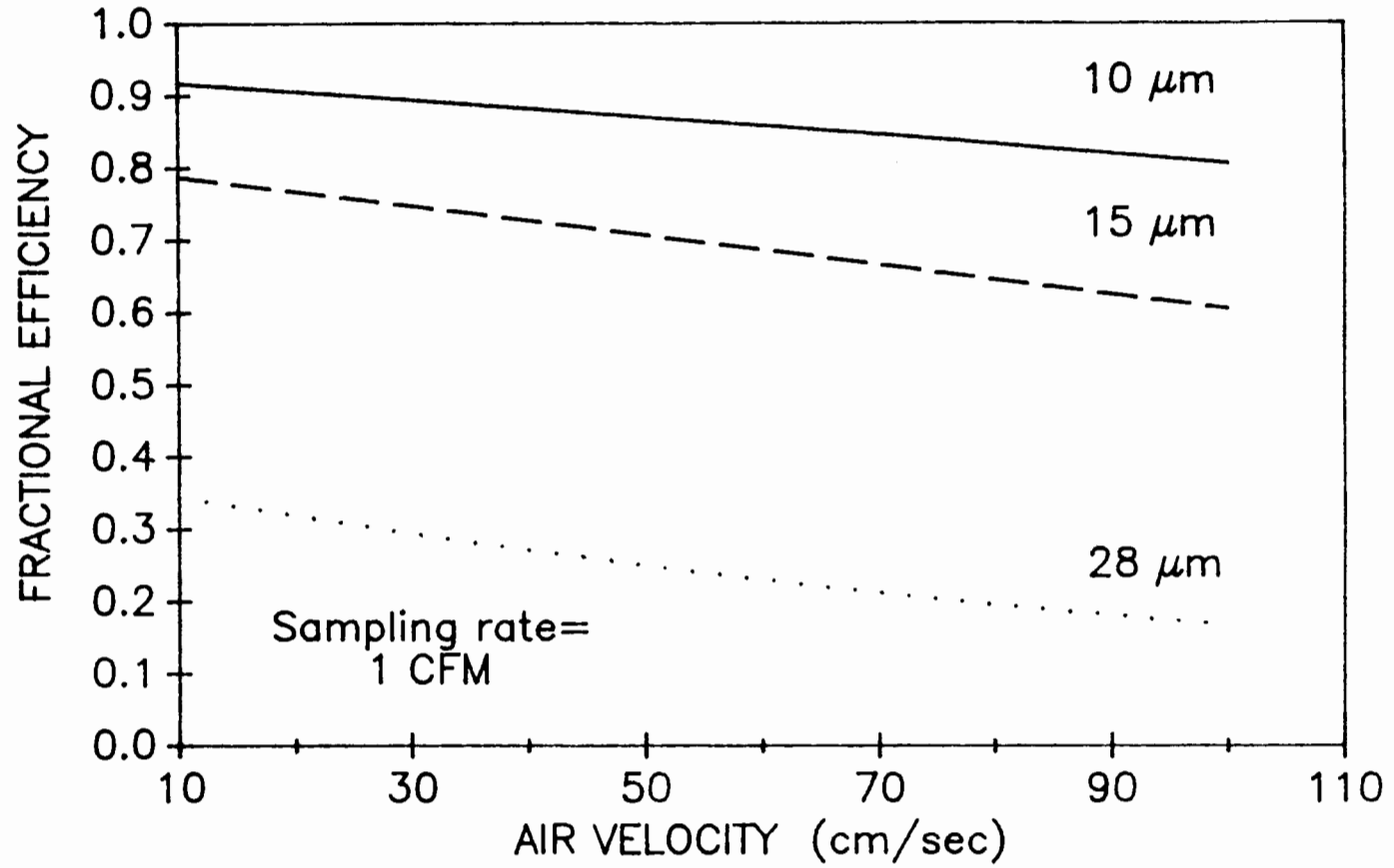FLOW CONDITIONS

Figure 9

3/4" nozzle and sampling at 1 CFM. As other sections of this report indicate, there could be serious particulate impaction problems associated with sampling at 1 CFM in a dusty environment such as may be found in some of the rooms underground. Such interaction effects between sampling rate requirements for adequate sensitivity, consequences of sampling rate on sample loss due to impaction, and sampling rate effects on aspiration and sample transport efficiency clearly need to be investigated systematically and potential conflicting tendencies resolved in the final design and operational characteristics chosen.

3.3 High Airflow Conditions

In contrast to conditions which may be found in work rooms on the surface or underground, the airflow in the haulageway and ventilation drifts may be much higher, particularly in the exhaust air drifts. The various airflow conditions expected in the drifts throughout the mine could vary from 100 cm/s to 750 cm/s, depending on the geometry of the drift, the operating conditions of the mine, and other factors. Inasmuch as the individual skid mounted CAM units are designed to operate at a fixed flow rate (maintained steady by a thermoanemometer flow control system in the exhaust lines of the x-ray and beta CAM detector systems), sampling cannot be isokinetic.

