# THE INFLUENCE OF SALT AEROSOL ON ALPHA RADIATION DETECTION BY WIPP CONTINUOUS AIR MONITORS

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#### **FOREWORD**

The purpose of the New Mexico Environmental Evaluation Group (EEG) is to conduct an independent technical evaluation of the Waste Isolation Pilot Plant (WIPP) Project to ensure the protection of the public health and safety and the environment. The WIPP Project, located in southeastern New Mexico, is being constructed as a repository for the disposal of transuranic (TRU) radioactive wastes generated by the national defense programs. The EEG was established in 1978 with funds provided by the U.S. Department of Energy (DOE) to the State of New Mexico. Public Law 100-456, the National Defense Authorization Act, Fiscal Year 1989, Section 1433, assigned EEG to the New Mexico Institute of Mining and Technology and continued the original contract DE-ACO4-79AL10752 through DOE contract DE-ACO4-89AL58309. The National Defense Authorization Act for Fiscal Year 1994, Public Law 103-160, continues the authorization.

EEG performs independent technical analyses of the suitability of the proposed site; the design of the repository, its planned operation, and its long-term integrity; suitability and safety of the transportation systems; suitability of the Waste Acceptance Criteria and the generator sites' compliance with them; and related subjects. These analyses include assessments of reports issued by the DOE and its contractors, other federal agencies and organizations, as they relate to the potential health, safety and environmental impacts from WIPP. Another important function of EEG is the independent environmental monitoring of background radioactivity in air, water, and soil, both on-site and off-site.

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## **EXECUTIVE SUMMARY**

Alpha continuous air monitors (CAMs) will be used at the Waste Isolation Pilot Plant (WIPP) to measure airborne transuranic radioactivity that might be present in air exhaust or in work-place areas. WIPP CAMs are important to health and safety because they are used to alert workers to airborne radioactivity, to actuate air-effluent filtration systems, and to detect airborne radioactivity so that the radioactivity can be confined in a limited area.

In 1993, the Environmental Evaluation Group (EEG) reported that CAM operational performance was affected by salt aerosol, and subsequently, the WIPP CAM design and usage were modified. In this report, operational data and current theories on aerosol collection were reviewed to determine CAM quantitative performance limitations. Since 1993, the overall CAM performance appears to have improved, but anomalous alpha spectra are present when sampling-filter salt deposits are at normal to high levels.

This report shows that sampling-filter salt deposits directly affect radon-thoron daughter alpha spectra and overall monitor efficiency. Previously it was assumed that aerosol was mechanically collected on the surface of CAM sampling filters, but this review suggests that electrostatic and other particle collection mechanisms are more important than previously thought. The mechanism of sampling-filter particle collection is critical to measurement of acute releases of radioactivity.

Specific Recommendations. Aerosol collection mechanisms are complex, and extensive research would be necessary to define and quantify all the variables associated with WIPP air monitoring. To insure adequate monitor performance:

- salt dust accumulations on CAM sampling filters should be kept to a minimum;
- CAMs should be designed to automatically detect sampling-filter salt deposits and alarm when deposits reach a predetermined level;
- underground CAMs should be strategically located in low-salt-aerosol airflows,
   downstream from radioactive waste; and
- performance limitations should be quantified and documented.

### INTRODUCTION

Alpha continuous air monitors (CAMs) are used at the Waste Isolation Pilot Plant (WIPP)<sup>1</sup> to monitor airborne radioactivity. The WIPP CAMs and air sampling systems have been meticulously studied and redesigned to insure adequate performance. Despite these efforts, anomalous alpha spectra appear as a broad range of energies, rather than as characteristic peaks of discrete energy. This report is a technical evaluation of the cause and significance of anomalous alpha spectra to alpha CAM operations.

The WIPP is a proposed geological repository for defense transuranic<sup>2</sup> wastes that contain plutonium and other alpha emitting radionuclides and is located in a bedded-salt formation approximately 655 m (2150 ft) below the surface. Mining and other operations create high concentrations of salt aerosol<sup>3</sup> in mine work areas and in the underground air effluent where CAMs are located. CAM measurements are essential to worker safety and the detection and prevention of possible environmental releases from the underground repository.

The basic operation of alpha CAMs, particle collection theory, and WIPP operational data are systematically reviewed and evaluated with the intent of defining alpha CAM operational limitations. An understanding of performance limitations is a prerequisite to successfully using CAMs in the radiation safety program, as part of a confinement scheme, or in methods employed by the defense-in-depth philosophy<sup>4</sup>.

<sup>&</sup>lt;sup>1</sup> The WIPP is located 42 km (26 mi) east of Carlsbad, New Mexico. The WIPP mission is to dispose of 176,000 m<sup>3</sup> (6.2 million cubic feet) of contact-handled (CH-TRU) waste and 7,080 m<sup>3</sup> (250,000 cubic feet) of remote-handled (RH-TRU) waste (US DOE 1990). The total radioactivity from CH-TRU will be about 1.01 x 10<sup>7</sup> Curies (Ci)(US DOE, OERWM 1990) and a maximum of 5.1 x 10<sup>6</sup> Ci from RH-TRU waste (NM and US DOE 1984).

<sup>&</sup>lt;sup>2</sup> Transuranic refers to elements containing nuclides with atomic numbers greater than uranium(92).

<sup>&</sup>lt;sup>3</sup> As used in this report, aerosol is defined as a gaseous suspension of ultramicroscopic solid particles (Parker 1989).

<sup>&</sup>lt;sup>4</sup> Defense-in-depth is a Department of Energy (DOE) approach to facility safety based on several layers of protection (U.S. DOE 1994b).

#### BACKGROUND INFORMATION

In 1988, the Waste Isolation Division (WID)<sup>5</sup> of Westinghouse Electric Corporation procured Eberline<sup>6</sup> Alpha-6 CAMs because radon and thoron progeny<sup>7</sup> interfered with the operation of WIPP L x-ray CAMs (Rodgers and Kenney, 1988). Although the Alpha-6 CAM was a state-of-the-art system and could subtract radon-thoron counts, anomalous alpha spectra were prevalent (Bartlett, 1993a). To improve alpha CAM performance, the WIPP effluent CAMs were redesigned (Arthur 1993). Overall reliability improved, but anomalous and poorly resolved alpha spectra were apparent as salt-aerosol concentrations increased, indicating a more fundamental problem than CAM equipment design. It appeared that the stopping power of the medium increased between the sampling filter and detector, and alpha particle kinetic energy was reduced before reaching the CAM detector. A broad spectrum of alpha energies was detected for each alpha species, rather than a more limited range of energies.

## Alpha Particle Detection

Alpha particle detection is not a straightforward methodology. The transuranic radionuclides <sup>238</sup>Pu, <sup>239</sup>Pu and <sup>241</sup>Am decay by emitting alpha particles more than 99% of the time (ICRP 38, 1983). The high alpha particle yield is a favorable detection characteristic, but alpha particles have limited range in air and other materials. For monitors to operate effectively, alpha emissions must be in close proximity to the CAM detector, with little interposed material between the point of emission and the detector. For example, a 5.1 MeV<sup>8</sup> <sup>239</sup>Pu alpha particle has a maximum range of about 42 mm (1.6 in) in air (U. S. HEW, 1970; Turner,1995) and can not penetrate a salt layer of more than 6.3 mg cm<sup>-2</sup> (Ziegler,1985; Bartlett, 1993b). If the sampling-filter-to-detector gap is greater than 42 mm (1.6 in), or if <sup>239</sup>Pu is buried beneath 6.3

<sup>&</sup>lt;sup>5</sup> The WID operates the WIPP for the DOE and is responsible for the CAM modifications referenced in this report.

<sup>&</sup>lt;sup>6</sup> Eberline is a division of Thermo Bioanalysis, P. O. Box 2108, Santa Fe, New Mexico 87504-2108.

<sup>&</sup>lt;sup>7</sup> The terms progeny, or "daughter" products, are defined as nuclides formed by the radioactive decay of another nuclide. Radon and thoron decay produces a series of radioactive daughters.

<sup>8</sup> MeV is million electron volts and is a term describing the kinetic energy of the alpha and other radioactive emissions.

mg cm<sup>-2</sup> of salt, then the plutonium alphas will be completely stopped before they reach the detector. If there is less attenuating salt, alpha particles may reach the detector with less energy.

# Alpha CAMs

The commonly used alpha CAMs are designed to account for the limited alpha particle range. Sampled air passes through a membrane filter of either 25 mm (1 in) or 47 mm (1.8 in) in diameter, and salt dust collects on the

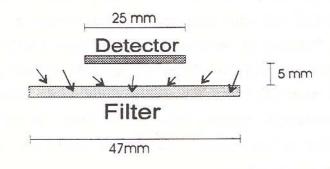


Figure 1. Detector-Filter Geometry (arrows indicate air-flow path).

surface of the sampling filter. The sampling filter is juxtaposed approximately 5 mm (0.2 in) from a 25-mm (1 in) diameter alpha detector (Figure 1). The Figure 1 filter-detector geometry allows alpha particle detection efficiency as high as 11% of the particles emitted (i.e.  $4\pi$  efficiency, Eberline, 1991).

