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

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Abstract

Waste Isolation Pilot Plant (WIPP) alpha continuous air monitor (CAM) performance was evaluated to determine if CAMs could detect accidental releases of transuranic radioactivity from the underground repository. Anomalous alpha spectra and poor background subtraction were observed and attributed to salt deposits on the CAM sampling filters. Microscopic examination of salt laden sampling filters revealed that aerosol particles were forming dendritic structures on the surface of the sampling filters. Alpha CAM detection efficiency decreased exponentially as salt deposits increased on the sampling filters, suggesting that sampling-filter salt was performing like a fibrous filter rather than a membrane filter. Aerosol particles appeared to penetrate the sampling-filter salt deposits and alpha particle energy was reduced. These findings indicate that alpha CAMs may not be able to detect acute releases of radioactivity, and consequently CAMs are not used as part of the WIPP dynamic confinement system.

I. Introduction

This paper discusses how aerosol particle collection on alpha continuous air monitor (CAM) sampling filters influences the reliability of CAM measurements. As a consequence of this study, the design of the Waste Isolation Pilot Plant (WIPP) radioactive confinement system was reevaluated, and a number of additional facility safeguards were added to reduce risks to workers and the environment.

Alpha CAMs were installed to monitor for transuranic radionuclides in the WIPP mine exhaust air. Mine exhaust air is normally unfiltered and flows at a rate as high as 425,000 cubic feet per minute (CFM). If radioactive aerosol were detected, then air flow would be diverted to high efficiency particulate (HEPA) filters and lowered to 60,000 CFM. At the lower air flow rate, some underground operations are not allowed; at high air flow, full underground operations are allowed. Thus, effluent CAMs are important in monitoring unfiltered air flow.

The 1990 WIPP Safety Analysis Report (SAR) required CAMs to be operational whenever unfiltered air was vented.(1) If an effluent CAM was non-operational for an hour, then operations were to be curtailed. Because of the importance of effluent CAMs, the Environmental Evaluation Group (EEG) recommended laboratory and in-situ testing to establish the reliability and detection efficiency of alpha CAMs.(2)

The WIPP repository is located in a bedded-salt formation 655 m (2150 ft) below the surface. The exhaust air contains high-salt-aerosol concentrations during mining, backfilling and other underground operations. It was assumed that alpha particle detection efficiency would not be significantly affected by the salt aerosol. Aerosol particles were expected to impact on the surface of the CAM sampling filters, or on the surface of sampling-filter salt deposits. Because radioactivity would be on the surface of the filter or salt deposit, alpha particle energy would not be reduced before interacting with the CAM detector. For chronic radioactive releases, it was suggested that a saturation plutonium count rate would occur when sampling-filter-salt-deposit thickness exceeded the range of the alpha particles.(3)

The EEG reviewed CAM operational data and found that alpha spectra and radon-thoron daughter background subtraction were significantly affected by the magnitude of sampling-filter salt deposits.(2) These problems were persistent, once a salt deposit accumulated on the sampling filter. The Waste Isolation Division (WID) of the Westinghouse Electric Corporation, located at the WIPP site, addressed this problem by modifying the alpha CAM detector-filter chamber design, but spectral anomalies and poor background subtraction were still observed.(4) It became apparent that aerosol was not collecting on the surface of sampling-filter salt deposits.

In the following discussion, the alpha CAM design is reviewed, particle collection mechanisms discussed, and operational data are presented. From this information, it was concluded that CAMs could not perform their intended function, as required in 1990 SAR. Changes in the facility operations and safeguards were instituted.

II. WIPP Alpha CAM Design, Location and Particle Collection Efficiency

The WIPP alpha CAMs are modified Eberline Model Alpha-6 CAMs and are designed to account for the limited range of alpha particles. Sampled air passes through a membrane filter of either 25 mm (1 in) or 47 mm (1.8 in) in diameter, and when present, salt dust collects on the 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 (4π efficiency).

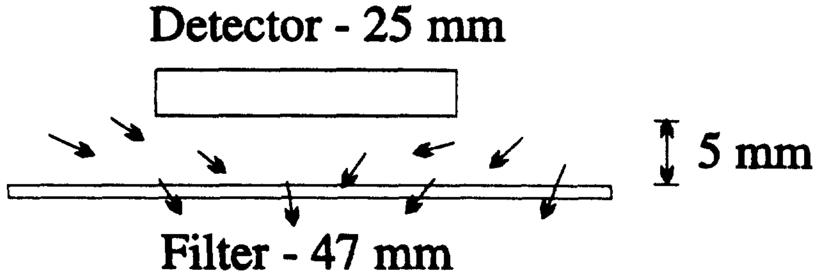

Figure 1 Detector-filter geometry (arrows indicate air-flow path).