The CAMs in the stack monitoring system are designed to operate isokinetically. (The expected performance of the stack CAMs are assessed in report EEG-37.)[13]

Considering the fixed flow CAM, and assuming a sampling rate of 1 CFM, the expected performance of the sampling system up to the CAM head is calculated to be generally very poor for particles 10 μm or larger, even at flows of 100 cm/s. The principal sources of sample distortion with this second configuration are poor aspiration efficiency and losses in the horizontal portion of the transport line (see Fig. 10).

It should be noted that the expected losses and distortion of the sample within the CAM head itself, to be discussed in Chapter 4, compound the sampling line effects. It will be seen, for example, that at 1 CFM the D_{50}
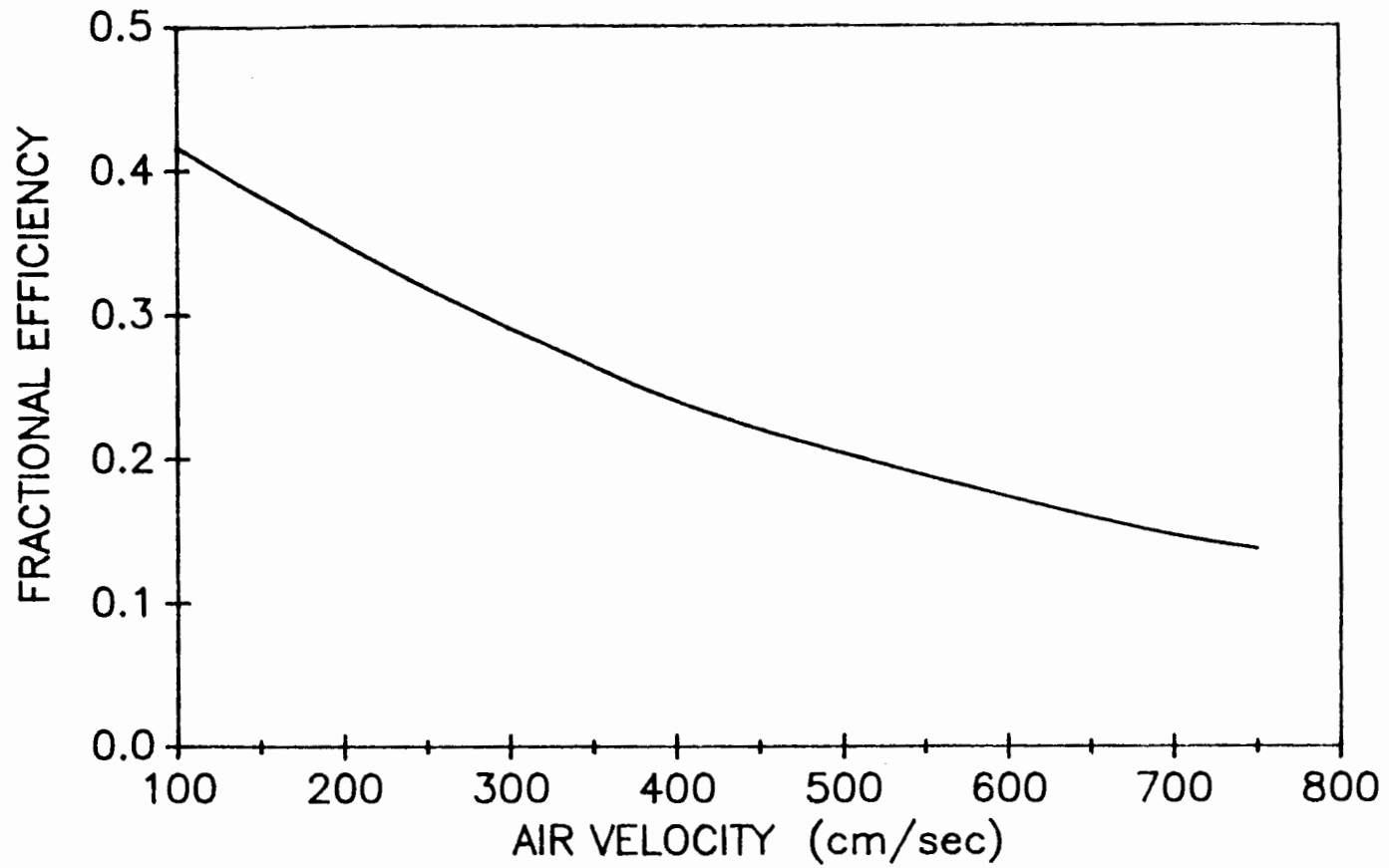
43

SAMPLING EFFICIENCY UNDER HIGH
FLOW CONDITIONS

Figure 10

cut is at about 1.5 μm, so that even those larger-size particles that survive transport in the sampling lines are very likely to be lost to impaction within the CAM head.