# **Operational Considerations**

WIPP CAMs are located in the waste handling bay, underground repository, and at a monitoring station (Station A) that samples the underground air exhaust (Bartlett, 1993a). High concentrations of salt aerosol occur underground and at Station A because of mining, backfilling<sup>10</sup>, and other underground operations. By contrast, the waste handling bay is a clean, air-conditioned area. Some underground CAMs encounter relatively little salt aerosol, depending

<sup>&</sup>lt;sup>9</sup> The detection efficiency is dependent on a source-detector geometry factor. If the detector intercepts a window area on a whole sphere, the geometry is expressed as  $4\pi$ . If only half the sphere is considered, the geometry is  $2\pi$ . Most equipment manufacturers express efficiency as  $2\pi$ . A  $4\pi$  efficiency is derived by dividing  $2\pi$  efficiency by a factor of 2.

<sup>&</sup>lt;sup>10</sup> Backfill is the material that will be used to cover emplaced contact handled (CH) drums in the underground repository, as required by the DOE/State of New Mexico Consultation and Cooperation Agreement. In January 1994, the backfilling methods were demonstrated. This was a unique time to evaluate the effect of high salt aerosol on CAM performance.

on the location of mining and other operations. The most practical method for evaluating the effect of salt aerosol is comparison of the relative performance of CAMs in different locations.

All air effluent from the waste handling building passes through high efficiency particulate air (HEPA) filters, but underground air effluent is normally unfiltered. To prevent radioactive and other hazardous material releases to the environment from the underground, either Station-A or two underground CAMs must alarm to cause air effluent to be automatically diverted through HEPA filters (Bartlett 1993a; Westinghouse 1990,1995; U.S. EPA 1990). If CAM sensitivity is reduced by salt aerosol, then CAMs may not alarm and air effluent will not be automatically diverted to the HEPA filters. In such a scenario, other remedies such as fire detection or underground intervention would take on increased importance in preventing environmental releases.

## **CAM Testing**

The U. S. Department of Energy (DOE) investigators at the DOE Inhalation Toxicology Research Institute (ITRI) suggested that CAM detection efficiency may be reduced if chronically released radioactivity is buried by sampling-filter salt deposits (Seiler *et al.* 1988; Hoover *et al.* 1988,1990, Hoover and Newton 1993b). These same investigators conclude that acutely released radioactivity will not be buried by sampling-filter salt deposits and will easily be detected. If these hypotheses are correct, then CAMs may lose some sensitivity to chronic releases, but will reliably detect accidental radioactivity releases. These hypotheses do not explain the cause of the observed degraded radon-thoron progeny alpha spectra, and consequently, there is doubt that aerosols collect in the manner described.

The DOE Carlsbad Area Office (CAO) contracted with the ITRI to evaluate the effect of sampling-filter salt deposits on laboratory generated plutonium aerosol (Arthur 1993). The ITRI tests are of limited value because they do not duplicate the WIPP mining conditions, particularly the size distribution of the salt aerosol and the attachment of radioactivity to airborne particles (U. S. DOE 1994a). It is extremely difficult to simulate actual monitoring conditions in the laboratory. The testing would have to consider the full range of particle sizes, the ratio of radon-thoron progeny attached to salt aerosol, and the environmental conditions where CAMs are located (ANSI 1980, ANSI 1989).

## Use of Operational Data

An alternative to laboratory testing is to evaluate the CAM performance in the actual location where CAMs are used. Radon and thoron radioactive daughters occur naturally and emit alpha particles of energy similar to transuranic alpha radionuclides (U. S. HEW 1970, ICRP 38). The radon and thoron daughters are the source of background interference on WIPP alpha CAMs, but they are also useful indicators that CAMs are detecting radioactivity. By studying the in-situ CAM response to radon-thoron daughters, the effect of salt aerosol can be more appropriately assessed than in the laboratory where actual operating conditions are not easily reproduced.

Aerosol collection theories (Brown 1993) suggest that particle-particle forces may cause dendritic structures to form on sampling filters, and the dry-electrostatic conditions in the WIPP mine are ideal for this kind of particle interaction. Electron micrographs of WIPP filters indicate that dendritic formations are present on the sampling-filter surfaces (Bartlett, 1993a). When a sampling-filter salt deposit is formed by a dendritic process, then airborne particles will penetrate into the deposit, alpha particle energy will be absorbed, and anomalous alpha spectra will be observed. If airborne particles penetrate the sampling-filter salt deposit, then WIPP anomalous alpha spectra can be explained. More importantly though, if transuranic aerosol particles penetrate the sampling-filter salt deposit, CAMs may not be able to accurately measure acute releases of radioactivity. Consequently, it is very important to understand the mechanism of sampling-filter particle collection.

The WID has been collecting alpha CAM operational data since 1991. These radonthoron background data are recorded electronically and made available to the Environmental Evaluation Group (EEG) for analyses. This report is primarily based on operational data provided after CAM equipment modifications made in 1992.

# EQUIPMENT AND METHODS

The WIPP alpha CAMs are modified Eberline Model Alpha-6 CAMs. At strategic locations, CAMs are interfaced with specially-designed air sampling systems that collect representative air samples from the underground exhaust air. The basic CAM operation and special equipment modifications are described below.

# WIPP Alpha CAM Design

The Alpha-6 monitor has a 256-channel spectrometer capable of discriminating 238Pu and 239Pu (5.1 to 5.5 MeV) alpha particles from naturally occurring alpha radiation, particularly alpha peaks from 218Po and 212Bi (6.0 to 6.09 MeV), 214Po (7.69 MeV) and 212Po (8.78 MeV). Figure 2 shows a well-resolved, 24-hr-accumulated alpha background spectrum11 from a waste-handling-bay CAM. Background counts are subtracted using a fixed region-of-interest (ROI) method proposed by Unruh (1986) and adopted by the Alpha-6

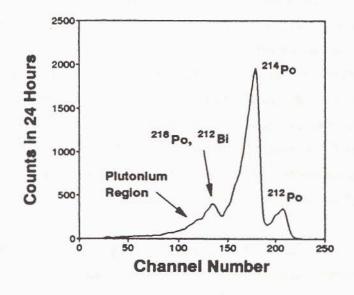


Figure 2. Accumulated Alpha Background.

<sup>&</sup>lt;sup>11</sup> At Station A, approximately 81.5 m<sup>3</sup> (2880 ft<sup>3</sup>) of air will pass through a sampling filter in a typical 24-hour sampling period.

manufacturer (Eberline 1991). As shown in Figure 3, background counts occurring in ROI-1 will be subtracted by the following method:

 $Pu_{max} = (ROI-1) - [k * {(ROI-2) * (ROI-3)} / (ROI-4 + 1)]$  (Equation 1) where

Pu<sub>net</sub> = Net counts in plutonium region

k = k-factor, constant

ROI-1 = Counts in region 1, plutonium (channels 92-126)

ROI-2 = Counts in region 1, <sup>218</sup>Po, <sup>212</sup>Bi (channels 136-143)

ROI-3 = Counts in region 1, <sup>214</sup>Po (channels 148-178)

ROI-3 = Counts in region 1, <sup>212</sup>Po (channels 179-186)

If the subtraction method is working properly, the average Pu<sub>net</sub> count rate will be zero. If the alpha spectrum is anomalous or degraded, ROI-4 may be disproportionately low and cause Pu<sub>net</sub> to be negative.

## Location of Test CAMs

There are five WIPP test CAMs discussed in this report. CAM 129 is located at the north end of room 1, panel 1 of the underground

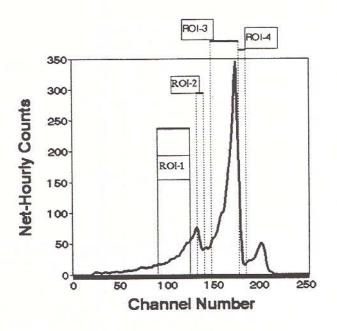


Figure 3. Regions of Interest (ROI) for Alpha Background Subtraction.

repository and in front of a room bulkhead and exhaust vent (Figures 4 and 5). There is usually little salt aerosol in room 1, panel 1.