The Alpha-6 monitor has a 256-channel spectrometer capable of discriminating ^{239}Pu and ^{238}Pu (5.1 and 5.5 MeV) alpha particles from naturally occurring alpha radiation, particularly alpha peaks from ^{218}Po and ^{212}Bi (6.0 to 6.09 MeV), ^{214}Po (7.69 MeV) and ^{212}Po (8.78 MeV). Net plutonium channel counts are derived by using a fixed region-of-interest (ROI) subtraction method(5) as shown below:

$$\mathbf{Pu_{net} = (ROI\text{-}1) - [k * \{(ROI\text{-}2) * (ROI\text{-}3)\} / (ROI\text{-}4 + 1)]} \qquad (1)$$

where

Pu_{net} = Net counts in plutonium region
ROI-1 = Counts in region 1, plutonium (channels 92-126)
k = k-factor, constant
ROI-2 = Counts in region 2, ^{218}Po, ^{212}Bi (channels 136-143)
ROI-3 = Counts in region 3, ^{214}Po (channels 148-178)
ROI-4 = Counts in region 4, ^{212}Po (channels 179-186)

Figure 2 shows a typical alpha background spectrum, and designated ROIs. If the subtraction method is working properly, the average Pu_{net} count rate will be zero. If the alpha spectrum is anomalous or degraded, ROI-4 may be disproportionately low and cause Pu_{net} to be negative.

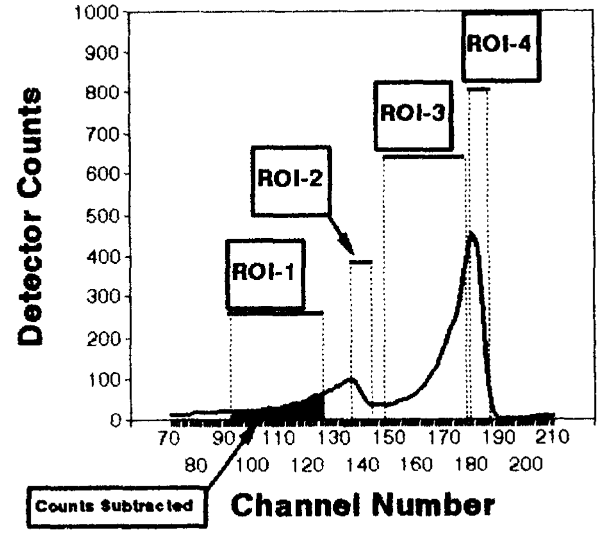

Figure 2 ROIs for alpha background subtraction.

There are four WIPP test CAMs discussed in this report. Three CAMs (153, 157 and an in-line prototype) are located in an above-ground sampling station (Station A) directly above the air exhaust shaft (see Figure 3). All mine air vents through the air exhaust shaft, and consequently, any salt aerosol produced underground can potentially affect the Station-A CAMs. A forth CAM (129) is at the north end of room 1, panel 1 of the underground repository (Figure 3). The repository horizon is approximately 655 m (2150 ft) below the surface. There is usually little salt aerosol in room 1, panel 1 and the performance of CAM 129 in a low aerosol location was compared to the CAMs at Station A where salt aerosol is most likely.

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^{-1} (6.5 to 46 ft s^{-1}) and at a rate of 170 L min^{-1} (6 CFM).(6) The sampled air is pulled into three separate collection ports at 56 L min^{-1} (2 CFM). The transmission ratio of particle sizes up to 10 μm AD (aerodynamic diameter) through the shrouded probe is expected to be 0.93 to 1.11.(6)