Overall, therefore, the performance of the underground or work place CAMs is predicted to be decidedly substandard, and cannot be counted on to provide the crucial early warning of a radioactive release. It is recognized that these CAM units do not have to produce a representative sample in the sense of a sample for record, that task being accomplished by open-faced fixed air samplers. However, the serious distortion and loss of sample predicted by the foregoing calculations indicates that even the alarm task may very well be compromised.

4.0   WIPP CAM SAMPLING HEAD DESIGN ISSUES

The terminus of the samples transport line in the various CAM units at WIPP is characteristically a 0.75" diameter line entering a shielded detector housing at a right angle to the long axis of the cylindrical housing. As shown in Fig. 1, the flow of air must enter in a jet from the inlet line, and then turn through 90° and spread over and through the filter before exiting the outlet line. Note that the inlet line undergoes a restriction in diameter just before entering the filter/detector chamber such that the inlet jet is approximately 1/8" in diameter.

Under the proposed sampling conditions of CAMs, particularly in the applications where the CAM must sample heavily dust laden air at relatively high sampling rates to achieve isokinetic sampling conditions, there is the possibility that the CAM filter and detector assembly (the CAM head) will act as an impactor with attendant loss and distortion of the sample collected. The large particle trajectories will diverge from the flow lines to impact on the far wall. Clearly, higher sampling rates, and hence higher jet velocities, would amplify this effect. The effect can be approximately modeled as jet impingement on a plate such as is used in the design of cascade impactors.

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4.1 Impactor Concepts

The process of airflow impingement on a plane surface depends[14] on several factors in the geometry and flow conditions of the jet including:

   a) the average flow velocity U_o of the jet (equal to the volumetric flow rate divided by the area of the opening),

   b) the jet opening diameter W,

   c) the aerosol size, shape and density,

   d) the viscosity of air, η,

   e) the jet throat length T, and

   f) the jet to impingement surface distance S.

The airflow streamlines themselves will follow the 90° bend created by airflow through the filter, but particles entrained in that flow may well separate and change direction, leading to impaction. The measure of the amount of separation from the airflow can be characterized in terms of the particle stopping distance. For a particle initially traveling at velocity U_o, the distance traveled in the forward direction before coming to rest with respect to the surrounding air defines the stopping distance.[15] The stopping distance L for a spherical particle with aerodynamic diameter D is given by:

$$L = \frac{U_o D^{2*} \rho}{18\eta}$$

where $\rho*$ is unit density (12).

A dimensionless parameter Ψ, obtained by dividing the stopping distance by the jet diameter, can be used to parameterize the impaction process:

$$\Psi = L/W$$

The square root of this parameter is proportional to particle aerodynamic diameter, hence

$$\sqrt{\overline{\Psi}} = D \left[\frac{U_o \rho^*}{18W} \right]^{0.5} \quad .$$

When $\sqrt{\overline{\Psi}}$ is plotted vs efficiency of collection by impaction it has been found (12) that the 50% collection point for round jets has a value approximately equal to 0.32. Therefore, the equation for Ψ in terms of aerodynamic diameter can be solved for the 50% cutoff diameter for the jet assuming $T/N = 1.0$, and $S/W = 1.0$:

$$D_{cut}(\mu m) = 1.257 \times 10^3 \; (W^3/F)^{0.5}$$

where W is the jet diameter in cm and F in the volumetric flow in cm^3/min.

4.2 Predicted Impaction Losses

Assuming a jet diameter of 0.32 cm (0.125") and a variable flow rate of 0.2 CFM-6 CFM, the effective aerodynamic diameter for a 50% cut varies between $3.6\mu m$ and $0.66\mu m$ AED (see Fig. 11). The corresponding percentage of sample loss due to impaction cannot be predicted without knowing the particle size distribution of the sample. But clearly, if the mass median aerodynamic diameter is large, a very substantial fraction of the sample will be lost.