CAM 153, CAM 157 and a prototype in-line CAM are located in the above-ground sampling station (Station A) directly above the air exhaust shaft shown in Figure 6. All mine

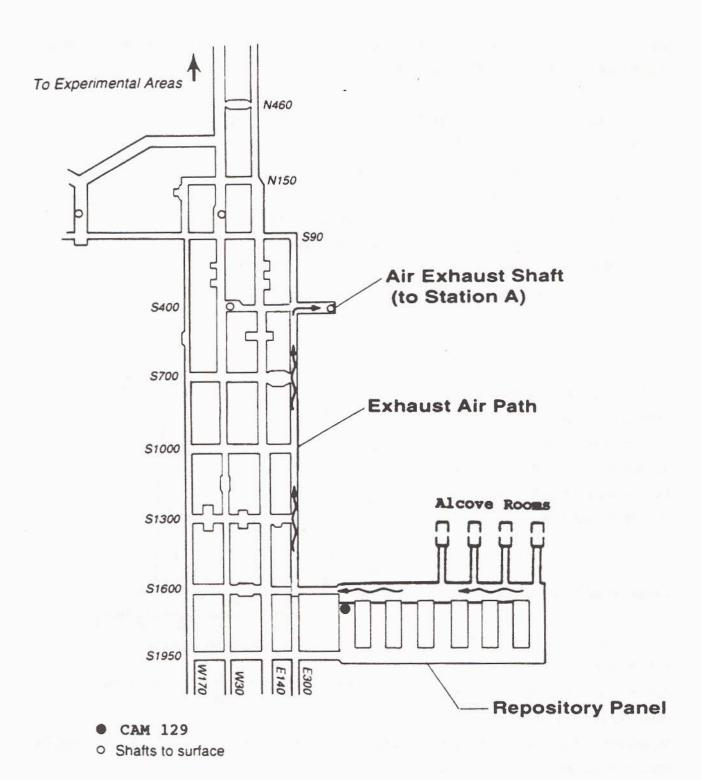


Figure 4. Main Areas of Underground Repository (CAM 129 is located in room 1, panel 1).



Figure 5. CAM 129 location in room 1, panel 1 at north end in front of barrier.

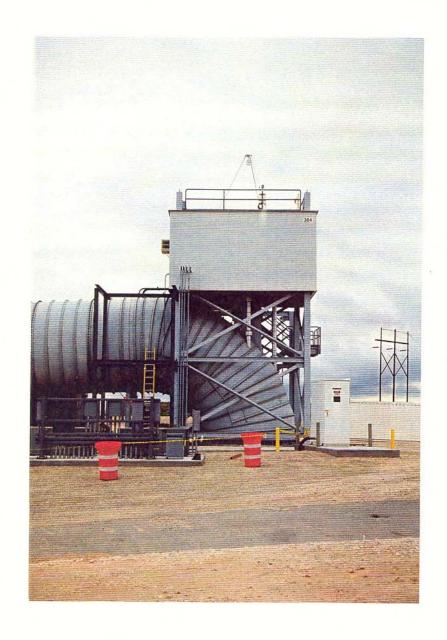


Figure 6. Station-A sampling room above the underground exhaust ventilation duct.

air is vented through the air exhaust shaft, and consequently, any salt aerosol produced underground can potentially affect the Station-A CAMs. CAM 55 is located in the waste handling bay, a clean air-conditioned receiving area for the TRUPACT-II<sup>12</sup>.

## Air sampling

Station-A CAMs are off-line monitors. Sample lines equipped with specially-designed shrouded probes extend from the Station-A sampling room into the exhaust shaft and can continuously sample the underground air effluent at a free stream velocity range of 2 to 14 m s<sup>-1</sup> (6.5 to 46 ft s<sup>-1</sup>) and at a rate of 170 L min<sup>-1</sup> (6 CFM<sup>13</sup>)(McFarland *et al.* 1989). The sampled air is pulled into three separate collection ports at 56 L min<sup>-1</sup> (2 CFM). The transmission ratio of particle sizes up to 10  $\mu$ m AD (aerodynamic diameter) through the shrouded probe is expected to be 0.93 to 1.11 (McFarland *et al.* 1989). This unique sampling system allows collection of the full range of respirable particles, including large particles.

## Particle size characteristics

In diesel-equipped mining operations similar to the WIPP, a bimodal distribution of airborne particles of 0.2  $\mu$ m and 5  $\mu$ m average aerodynamic diameter is typical (Cantrell *et al.* 1993). WIPP measurements by Newton *et al.* (1983) indicated a similar distribution and that the aerosol is primarily NaCl.

CAMs 129 and 55 are equipped with radial annulus sample heads in which aerosol enters the head from any direction around the rim at 28 L min<sup>-1</sup> (1 CFM). The radial annulus sampler allows essentially 100% penetration of particle sizes up to 6 to 8  $\mu$ m AED (aerodynamic equivalent diameter) at 28.3 L min<sup>-1</sup> to 85.0 L min<sup>-1</sup> (1 to 3 CFM) and wind speed of 1 m s<sup>-1</sup>

<sup>&</sup>lt;sup>12</sup> The TRUPACT-II is the transportation shipping container used to deliver contact handling drums and boxes to the WIPP. The TRUPACT-II will be unloaded in the waste handling bay, and drums and boxes will be taken to the underground repository.

<sup>&</sup>lt;sup>13</sup> CFM = cubic feet per minute (ft<sup>3</sup> min<sup>-1</sup>).

(3.28 ft s<sup>-1</sup>) (McFarland 1992). Both CAMs are on free-standing platforms with extended sampling heads as shown in Figure 5.

The ratio of radon-thoron progeny attached to WIPP salt-diesel aerosol is unknown. The NCRP (1989) states that mine aerosol concentrations would have to be extremely low to allow unattached fractions to exist. Other investigators suggest that the unattached fraction in diesel-equipped mines is much less than 1% (NRC 1991). The WIPP is primarily a day-shift operation, and aerosol concentration may vary widely over a 24-hr period. Alpha spectra shown in this report are primarily from day-shift operations when aerosol concentrations are expected to be high.

# Effects of filter sampling media

Fibrous sampling filters will cause alpha spectra to be degraded. Unruh (1986) noted alpha particle straggling accounts for the typical alpha detector resolution and attributed resolution problems to the filter composition. For optimum resolution, the filtering medium must be carefully selected (Hoover and Newton 1993a; Moore *et al.* 1993). Moore *et al.* (1993) showed that alpha resolution can be affected by a number of factors including filter media, self-absorption, mass loading, filter-detector gap spacing, and non-uniform aerosol deposits. In December 1992, special equipment to minimize filter-detector gap spacing variations was installed in WIPP effluent CAMs. WIPP used Versapor-3000<sup>14</sup> membrane sampling filters with CAMs 153 and 157, but in June 1995, WIPP started using the Fluoropore PTFE membrane filter<sup>15</sup> because of its reported superior performance characteristics (Hoover and Newton 1993a). The Fluoropore filter has been used with the in-line prototype CAM since August 1993.

<sup>&</sup>lt;sup>14</sup> from Gelman Sciences, 600 South Wagner Road, Ann Arbor, MI 48103-9019.

Available from Millipore, 80 Ashby Road, Bedford, MA 01730.

## Data collection

Alpha spectrometer counts are updated hourly and accumulated until the next filter change. Net-hourly spectra were derived by taking the difference of hourly accumulated spectra. Net-hourly spectra indicate immediate effects of salt loading. Accumulated spectra can mask short periods of spectral degradation as will be shown in the review of operational data.

The calculated Pu<sub>net</sub> count data are recorded every minute by a microprocessor. Alpha monitor data are retrieved and stored on a personal computer in a Turbo Pascal format (Hofer and Clayton 1991). The data, collected for several years, have been converted by the EEG for use in a Quattro Pro<sup>16</sup> spreadsheet program.

The EEG collects a sampling filter from a fixed-air-sampler (FAS) at Station-A, adjacent to the alpha effluent CAMs. The effluent CAM sampling conditions are duplicated, i.e. shrouded sampling probes are used, air is sampled at 56 L min<sup>-1</sup> (2 CFM), Versapor-3000 membrane filters are used, and filters are changed daily. Filter mass loading is determined gravimetrically after desiccation.

<sup>&</sup>lt;sup>16</sup>From Quattro Pro, Version 4.0, Borland International, Inc., Scotts Valley, CA.

## THEORY

Two questions are theoretically addressed to determine if filter mass loading could be the cause of WIPP CAM spectral anomalies. First, is filter salt loading of sufficient magnitude to cause degraded alpha spectra? If so, is there a mechanism by which incoming aerosol could penetrate a filter salt deposit? These simple questions do not consider other factors that may also contribute to spectral degradation.

# Influence of filter loading

The influence of filter salt loading is dependent on the CAM filter-detector geometry. The filter-detector geometry is two disks of 47 mm (1.8 in, filter) and 25 mm (1 in, detector) diameter with a 5-mm (0.2 in) separation (Figure 1). If radioactivity collects on the filter surface, then alpha particles will be emitted isotropically from the filter surface. Using the filter surface as the source, alpha particle direction, path length and kinetic energy can be predicted and calculated, with consideration of the influence of air and varying salt thicknesses.

A Monte Carlo computer code, termed TRIM85, was used to predict alpha particle kinetic energy after the particles pass through air and a salt layer (Ziegler *et al.* 1985). The TRIM85 accuracy for the calculated stopping cross sections averages 4% between the theoretical and empirical data for the energy range of interest (Ziegler *et al.* 1985). A theoretical <sup>214</sup>Po alpha peak spectrum is shown in Figure 7 along with spectra attenuated by 1.13 and 1.67 mg cm<sup>-2</sup> of salt. These relationships were derived by calculating the alpha particle energy after traversing an air-salt path. The particle pathway to filter angle was considered for each theoretical emission.

By numerical integration, the number and energy of emitted particles (source emissions) reaching a receptor (detector) was quantitatively determined, by a method previously described (Bartlett 1987). The calculations confirmed that sampling-filter salt deposits are of sufficient magnitude to cause <sup>214</sup>Po alpha peak amplitude to be reduced and the peak resolution to increase. The theoretical spectra are consistent with previous calculations (Seiler *et al.* 1988).

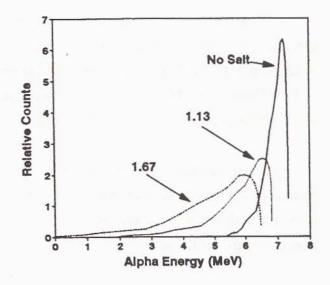


Figure 7. Theoretical reduction in <sup>214</sup>Po peaks with 1.13 and 1.67 mg cm<sup>-2</sup> of salt.

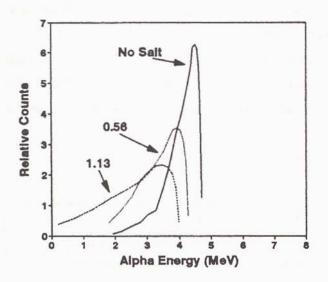
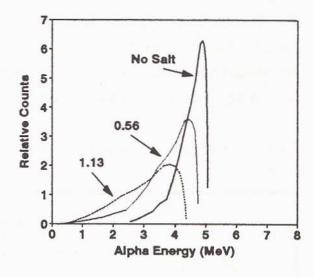


Figure 8. Theoretical reduction of 5.1 MeV (<sup>239</sup>Pu) peak with 0.56 and 1.13 mg cm<sup>-2</sup> of salt burial.



Theoretical reduction of 5.5 MeV (238Pu) peak with 0.56 and 1.13 mg cm<sup>-2</sup> of salt burial.

Figure 9.

<sup>239</sup>Pu and <sup>238</sup>Pu alpha spectra (Figures 8 and 9) were calculated in the same manner as <sup>214</sup>Po spectra. From the plutonium spectra, theoretical detector efficiencies were determined for the standard plutonium ROI-1 of 92 to 126 channels and a proposed wider plutonium ROI-1 of 65 to 126 (Hoover and Newton 1992). The results indicate that as little as 0.5 mg cm<sup>-2</sup> of salt can reduce efficiency by as much as 50% (Table 1).

These calculations presume that the salt deposit is interposed between the source and detector. The above calculations indicate the potential to degrade alpha energy. If radioactivity is mixed with the salt accumulation, then there would be less absorption of the alpha particle kinetic energy by the salt.

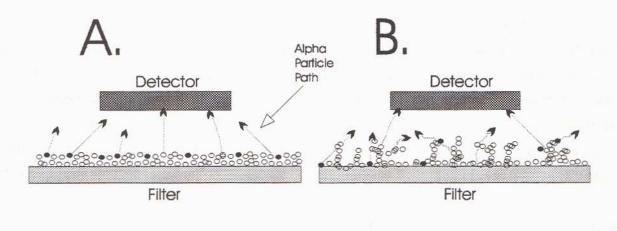
## Mechanism of aerosol collection

The data above indicate that filter salt loading is sufficient to seriously affect detection efficiency. Consequently, the mechanism of aerosol collection on a filter surface is important. Two possible particle collection mechanisms were hypothesized, although it is recognized that particles probably do not collect by a singular mechanism. If particles collect in layers, then the late-arriving particles will be near the surface of the salt deposit (Figure. 10 A.). If particles carry a high electrostatic charge, then they will be attracted to one another and form tree-like

Table 1. Theoretical <sup>238</sup>Pu and <sup>239</sup>Pu Alpha Efficiencies\*.

	Efficiency, % $(4\pi)$				
	<sup>239</sup> Pu (5.1 MeV)		<sup>238</sup> Pu (5.5 MeV)		
Depth in Salt mg cm <sup>-2</sup>	ROI 92-126	ROI 65-126	ROI 92-126	ROI 65-126	
0	10	12	11	12	
0.56	6	10	5	11	
1.13	1	6	2	10	

<sup>\*</sup>Efficiencies are relative to a 10% no-load efficiency.



Late Arriving SaltEarly Arriving Salt

Figure 10. Sampling filter particle collection mechanisms, A. mechanical collection, B. electrostatic collection.

structures on the surface of the filter (Figure. 10 B.). In Figure. 10 B., the alpha particle path may intersect salt structures and cause alpha particle kinetic energy to be reduced before reaching the detector.

The hygroscopic nature of the WIPP salt mine promotes dry, electrostatic conditions and the formation of tree-like structures called dendrites. Dendrites are common in heavily-loaded filters when examined microscopically, and they are formed by various processes including diffusional deposition and electrostatic forces (Brown 1993). The presence of dendritic structures on WIPP filters was previously reported (Bartlett 1993b), and a clean WIPP filter and one with clustering particle formations are shown in Figure 11.

Brown (1993) noted that diffusional deposition will cause particles to be captured along the entire length of a dendrite, and the rate of dendrite growth will increase as the dendrite becomes longer and more branched. Nielsen and Hill (1980) agree that electrostatic effects cause particles to collect at any point along a dendrite, in a manner similar to the diffusional deposition described by Brown (1993).

Other particle collection mechanisms are also a possibility. Very coarse dust particles tend not to clog filters, but to form a cake which contributes little to either resistance or the filter efficiency (Smissen 1971). As mentioned previously, a specially-designed sampling system is used at Station A that efficiently collects particle sizes up to and greater than  $10~\mu m$  AD. As a result, Station-A filters are likely to collect more mass and form cake-like deposits. Filter cake deposits were observed during the backfill demonstrations. Some reduction in flow rate was observed during backfill demonstrations, but filters did not clog. Fine dust has been shown to cause filter resistance to increase and clogging (Smissen 1971). Filter clogging has been rare, except when exhaust shaft water in-leakage was a contributing factor. This observation suggests that low-resistance salt matrices are forming on the filter surface when the filters are dry.

If a porous salt matrix forms on the surface of sampling filters, then the matrix can become the sampling medium, rather than the filter. If aerosol penetrates a porous matrix formed by dendrites or other particle collection mechanisms, then the alpha particle kinetic energy will be reduced. As discussed on page 12 (Effects of filter sampling media), researchers have historically recognized that alpha spectra will become degraded when the sampling media is porous, such as with a fibrous filter, or in this case a porous salt matrix.

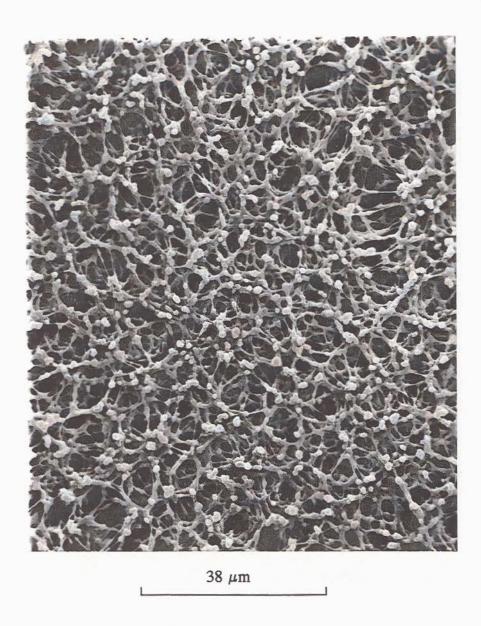


Figure 11 A. Scanning electron micrograph (top view) of a clean Versapor-3000 filter<sup>17</sup>.

<sup>&</sup>lt;sup>17</sup>Photographs in Figures 11 A. and B. were provided by the Department of Biological Sciences, Texas Tech University, Lubbock, Texas.

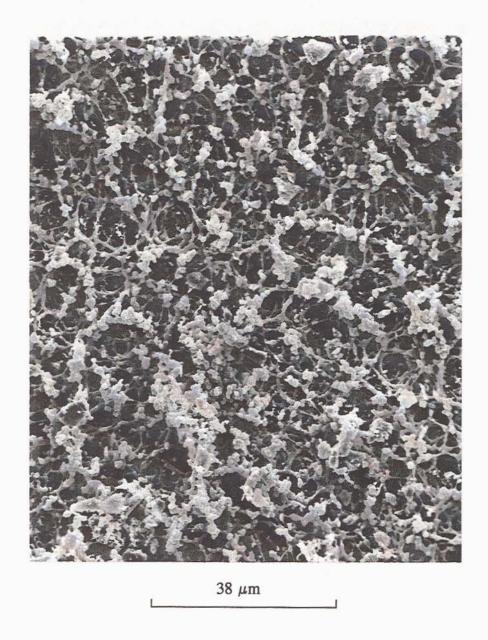
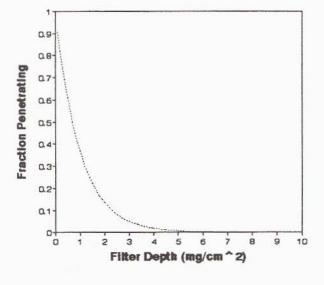


Figure 11 B. Scanning electron micrograph (top view) of Versapor-3000 filter with 0.17 mg cm<sup>-2</sup> salt loading, a very low salt loading, but particles have started piling on top of one another.