There would be varying amounts of smaller-size particles lost as well to impaction on the far wall of the CAM chamber. Depending on location in the underground drifts or rooms or in the exhaust stack, losses of substantial fractions of larger aerodynamic diameter particle components of sample, as predicted by this model, could have a severe effect on CAM performance. In underground areas near mining operations, or in areas of high traffic the mean diameter has been found to be much larger than in more remote areas. Also, in the case of accidental release, the mean aerodynamic diameter could be much larger than under normal conditions. Hence, impaction losses will have to be demonstrably controlled before the CAM can be considered a truely operational instrument.
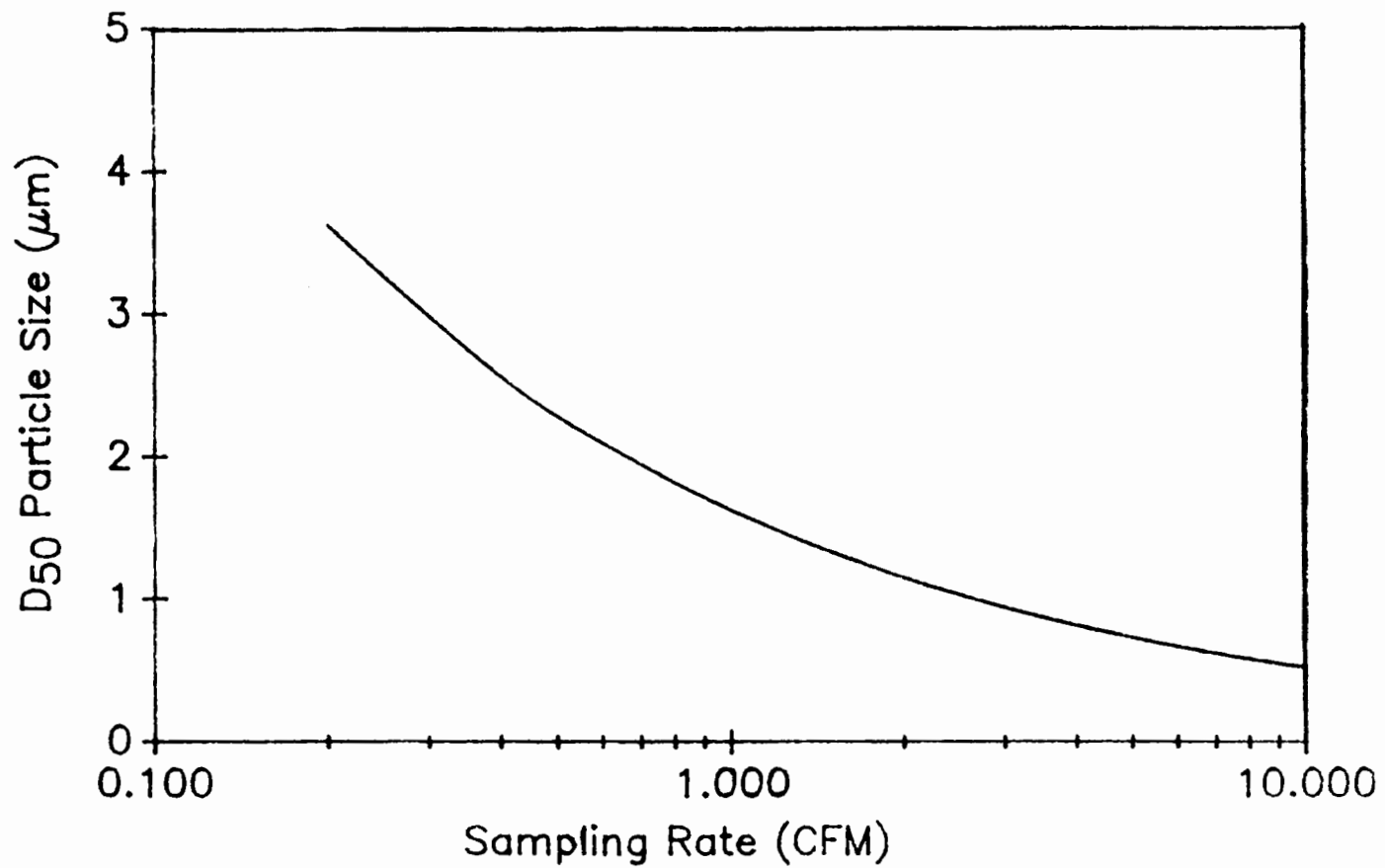
D50 CUT OF CAM HEAD NOZZLE

Figure 11

Another factor affecting CAM performance is the pattern of deposition of particles which do not impact on the sidewall, but still become displaced from the airstream and therefore collect preferentially on the far side of the filter opposite the jet entrance. If significant pile-up of particles in one region of the filter occurs, the effective counting geometry changes, and the potential for self-absorption of the radiation being monitored increases.

4.3 Asymmetrical Deposition Effects

The process of asymmetrical deposition depends so much on the properties of the filter medium (e.g., resistance to flow), and the geometry of the counting chamber, it is very difficult to predict the pattern from theoretical considerations.