## Penetrating a filter matrix

If aerosol forms a porous matrix on the filter surface, then particle collection should be similar to that of a fibrous filter. Brown (1993) suggested that a monodisperse particulate aerosol will collect differentially on a fibrous filter with the fewest particles penetrating to the greatest depth in the filter. The degree of penetration can be described by a simple differential equation with the following solution:

where 
$$N(x) = N(o) \ e^{-\alpha x} \qquad (\textit{Equation 2})$$
 where 
$$N(x) = \text{particle concentration at depth } x$$
 
$$N(o) = \text{particle concentration at surface, } x = 0$$
 
$$\alpha = \text{layer efficiency } (\text{cm}^2 \text{ mg}^{-1})$$
 
$$x = \text{layer thickness } (\text{mg cm}^{-2})$$
 and 
$$P = N(x) \ / \ N(o) = e^{-\alpha x} \qquad (\textit{Equation 3})$$
 where 
$$P = \text{penetration fraction}$$



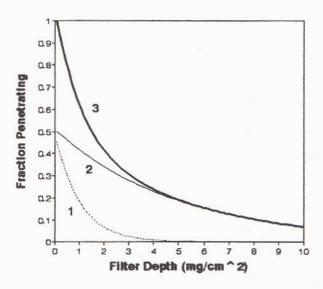


Figure 12. Monodisperse penetration of aerosol into a filter.

Figure 13. Polydisperse penetration of aerosol into a filter.

A plot of a typical monodisperse aerosol penetrating a fibrous matrix appears in Figure. 12. A polydisperse aerosol, or an aerosol with a range of particle sizes, will have a more complex penetration pattern. Brown (1993) described a two-component polydisperse particulate aerosol penetration as follows:

$$P = (1-\beta) e^{-\alpha 1 x} + \beta e^{-\alpha 2 x}$$
 (Equation 4) where

P = penetration fraction

 $\alpha 1$  = layer efficiency of first aerosol fraction (cm<sup>2</sup> mg<sup>-1</sup>)

 $\alpha 2$  = layer efficiency of second aerosol fraction (cm<sup>2</sup> mg<sup>-1</sup>)

 $\beta$  = fraction of the  $\alpha$ 2 aerosol particles

x = layer thickness (mg cm<sup>-2</sup>)

A semi-log plot of a typical monodisperse aerosol penetrating a matrix would appear as Line 1 or 2 in Figure 13. Line 1 shows a less penetrating fraction while Line 2 shows a highly penetrating fraction. Line 3 is a combination of two monodisperse aerosol fractions and is characteristic of a bimodal or polydisperse aerosol.

The WIPP effluent aerosol was reported to be bimodal (Newton *et al.* 1983). Either a monodisperse or polydisperse particle distributions will penetrate differentially into a sampling-filter matrix, as shown in Figures 12 and 13. If CAM performance variables decrease exponentially with sampling-filter salt loading, it is indicative of a particle penetration mechanism for either a monodisperse or polydisperse particle distribution.

This section establishes that normal salt loading at Station A is sufficient to cause degraded alpha spectra. In addition, the literature indicates that dendritic sampling-filter deposits are a probable collection mechanism, and the porous salt matrices will allow differential aerosol penetration into the sampling-filter deposit. These theoretical considerations suggest a more feasible method of sampling-filter particle collection than previously assumed, and the collection method provides an explanation of why alpha spectra become degraded.

## REVIEW OF OPERATIONAL DATA

In this section, operational data are reviewed in the context of the theory presented in the previous section. Radon-thoron peak height, peak resolution, and relative efficiency data were correlated with sampling-filter salt loading to determine if differential relationships were present and if the effects could be quantified. The electronically recorded alpha CAM operational data and EEG gravimetric data are used in the following analyses.

# <sup>214</sup>Po Peak Height Data

Early in 1991 the EEG graphed <sup>214</sup>Po peak height versus sampling-filter loading, and a declining relationship was found. At the time, the WID was primarily concerned with CAM redesign maintenance, and these efforts had the potential to correct the observed anomalies. Following the WIPP CAM modifications, anomalous spectra were again observed, and the EEG again found maximum 214Po peak heights were lower as sampling-filter salt deposits increased. Because anomalous spectra were present before and after equipment modifications, it became more

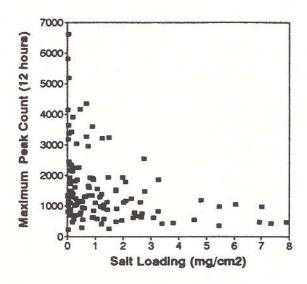


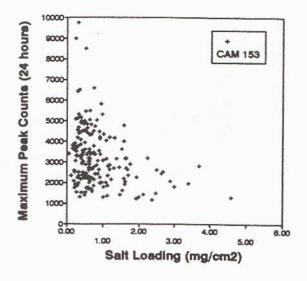
Figure 14. Maximum peak heights of <sup>214</sup>Po, CAM 153, 1991, 12-hr sampling period.

obvious that the monitor design was not the cause of the observed degraded spectra. These simple data analyses are discussed below.

From January to April 1991, CAM 153 filters were changed approximately every 12 hours. The <sup>214</sup>Po maximum peak height and filter salt loading were graphed as shown in Figure

14. The maximum peak heights in discrete salt loading ranges were fit exponentially  $^{18}$  (97% correlation) with the monodisperse particulate aerosol equation described previously (Equation 2, page 21). The resulting curve predicts the highest range of peak heights expected at any level of sampling-filter salt loading. The layer efficiency,  $\alpha$ , was derived from the curve fit, and  $\alpha$  was approximately  $0.7 \text{ cm}^2 \text{ mg}^{-1}$ . The  $\alpha$  value indicates that a 1 mg cm<sup>-2</sup> salt layer is sufficient to reduce the peak height by one half.

In November 1991, CAM 157 was installed adjacent to CAM 153, and filters were changed every 24 hours instead of every 12 hours. Corrosion resistant detectors (August 1992) and detector-source gap positioning devices (November 1992) were installed to improve CAM performance. These and other equipment modifications (Arthur 1993) seem to enhance CAM reliability, but degraded spectra were still apparent. The 1993 CAM 153 and 157 <sup>214</sup>Po peak-height data were evaluated and graphed. The trend noted in the 1991 data (Figure 14) was apparent in the 1992 and 1993 data (Figures 15 and 16). The layer efficiencies, α, were calculated as above and found to be approximately 0.47 and 0.45 cm² mg¹ and indicate 1.4 to 1.5 mg cm² salt layer is sufficient to reduce the peak height by one half. As will be shown later, averaging off-shift and day-shift data may account for the increased half-value thickness.



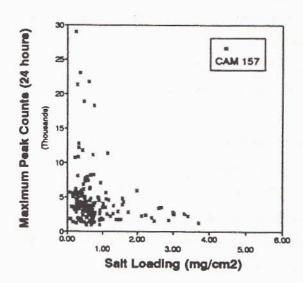


Figure 15. Maximum peak heights of <sup>214</sup>Po, CAM 153, 1992-3, 24-hr sampling period.

Figure 16. Maximum peak heights of <sup>214</sup>Po, CAM 157, 1992-3, 24-hr sampling period.

<sup>&</sup>lt;sup>18</sup> Curve fitting and statistical analyses were with PSI-Plot from Poly Software International, P.O. Box 526368, Salt Lake City, UT 84152.

# Filter Salt Loading

The 1994 filter salt loading data were derived from the EEG Station-A FAS sampling filters (Figure 17). In January 1994, backfill demonstrations in the mine produced salt aerosol concentrations that resulted in filter loading as high as 17 mg cm<sup>-2</sup>.

The January backfill demonstrations were located in an alcove room connected to the north drift \$1600 (Figure 4, page 8). The alcove air vented through the north drift of panel 1 and eventually past Station A, but did

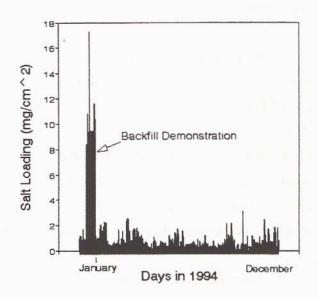


Figure 17. Station A FAS sampling filter salt mass loading.

not affect CAM 129, located in room 1, panel 1 (Figure 4). The high-flow-rate, once-through mine ventilation method subjects Station-A and underground CAMs to similar radon-thoron daughter background. CAM 129 was used as a control CAM to evaluate Station-A CAM performance because CAM 129 and Station-A CAMs have similar radon-thoron background, and CAM 129 has less filter salt loading.

The 1995 Station-A filter loading and CAM operational data were considered suspect and not used because of decreased FAS air-flow sampling rates. In March 1995, the WID examined the exhaust shaft and found significant leakage of water into the shaft at 32.2 m (105.5 ft) below the surface. The water leakage apparently contributed to the formation of salt encrustation on the EEG sampling line, moisture on sampling filters, and partial blockage of air flow. The sampling line was removed and cleaned in May 1995, but the water leakage problem has not been resolved. During this time period, crystalline salt formations were observed in electron micrographs of Station-A sampling filters, and these formations were probably a result of moisture on the sampling filters.

#### Air Flow

A reduction of air flow will proportionately reduce CAM detection sensitivity to airborne radioactivity concentrations. The EEG Station-A FAS air flow rate is regulated to 56 L min<sup>-1</sup> (2 CFM) and is expected to remain constant through the sampling period. Flow rate is recorded after filters are changed and at the end of the sampling period. In 1993, there were only 2 days when flow rate decreased more than 10% at the end of a sampling period. During the exceptionally high-salt-loading days recorded in January 1994, there were 6 days when flow rate decreased more than 10%, with a high of 29% reduction when a filter accumulated 17.3 mg cm<sup>-2</sup> of salt. The January flow rate reductions were expected because of the very high filter loading.

After the January 1994 backfill demonstrations, reduced flow rate was more frequent, even with normal levels of filter salt loading. There were 33 days (February to December 1994) when flow rate decreased more than 10%. In the first 4 months of 1995, there were 24 days when flow rate decreased more than 10%, and in 7 of these days, the flow rate was 50% to 96% less. The air flow rate reductions correspond with the identified water in-leakage and sampling-line blockage problems. Moisture and salt combined to form salt encrustation on sampling lines and filters<sup>19</sup>, and the encrustations are the most probably cause of air-flow reductions. If water or air-flow problems existed at Station A, the operational data were not used in this report to evaluate the effects of sampling-filter deposits on CAM performance data.

#### Peak Resolution

Alpha particles have discrete kinetic energies. For example, the radionuclide <sup>239</sup>Pu emits alpha particles of 5.1 MeV (kinetic energy), and if all the kinetic energy is detected, a discrete peak of 5.1 MeV will be recorded by the alpha CAM.

Alpha particles will lose kinetic energy as they traverse an air gap or interact with salt molecules. When <sup>239</sup>Pu alpha particles finally interact with the alpha CAM detector, the CAM

<sup>&</sup>lt;sup>19</sup> For the last several years, the WID has routinely removed and cleaned Station A exhaust-shaft sampling lines. Photographs of the shrouded sampling heads and lines clearly show formation of crystalline formations. The clogged sample head and large stalactitic formations were obvious in 1995.

detects a spectrum of alpha energies. For example, a discrete alpha emission of 5.1 MeV will have a broad range of energies less than 5.1 MeV (Figure 8, page 15). The width of the alpha peak is characteristic of the detector-filter geometry, the type of the sampling filter, and any other factor that causes the alpha particles to loose energy before interacting with the detector (Moore 1993).

Traditionally, alpha peak resolution is quantified by measuring the width of the peak at half the maximum height of the peak (i.e. - Full Width at Half Maximum - FWHM). Measuring the FWHM of alpha peaks is a good indicator of how alpha particle energy is affected by equipment modifications or interfering material, such as sampling-filter salt deposits.

# Accumulated versus Net-hourly Counts

Analysis of accumulated spectra may be misleading because periods of optimal performance may mask periods of poor performances. For example, resolution is usually good after a filter change. If spectra subsequently become degraded, then the degraded spectra are added to the well-resolved spectra. It may take a long time before counts from the poor spectra will significantly change the resolution of the spectrum that was initially good.

Net-hourly spectra are derived by taking the difference between two consecutive hourly-accumulated spectra. As a result, only the counts recorded in a single one-hour period are graphed and analyzed. With this method, it is immediately obvious when a spectrum becomes degraded.

The two spectral analysis methods were compared on January 25, 1994 when there were high-salt-aerosol concentrations. In Figure 18, graphs A., C., and E. are accumulated spectra on each of the Station A CAMs. At 9:00 a.m., each of the spectra had good peak resolution. At 10:00 and 11:00 a.m., the spectra were not as well resolved, but the <sup>214</sup>Po peak appears to be present. Graphs B., D. and F used the same data, but the data are displayed as net-hourly spectra. After 9:00 a.m., the <sup>214</sup>Po alpha peak is completely lost, and it is obvious that there is no peak resolution. The net-hourly method clearly indicates a loss of CAM function while the accumulated spectral method indicates only a change in peak resolution.

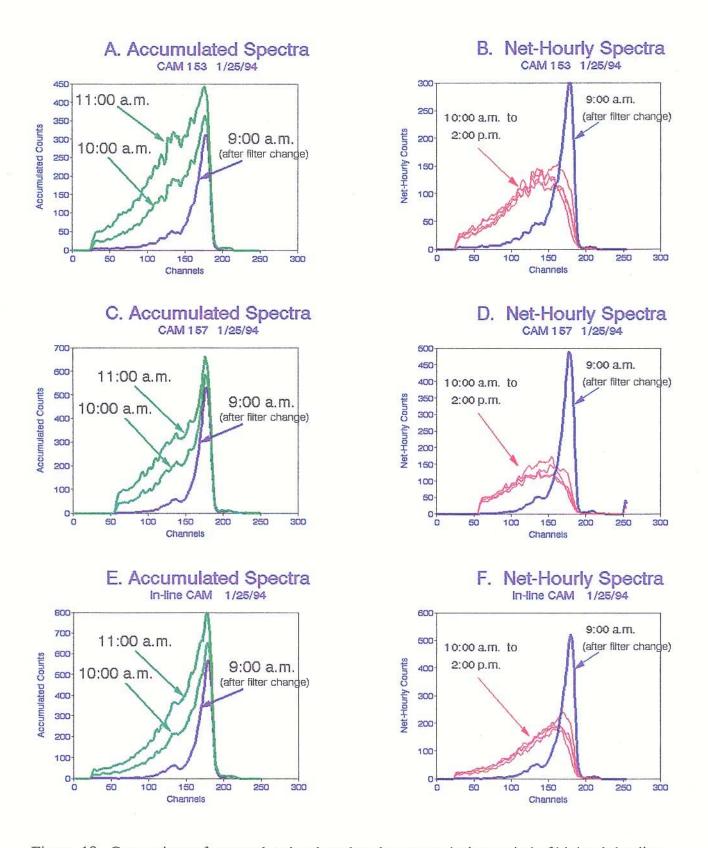


Figure 18. Comparison of accumulated and net-hourly spectra during period of high salt loading of sampling filter (11 mg cm<sup>-2</sup>).

Net-hourly <sup>214</sup>Po alpha peak resolutions with essentially no salt loading (< 0.5 mg cm<sup>-2</sup>) are shown in Figure 19. CAM 129 resolution (FWHM = 14 channels)<sup>20</sup> is slightly better than CAM 153 (FWHM = 20 channels) resolution. A slightly better resolution was expected from CAM 129 because the sampling filter has a smaller diameter.

The typical <sup>214</sup>Po alpha peaks for CAMs 55, 129 and Station-A are compared in Table 2. To obtain these data, ten low-salt loading days were chosen during March and April 1994, and the mean and standard deviation of <sup>214</sup>Po peaks calculated. As expected, CAMs with the smaller diameter filters had better peak resolution. The in-line CAM, equipped with Fluoropore filters, appeared to have slightly better resolution than CAMs 153 and 157, equipped with Versapor filters. The better in-line performance could be attributed to either the type of filter or improved particle collection characteristics of the CAM, but the data are insufficient to test these hypotheses.

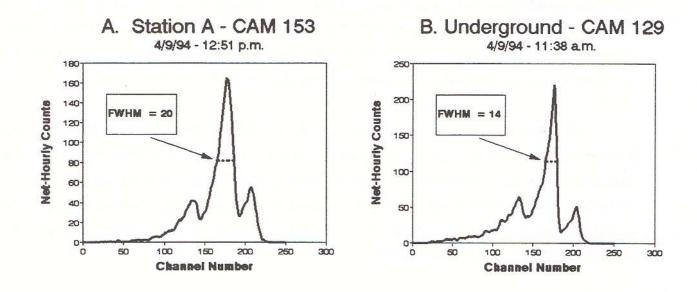


Figure 19. Ideal resolution of net-hourly spectra.

<sup>&</sup>lt;sup>20</sup> Each channel nominally represents 40 keV in alpha kinetic energy.

Table 2. Optimal 214Po Full Width Half Maximum (FWHM) Resolution.

Location	CAM Number	FWHM (Average)	FWHM (1 σ)	Filter (mm)
waste handling bay	55	14	1.4	25
room 1, panel 1	129	14	2.4	25
Station A	153	24	2.4	47*
Station A	157	24	3.9	47*
Station A	In-line	19	2.8	47*

<sup>\*</sup>Filter salt loading ≤0.5 mg cm<sup>-2</sup>

### Salt Loading and 214Po Peak Resolution

<sup>214</sup>Po peak resolutions from Station-A CAMs and CAM 129 were compared on a day when the backfill demonstration produced high concentrations of salt aerosol (9 mg cm<sup>-2</sup> salt loading on January 21, 1994; spectra at 2:00 p.m.). Station-A CAM spectra were visually poor compared to CAM 129 (Figure 20). The in-line, 153 and 157 CAM FWHMs were 50, 87, and 82 respectively, while CAM 129's resolution was ideal (FWHM = 17).