In order to further study the problem of non-uniform sample deposition, the consequences for detector efficiency of particulate deposition location on the filter was explored with the detector response model described in Chapter 2. The surface of the filter, which is coaxial with the detector, was modeled as if it were divided into 9 concentric rings, each of which having about equal area. Then the source of each photon arriving at the detector (whether absorbed or not) was determined, as well as the total number leaving each area. The ratio of the total number of hits on the detector to the number leaving each area is then a measure of the efficiency for detection as a function of distance from the center. The result, shown in Fig. 12, reveals that indeed for the geometry of the NRC CAM head, the efficiency drops rapidly and significantly from about 80% of 2π near the center of the filter to less than 2% near the edge. Therefore, for the larger-particle size fraction of the sample deposited toward the far side of the chamber from the jet entrance, the detection efficiency would be very significantly reduced, even more than reported by Biermann and Velen for the alpha CAM they investigated.[16] Increasing the CaF_2 detector size to match the filter size does apparently improve the detection of off-center sources, based on model simulation. The efficiency near the edge is then closer to 75% of 2π.

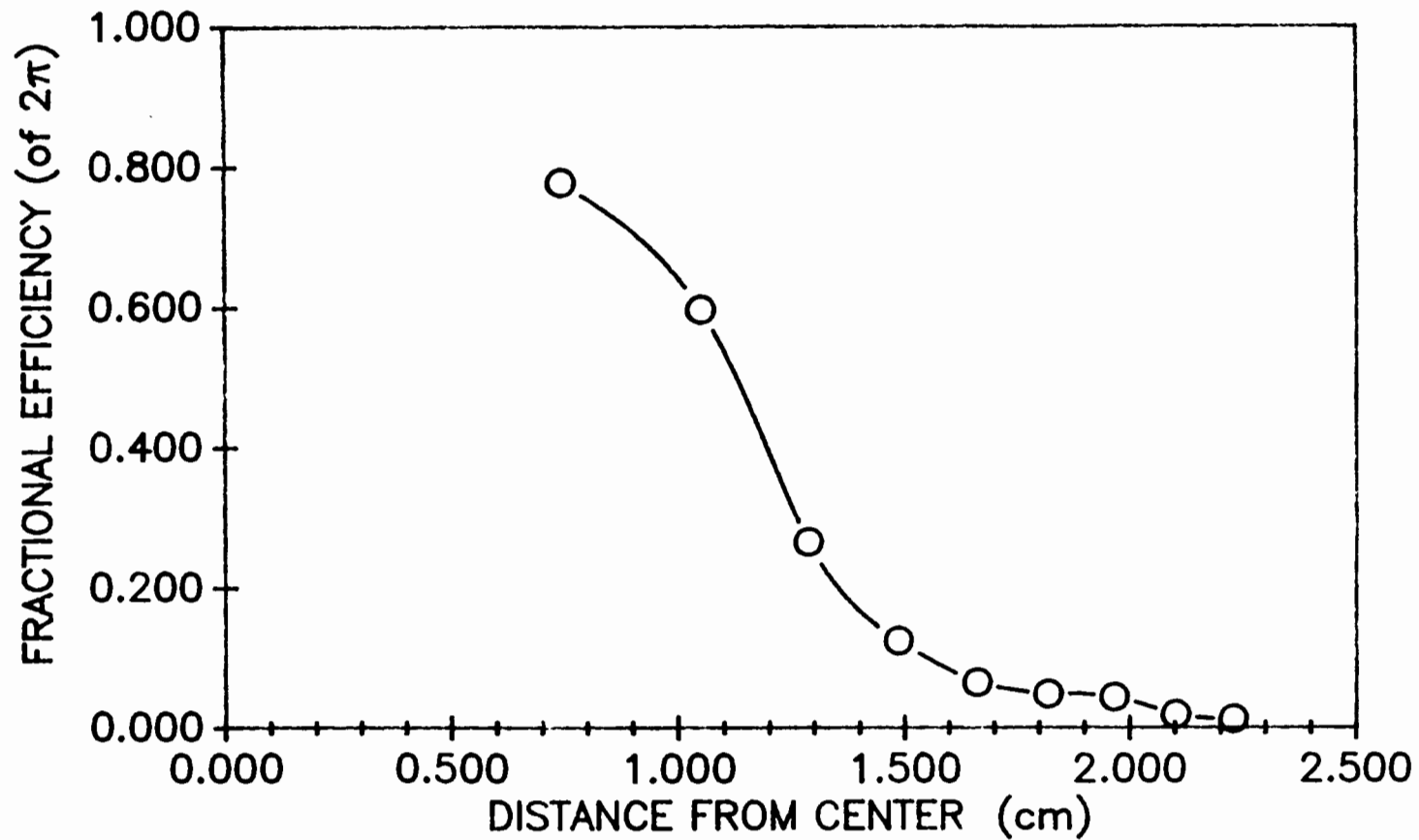DETECTOR RESPONSE AS A FUNCTION OF SOURCE
DISTANCE FROM CENTER OF DETECTOR

Figure 12

Empirical studies, such as those of Biermann and Velen of an alpha CAM using monodisperse aerosols clearly show a distortion of particle deposition, particularly for particles above 2-μm AED. For the largest size aerosols considered (6.0 μm) it was estimated that only 15% would actually be detected, for 2.7-μm particles 32% would be detected, and for the smallest aerosols (0.6 μm) the amount detected approaches 35%. For an L x-ray detection scheme, and for the sampling head geometry under review these fractions will undoubtedly be different, but they do illustrate the problem.

On the basis of the above analysis, there is sufficient evidence of impaction losses in the CAM head and asymetrical distribution on the filter leading to loss of sensitivity and possibly loss of accuracy to warrant a complete and thorough investigation of the sample collection characteristics of the WIPP CAM head using monodisperse aerosols having a range in size from less than 1-μm AED to over 10-μm AED. The series of tests should include a test incorporating a radioactive tracer label which emits L x-rays of appropriate energy (perhaps the progeny of radon and thoron which are known to readily attach to aerosols) as required by DOE regulations.

EEG would anticipate receiving a copy of the proposed test protocol and performance criteria for review as well as the data, and report of the outcomes.

5.0 CONCLUSIONS AND RECOMMENDATIONS

The foregoing several chapters have explored a number of apparently serious deficiencies in the design and construction of the WIPP CAMs. Much of the analysis, out of necessity, has been based more on theoretical analysis than on an analysis of actual performance data. As far as has been learned from discussions with the WIPP operations office, there have been no experimental studies conducted with these CAMs, with the exception of an early investigation of the L x-ray CAM by the ITRI. The results of that study still have not been made available to the State for evaluation. From

what has been said of that study informally, it would appear that very little, if any of the concerns raised in the present assessment were addressed in that study, and hence, the major findings would remain the same.

The principal conclusions of the review are these:

a) The specifications for the design of a CAM system for the WIPP site were written for an alpha detection scheme, not an L x-ray detection approach, and therefore, are notably deficient in performance specifications appropriate to the detection of L x-rays. It would appear that the use of the insoluble limit for Pu-238 together with a response time of 2 hours rather than the soluble limit for Pu-239 and an 8-hour response time has lead to a less restrictive integral response specification than DOE Orders require.

b) The CAM instrumentation that has actually been built and installed at numerous locations around the WIPP facility, both above ground and underground, was apparently built with the supposition that the concerns for background subtraction with the radiation detection devices used is limited to a relatively stable ambient field which can essentially be "zeroed" out at each instrument location. Even in the case of the L x-ray CAM where a two-channel counting method is used to correct for background, it would appear that specific concerns for the effect of collection of radon and thoron progeny along with airborne particulates on the detection of Pu or Am were not appreciated or dealt with as required by DOE Orders (and good radiological instrumentation engineering). The design of beta CAMs simply ignores the possibility of beta interference on the filter.

c) Both by Monte Carlo modeling of the scintillator detector response and by experimentation with a thin scintillator detector of similar design, it would appear that the detection of radon/thoron progeny L x-rays will be at least as efficient as the detection of Pu and Am L x-rays. The choice of $CaF_2(Eu)$ rather than $NaI(Tl)$ compounds the process of peak broadening which makes the use of narrow window settings an unattractive option in efforts to control interference.

d) Modeling of the buildup of x-ray emitters on the L x-ray CAM filter during sampling indicates that the background counts from radon and thoron daughters could be 10 times or more the counts from 1 MPC of a typical mixture of radionuclides on a filter for many hours after a release event occurs, making reliable, fast response unlikely with the present system. The beta CAM system is so at variance with normal schemes for coping with background beta emitters collected on a filter along with the target radionuclides, it is not clear just how to estimate the response time. The model of the buildup of background activity on a filter during continuous sampling was verified by on-site measurements of the alpha-emissions from these radionuclides, which enhances confidence in the prediction of difficulties for the x-ray system.

e) On-site measurements of radon and thoron progeny, both in ambient air and in exhaust air from the underground facilities, indicate that there is ample source strength at all potential CAM locations for background subtraction to be a practical necessity.

f) A study of particle transport in the nozzle and transport lines needed for some CAM locations suggests that significant sample distortion with respect to particle size will occur. A general strategy for determining optimum sampling rate at each CAM location (ranging from high-velocity air flow to calm air conditions) has not been reported to EEG.

g) The CAM head itself apparently was not designed with appropriate concern for aerosol transport into the counting chamber. The inlet line undergoes a 90° bend just before entering the head; there is an abrupt narrowing of tube diameter from approximately 3/4" to 1/8"; and then the air enters the chamber in a jet out of a 1/8" diameter opening. All of these conditions contribute to sample loss and distortion of representation of all particle sizes in the sample. The chamber itself is predicted to behave much like an impactor, with significant loss of larger particles through impaction on the far wall. Intermediate-size particles will deposit preferentially near the outer edge of the filter where detection efficiency is expected to be severely reduced based on the model of the detection processes.