<sup>214</sup>Po peak resolutions were calculated for days in January through March 1994 when salt loading varied from very high to essentially no loading. The average FWHM during a 5-hour afternoon period was calculated and graphed as shown(Figure 21). The normal range of 14 to 20 channels FWHM was marked as a shaded band on the graph. The data indicate that as little as 1 to 2 mg cm<sup>-2</sup> salt loading shifts the FWHM out of the ideal range. At high salt loading, resolution is consistently poor.

The loss in <sup>214</sup>Po resolution at relatively low sampling-filter salt loading (1 to 2 mg cm<sup>-2</sup>) is consistent with theoretical calculations in Figure 7, page 15. Resolution is extremely poor and highly variable when salt loading is high.

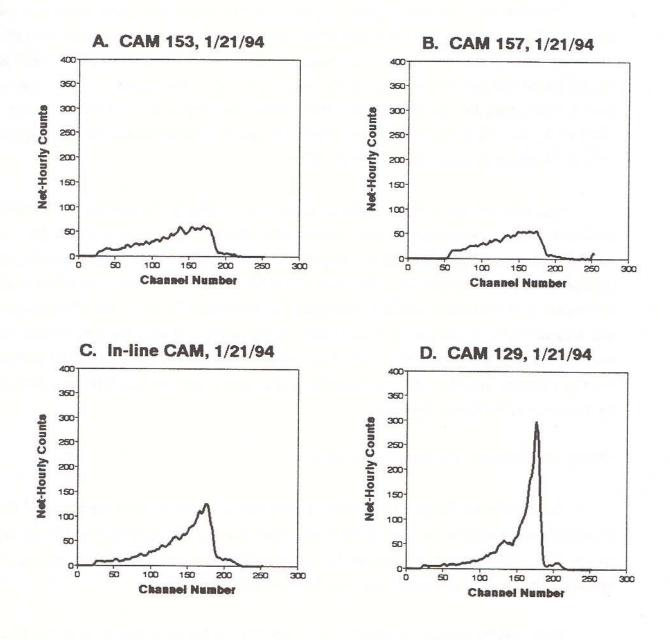


Figure 20. Station-A CAM net-hourly spectra on January 21, 1994 at 2:00 p.m.; A., B., C., are CAMs with 9 mg cm<sup>-2</sup> salt loading, D. is CAM 129 with low salt loading.

#### Resolution and Background Subtraction

The Alpha-6 CAM uses a fixed region of interest background subtraction method as shown in Figure 3, page 7. The major disadvantage of this method is that plutonium region count rate (Pu<sub>net</sub>, Equation 1, page 7) can become negative when resolution is poor. Net-hourly spectra shown in Figures 18 B., D., and F. (after 10:00 a.m.), clearly indicate poor resolution. During these times, Pu<sub>net</sub> became negative and reached hourly lows of -102 CPM<sup>21</sup> (in-line), -46 CPM (153), and -72 CPM (157). These levels are significant compared to the DOE suggested +40 CPM Station-A alarm level (Arthur 1993).

Pu<sub>net</sub> does not appear to be a reliable performance indicator. In January 1994 when salt loading was high, the operational data indicate very negative Pu<sub>net</sub> (< -100 CPM) for 3 to 5-hour periods, but subsequently the Pu<sub>net</sub> improved. In Figure 21, 5-hour averages of Pu<sub>net</sub> were compared with the corresponding <sup>214</sup>Po peak resolution data. At relatively low salt loading (0 to 2 mg cm<sup>-2</sup>), the background subtraction appears reasonably good, but at high salt loading there was a significant oversubtraction of plutonium region counts. At very high salt loading, there was only nominal oversubtraction. The Pu<sub>net</sub> variability and lack of correlation with sampling-filter salt loading indicates that Pu<sub>net</sub> depends on ROI ratios, and the ROI ratios are influenced by a number of different variables.

### <sup>214</sup>Po Peak Resolution (FWHM) Versus Time

Peak resolution changes during the sampling period. Short periods (one to several hours) of normal peak resolution are usually noted after filter changes and before salt is likely to collect on the filter. The loss of spectral resolution typically corresponds to the time of day when operations begin and salt aerosol concentrations are likely to increase.

Once spectra become anomalous, poor spectra are likely to persist until the filter is changed. A good example of this observation is a period from January 21 to 24, 1995 (Figure 22). Station-A CAMs accumulated 9 mg cm<sup>-2</sup> of salt beginning on the Friday day shift, and all Station-A CAMs exhibited poor resolution through the week end. The in-line CAM resolution

<sup>&</sup>lt;sup>21</sup> CPM = counts per minute.

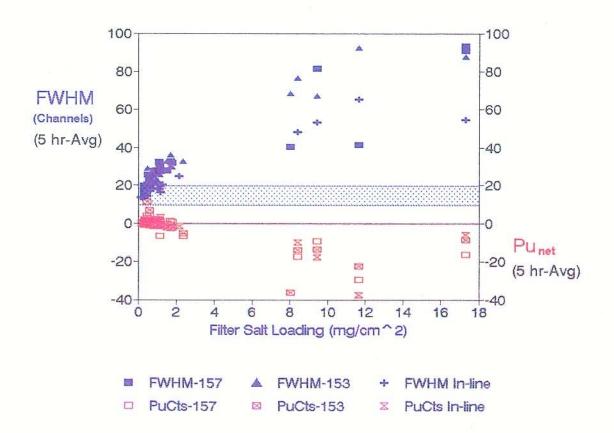


Figure 21. Comparison of Full Width Half Maximum (FWHM) peak resolution and net plutonium channel counts versus filter salt loading at Station A.

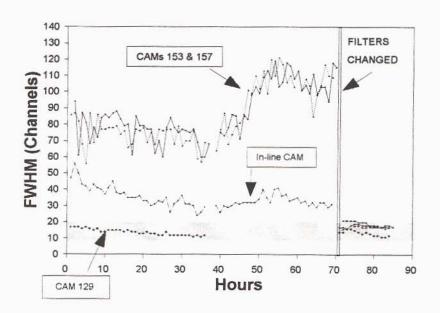


Figure 22. FWHM over week-end period and after filter change.

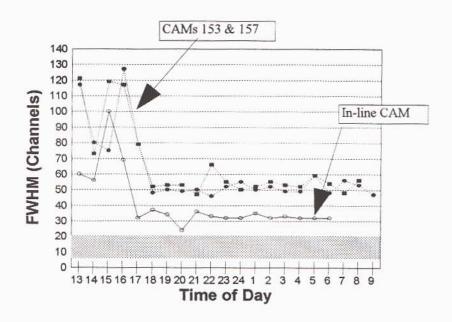


Figure 23. FWHM change during off-shift.

improved slightly on Saturday, but did not return to the normal range. After the Monday filter change, all three CAMs returned to the normal range. CAM 129 resolution was in the normal range during this period.

At other times when salt loading was high, CAM resolution improved during the evening shift, but did not return to the normal range. This trend is shown in Figure 23. In January 1995, the EEG met with WID engineers and discovered that mine exhaust is sometime reduced from 12K m³ (425K CFM) during the day, to 3.4K m³ (120K CFM) in the off-shifts. Changes in exhaust flow rate may affect particle-size distribution, the aerosol filtration mechanism, and peak resolution. The influence of these variables also needs additional study.

#### Relative Efficiency

Relative efficiency was determined by comparing total counts in each of the Station-A CAMs to total counts from CAM 129 during the same time period. The results were graphed as a ratio shown in Figure 24. When salt loading was low, all CAMs had similar total counts. As salt load increased, efficiency dropped quickly. This analysis is strong evidence that alpha particle efficiency is significantly reduced as sampling-filter salt deposits increase. The most straightforward explanation is that the aerosol is consistently buried in the sampling matrix (salt deposit) as described by Brown (1993).

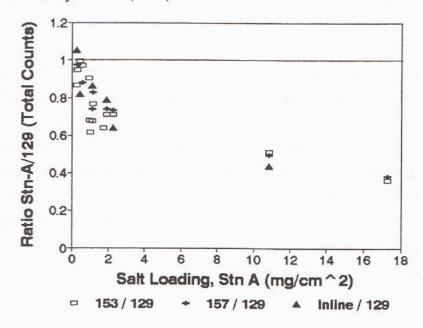


Figure 24. Efficiency of Station-A CAMs with sampling-filter salt loading.

#### DISCUSSION

The previous EEG CAM evaluation (Bartlett 1993b) was based on accumulated-alpha-spectral and net-plutonium-region-count data, and these data indicated CAMs had operational problems. In this report, analyses of net-hourly spectra, peak resolution, peak height and relative efficiency were evaluated as a function of sampling-filter salt loading. Consequently, the data give a better understanding of the root cause of spectral anomalies and the quantitative effect of sampling-filter salt loading. Theoretical calculations show that normally-occurring, sampling-filter salt deposits are sufficient to reduce alpha peak resolution and plutonium detection efficiency by 90% or more, and the empirical data indicate that alpha particle detection efficiency will be substantially reduced (50% or more) when salt loading is high.