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Taken together, these seven findings constitute a very pessimistic appraisal of the expected overall performance of the present WIPP CAM designs. Compensating factors may emerge in the course of detailed performance testing under the wide variety of applications intended at WIPP which could not be anticipated here. Nevertheless, based on what is presently known about these instruments and the conditions under which they operate, the expectation is that they will fail to prove acceptable for the WIPP applications.

The EEG recommendations are, therefore:

a) The NRC alpha and beta CAMs must be subjected to thorough performance testing by an experienced laboratory with the capability of creating test conditions covering the expected range of environmental conditions to be found at WIPP. The tests and the level of performance demonstrated must meet or exceed those specified in the DOE Orders (or by reference in the Orders, such as certain ANSI Standards).

b) In addition to laboratory testing, appropriate in-place testing must be performed using well characterized and labeled aerosols to demonstrate that suitable performance can indeed be achieved under actual operating conditions.

c) In light of the identified deficiencies in the L x-ray detection approach, the WIPP project should initiate a vigorous search for alternative CAM systems and detection schemes (both alpha and beta, if both are to be used). Well qualified alternative systems may exist which should be identified and tested for WIPP applications. It can be anticipated that an alternative approach based on alpha detection, for instance, will not be easily identified based on the initial difficulties encountered in the search for a suitable system. However, a multichannel alpha spectrometry approach is under development at Los Alamos National Laboratory, which offers the potential for greatly enhanced performance in rejecting radon and thoron background. Simply substituting an alpha detector for the x-ray detector in the present CAM head does not appear to be a workable solution.

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d) It may be necessary to consider approaches which are not widely used in nuclear facilities handling plutonium due to the fact that many of the WIPP environments which must be monitored are very much more dusty than ordinary rooms. One approach meriting consideration would be the use of a moveable filter system which would permit the interruption of the continuous buildup of background radionuclides on the filter, and also the decay of the short-lived component of the background radionuclides collected. However, this approach may be more costly to implement and suffers from the difficulty of obtaining a good seal between filter and pump.

e) A variety of detector devices should be considered. For example, x-ray detection may well be a workable approach if a detector with very much better energy resolution could be identified, thereby permitting a much improved background subtraction scheme to be employed utilizing multichannel spectrometry. Whatever approach is chosen, ample time must be allowed to permit thorough testing of every aspect of the instrument such as has been considered in the present review.

f) If EEG is to fulfill its role under the 1978 contract as well as the Consultation and Cooperation Agreement provisions, the process of demonstrating that WIPP does indeed have a fully operational CAM network which meets or exceeds the standards of performance of the U. S. Department of Energy (DOE) and the nuclear community should be conducted with the full participation of EEG. Participation in design reviews, peer reviews, test plan reviews, and evaluations of the outcome of critical test programs would contribute greatly to the success of the effort and enable a prompt review from the State of whatever final solution is proposed to the problems of continuous air monitoring at WIPP.

REFERENCES

1. Correspondence, Robert H. Neill, Director of Environmental Evaluation Group, to Jack Tillman, Site Manager, WIPP Operations Office, September 23, 1987.

2. ANSI N13.1-1969, American National Standards Institute, Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities.

3. ANSI N42.18-1979, American National Standards Institute, Specification and Performance of On-Site Instrumentation for Continuously Monitoring Radioactivity in Effluents, 1980.

4. U. S. Department of Energy (DOE) Order 5480.1A "Standards for Radiation Protection".

5. U. S. Department of Energy, "A Guide to Reducing Radiation Exposure to As Low As Reasonably Achievable (ALARA)", R. L. Kathren, Battelle Pacific Northwest Laboratory, Principal Investigator, DOE/EV/1830-TS, April 1980.

6. U. S. Department of Energy, Order 5480.4, Environmental Protection, Safety, and Health Protection Standards, Attachment 2, "Mandatory ES4H Standards (Policy Requirements), 5b.

7. Nuclear Research Corporation, WIPP Instrumentation Manual, 1984.

8. Kocher, D. C., Radioactive Decay Data Tables Dept. of Energy, Technical Information Center, DOE/TIC-11026, March 1985.

9. Rodgers, J. C., Personal communications with Los Alamos National Laboratory, Los Alamos, New Mexico, and Harshaw /Filtrol Partnership, Solon, Ohio.

10.   Newton, G. J., Y. Cheng, B. A. Wong, B. B. Boecker, Aerosol Measurements in the Partially Completed Underground Waste Isolation Pilot Plant: Final Report.   Inhalation Toxicology Research Report July 25, 1983.

11.   Kenney, J. W., J. C. Rodgers, J. Chapman, and A. Dhar, Environmental Monitoring at the WIPP Site.   (DRAFT EEG Report in preparation.)

12.   Buchan, R. M., S. C. Soderholm, and M. I. Tillery, Aerosol Sampling Efficiency of 37mm Filter Cassettes, Am. Ind. Hyg. Assoc. J., 47, 825-31, 1986.

13.   Rodgers, J. C., Stack Monitoring Issues at the Waste Isolation Pilot Plant, EEG Report EEG-37, November, 1987.

14.   Newton, G. J., O. G. Raabe, and B. U. Mokler, Cascade Impactor Design and Performance, J. Aerosol Science, 8, 339, 1977.

15.   Fuchs, N. A., The Mechanics of Aerosols, Pergamon Press, Oxford, 1964.

16.   Biermann, A., and S. Velen, CAM Particle Deposition Evaluation, Lawrence Livermore National Laboratory, Report UCRL-50007-83, June 15, 1984.

Environmental Evaluation Group
Reports

EEG-12   Little, Marshall S., Potential Release Scenario and
          Radiological Consequence Evaluation of Mineral Resources
          at WIPP, May 1982.

EEG-13   Spiegler, Peter., Analysis of the Potential Formation of a
          Breccia Chimney beneath the WIPP Repository, May, 1982.

EEG-14   Not published.

EEG-15   Bard, Stephen T., Estimated Radiation Doses Resulting if an
          Exploratory Borehole Penetrates a Pressurized Brine Reservoir
          Assumed to Exist Below the WIPP Repository Horizon, March
          1982.

EEG-16   Radionuclide Release, Transport and Consequence Modeling for
          WIPP. A Report of a Workshop Held on September 16-17, 1981,
          February 1982.

EEG-17   Spiegler, Peter, Hydrologic Analyses of Two Brine Encounters
          in the Vicinity of the Waste Isolation Pilot Plant (WIPP)
          Site , December 1982.

EEG-18   Spiegler, Peter, Origin of the Brines Near WIPP from the
          Drill Holes ERDA-6 and WIPP-12 Based on Stable Isotope
           Concentrations of Hydrogen and Oxygen, March 1983.

EEG-19   Channell, James K., Review Comments on Environmental Analysis
          Cost Reduction Proposals (WIPP/DOE-136) July 1982, November
          1982.

EEG-20   Baca, Thomas E., An Evaluation of the Non-radiological
          Environmental Problems Relating to the WIPP, February 1983.

EEG-21   Faith, Stuart, et al., The Geochemistry of Two Pressurized
          Brines From the Castile Formation in the Vicinity of the Waste
          Isolation Pilot Plant (WIPP) Site, April 1983.

EEG-22   EEG Review Comments on the Geotechnical Reports Provided by
          DOE to EEG Under the Stipulated Agreement Through March 1,
          1983, April 1983.

EEG-23   Neill, Robert H., et al., Evaluation of the Suitability of the
          WIPP Site, May 1983.

EEG-24   Neill, Robert H. and James K. Channell Potential Problems
          From Shipment of High-Curie Content Contact-Handled
          Transuranic (CH-TRU) Waste to WIPP, August 1983.

Environmental Evaluation Group
Reports

EEG-25 Chaturvedi, Lokesh, Occurrence of Gases in the Salado Formation, March 1984.

EEG-26 Spiegler, Peter, Environmental Evaluation Group's Environmental Monitoring Program for WIPP, October 1984.

EEG-27 Rehfeldt, Kenneth, Sensitivity Analysis of Solute Transport in Fractures and Determination of Anisotropy Within the Culebra Dolomite, September 1984.

EEG-28 Knowles, H. B., Radiation Shielding in the Hot Cell Facility at the Waste Isolation Pilot Plant: A Review, November 1984.

EEG-29 Little, Marshall S., Evaluation of the Safety Analysis Report for the Waste Isolation Pilot Plant Project, May 1985.

EEG-30 Dougherty, Frank, Tenera Corporation, Evaluation of the Waste Isolation Pilot Plant Classification of Systems, Structures, and Components, July 1985.

EEG-31 Ramey, Dan, Chemistry of the Rustler Fluids, July 1985.

EEG-32 Chaturvedi, Lokesh and James K. Channell, The Rustler Formation as a Transport Medium for Contaminated Groundwater, December1985.

EEG-33 Channell, James K., John C. Rodgers and Robert H. Neill, Adequacy of TRUPACT-I Design for Transporting Contact-Handled Transuranic Wastes to WIPP, June 1986.

EEG-34 Chaturvedi, Lokesh, (edi), The Rustler Formation at the WIPP Site, January 1987.

EEG-35 Chapman, Jenny B., Stable Isotopes in Southeastern New Mexico Groundwater: Implications for Dating Recharge in the WIPP Area, October 1986.

EEG-36 Lowenstein, Tim K., Post Burial Alteration of the Permian Rustler Formation Evaporites, WIPP Site, New Mexico, April 1987.

EEG-37 Rodgers, John C., Exhaust Stack Monitoring Issues at the Waste Isolation Pilot Plant, November 1987.