Fibrous-filter collection theories suggest that aerosol can collect on filters in a number of ways, depending on the environmental and sampling conditions. One method, electrostatic particle collection, can cause dendritic structures to form on the filter surface, and dendrites were observed in micrographs of WIPP filters. Once significant levels of dendrites build up on the surface of a membrane filter, the filter is likely to perform like a fibrous filter, rather than a membrane filter. The anomalous WIPP spectra are similar to the historical alpha spectra obtained with fibrous filters.

If salt aerosol collected strictly in the manner described by WIPP investigators, then acutely released radioactivity would always be at or near the surface of the sampling-filter salt mass. The salt on the sampling filter would have little or no effect on the alpha spectra, and monitor performance would be expected to be optimal for acutely released radioactivity. If background spectra were normal during periods of high salt aerosol, or returned to normal during off-shifts when salt aerosol concentrations became low, then the data would support the presumed aerosol collection mechanism. To the contrary, the empirical data indicate that alpha spectra become degraded during periods when salt collects on the sampling filter, and spectra do not return to normal until the sampling filter is changed.

The EEG evaluation indicates that there are many aerosol sampling variables that will influence alpha detection sensitivity, and relatively few of these variables have been characterized at the WIPP. For example, it is generally thought that radon-thoron progeny are attached to ambient salt aerosol, but there are no empirical data to substantiate this assumption. Progeny are thought to originate from intake air, rather than underground, but there is little empirical evidence to verify this assumption. Radioactive progeny may be preferentially attached to small or large dust particles, but there are again no empirical data. If radioactivity were found to attach only to the small particle fraction, then it should be established whether the small particles will penetrate or deposit near the surface of sampling-filter salt deposits. These kinds of performance data are not available.

From our review of particle penetration theories, it appears that different particle collection mechanisms, such as inertial, diffusional, or electrostatic collection, will greatly influence particle penetration in different ways. Electron micrographs provide some evidence that dendrites form under certain environmental conditions. If so, these data suggest an electrostatic collection mechanism. In recent months, moisture has been more prevalent at Station A. Moisture on a filter will probably reduce the electrostatic charge. The collection mechanism may then be more favorable to mechanical collection with particles tending to fill openings in the filter matrix and clog the filter. Recently, much more filter clogging has been observed.

WIPP CAM usage is based on mechanical collection of salt aerosol and radioactivity on the surface of a membrane sampling filters (Seiler 1988). Although Seiler's calculations provide insight into the alpha particle detection efficiency under a particular set of conditions, the calculations do not take into account other aerosol collection theories. At WIPP, salt aerosol is usually produced during the day shifts. It was observed that spectra will rapidly become degraded from one hour to the next when salt aerosol concentrations are high (Figure 17). Spectra do not return to normal resolution after this initial insult, but rather, anomalous spectra persist until sampling filters are changed (Figure 22). These observations are consistent with a particle penetration mechanism, not mechanical collection of aerosol.

There was one instance when resolution improved during an off-shift after resolution became poor during the day (Figure 23). This example might favor Seiler's hypothesis, but once

the peak resolution became poor, the resolution did not return to normal. Consequently, this particular example does not support a mechanical collection mechanism, but rather suggests that aerosol particles are continuing to penetrating the established salt matrix. It is likely that offshift changes in the exhaust air-flow rate, from 12K m³ (425K CFM) during the day to 3.4K m³ (120K CFM) in off-shifts, account for the change in resolution seen in Figure 23. At the very least, the variability of data between day and off-shift times indicates that it is inappropriate to average day-shift and off-shift spectra, or to use the last accumulated alpha spectrum in a sampling period as a performance indicator. These operational data emphasize the importance of carefully collecting and documenting experimental data to establish particle-size distribution, particle collection mechanisms, exhaust air-flow rates, and environmental conditions.

WID has continued to determine operational status based on net plutonium channel counts and average aerosol concentrations (mg cm<sup>-3</sup>) (Donovan 1995). Net plutonium channel counts may become very negative for short periods (< 6 hours), especially during the day-shift when salt aerosol is high. These shorter periods of poor performance are not obvious if net plutonium channel counts are averaged over the entire 24 to 72-hour sampling period. Similarly, using average aerosol concentration may not be indicative of the mass collected on the sampling filter. As shown in this report, relative efficiency is a function of sampling-filter-deposit mass (Figure 24), not necessarily aerosol concentration. The use of aerosol concentrations in performance evaluations is a byproduct of assuming a single particle collection mechanism.

Accumulated alpha spectra are viewed on the Alpha-6 instrument display in order to determine CAM operational status (Westinghouse 1990). The procedure is intended to be a simple way to identify <sup>214</sup>Po peak position and operational status. As shown in the comparison of net-hourly and accumulated spectra (Figure 17), visual observation of accumulated alpha spectra can be misleading. If this procedure is to have value, sampling filters should be free of salt deposit. Optimally, the CAMs should be designed so that net-hourly spectra rather than accumulated spectra are displayed.

The amount of salt on a sampling filter appears important to CAM performance. Because of this finding, CAMs should be modified to automatically alarm when sampling-filter salt deposits become significant. Depending on the CAM location and function, the data indicate

that sampling filters should be changed when salt deposits are in the range of 0.5 to 2.0 mg-cm<sup>-2</sup> (Table 1, Figures 21 and 24).

Station-A CAMs are used to automatically actuate underground air effluent HEPA filtration. The Station A monitoring location is likely to experience high salt aerosol produced anywhere in the underground, and Station A CAMs will be affected by this salt aerosol. The overall air monitoring strategy should consider the advantages of locating CAMs downstream from the waste and in places where salt aerosol is not as much of a concern.

In 1995, water leakage into the air exhaust shaft was identified as a problem. As salt aerosol and moisture combine and collect on a sampling filter, air flow will be reduced through the sampling filter. An unusually high number of low-flow days were noted at the Station-A FAS in 1994 and 1995. It is likely that moisture was the cause. CAMs should be programmed to actuate an alarm (Eberline, 1991) at a predetermined low-flow rate, and the WIPP control room staff should be able to observe these alarms. The water leakage problem shows the influence of environmental conditions on CAM performance. Steps should also be taken to remedy the water leakage into the exhaust shaft.

#### CONCLUSIONS

The WIPP alpha CAM operational data indicate that alpha spectra and efficiency are directly affected by the amount of sampling-filter salt deposit. The CAMs are not equipped to measure the presence of these deposits, and consequently, CAM operability can not be insured in a salt-aerosol environment. The data in this report indicate that CAM operability is affected by as little as 0.5 to 2.0 mg cm<sup>-2</sup> sampling-filter salt, and these levels are routinely present at the underground-exhaust monitoring station. Depending on the usage, CAMs should not be considered operational when 0.5 to 2.0 mg cm<sup>-2</sup> of sampling-filter salt is present.

DOE investigators have claimed that WIPP CAMs will be able to detect acute releases of radioactivity, even when significant levels of salt accumulate on the sampling-filter. This claim assumes that aerosol collects by a simple mechanical process, but the empirical data are not consistent with this hypothesis. To the contrary, empirical data indicate that radioactive radon-thoron progeny penetrate sampling-filter salt deposits and background alpha spectra are significantly affected. If acutely released transuranic aerosol is collected by similar processes, then detection of transuranic radioactivity will be uncertain, and probably will be greatly reduced.

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#### **ACRONYMS**

AED Aerodynamic equivalent diameter

AD Aerodynamic diameter

<sup>212</sup>Bi A radionuclide of bismuth with an atomic mass of 212.

CAM Continuous Air Monitor.

CPM Counts per Minute.

CAO U. S. Department of Energy, Carlsbad Area Office in New Mexico.

Ci Curie, a measure of radioactivity.

DOE U. S. Department of Energy.

EEG Environmental Evaluation Group.

FAS Fixed Air Sampler, without a radiation detector.

FWHM Full Width Half Maximum, peak width at half the maximum height of the peak.

HEPA High Efficiency Particulate Air (referring to a special type of filter).

ITRI Inhalation Toxicology Research Institute, Albuquerque, New Mexico.

keV Thousand electron Volts, a measure of particle kinetic energy.

MeV Million electron Volts, a measure of particle kinetic energy.

<sup>212</sup>Po A radionuclide of polonium with an atomic mass of 212.

<sup>214</sup>Po A radionuclide of polonium with an atomic mass of 214.

Pu<sub>net</sub> Net counts in plutonium region.

<sup>238</sup>Pu A radionuclide of plutonium with an atomic mass of 238.

<sup>239</sup>Pu A radionuclide of plutonium with an atomic mass of 239.

ROI Region of Interest, refers to a group of channels on a spectrometer.

Station A Above-ground air monitoring station for WIPP underground air effluent.

WID Waste Isolation Division of Westinghouse Electric Corporation, Carlsbad, NM.

WIPP Waste Isolation Pilot Plant, Carlsbad, NM.

# LIST OF EEG REPORTS

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