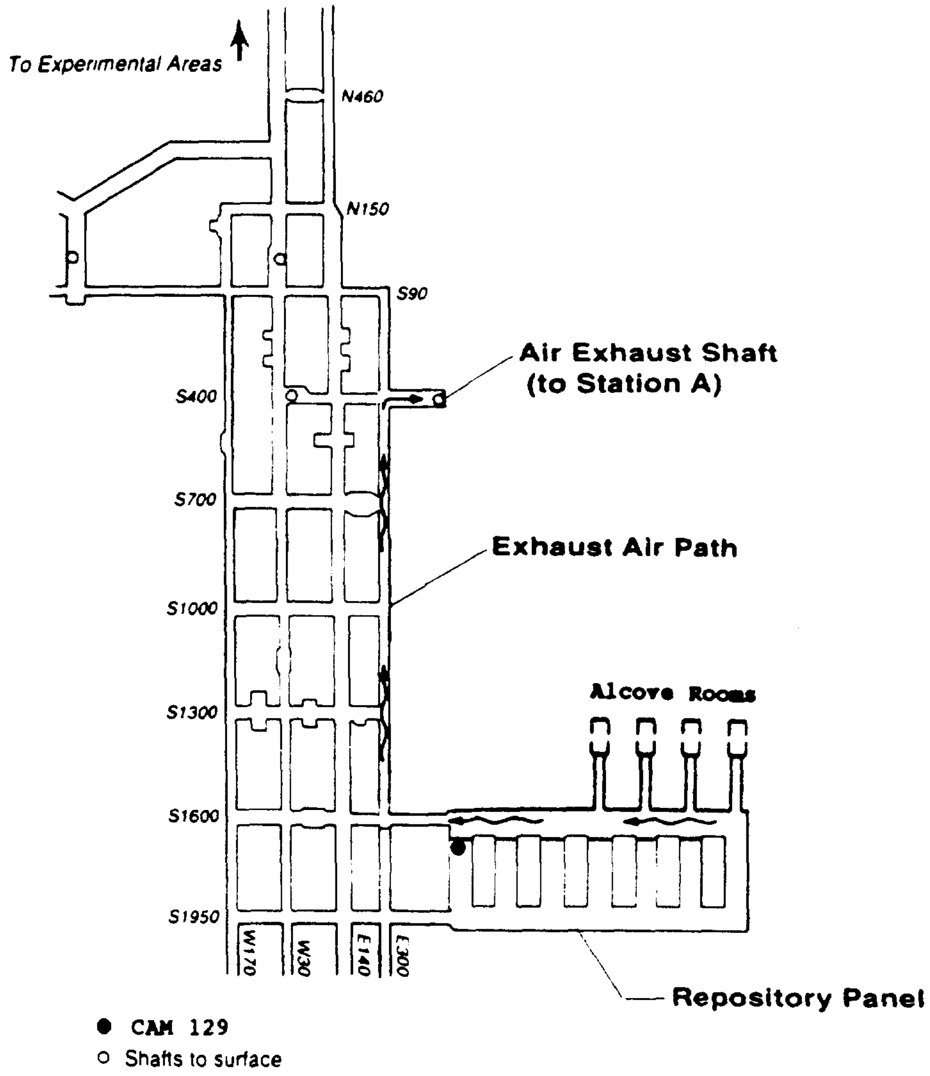

Figure 3 Main areas of underground repository (CAM 129 is located in room 1, panel 1, and station-A CAMs are located at ground level above the air exhaust shaft.)

CAM 129 is equipped with a radial annulus sample head in which aerosol enters the head from any direction around the rim at 28 L min^{-1} (1 CFM). The radial annulus sampler allows essentially 100% collection of particle sizes up to 6 to 8 μm AED (aerodynamic equivalent diameter) at 28.3 L min^{-1} to 85.0 L min^{-1} (1 to 3 CFM) and wind speed of 1 m s^{-1} (3.28 ft s^{-1}).(7)

In diesel-equipped mining operations similar to the WIPP, a bimodal distribution of airborne particles of 0.2 μm and 5 μm average aerodynamic diameter is typical.(8) WIPP measurements indicated a similar distribution and that the aerosol is primarily NaCl.(9)

The ratio of radon-thoron progeny attached to WIPP salt-diesel aerosol is unknown. The NCRP(10) 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%.(11) 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.

III. Potential for Degraded Alpha Spectra

CAM sampling-filter mass loading is as high as 2 to 3 mg cm^{-2} for underground operations, but can be in the range of 15 to 20 mg cm^{-2} during backfilling or some mining operations.(2) Twenty-four hour average air concentrations are as high as 0.3 to 0.5 mg m^{-3}, and up to 2.5 to 3.3 mg m^{-3} in extreme conditions.

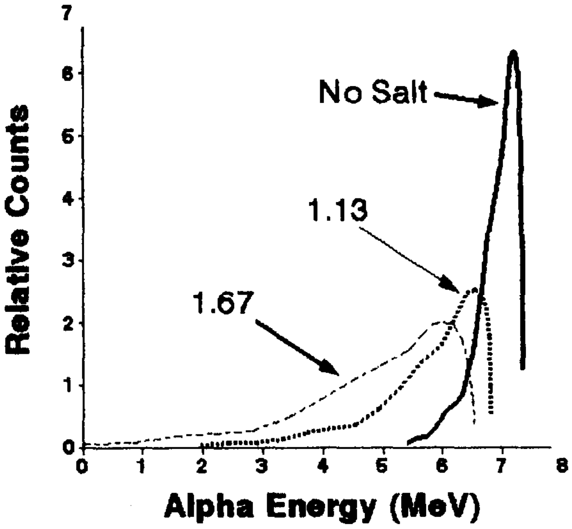

Figure 4 Theoretical reduction a ^{214}Po alpha spectrum with 1.13 and 1.67 mg cm^{-2} salt interposed between the source and detector.

Alpha measurements are dependent on the CAM filter-detector geometry (Figure 1). Radioactivity on the sampling-filter surface will emit alpha particles isotropically. Using the filter surface as the source, alpha particle direction, path length and kinetic energy were predicted and calculated, considering the influence of air and varying salt thicknesses.(4) Figure 4 shows a calculated ^{214}Po (7.69 MeV) alpha spectrum and the effect of interposing as little as 1 mg cm^{-2} of

Table 1 Theoretical ^{238}Pu and ^{239}Pu Alpha Efficiencies*.

Depth in Salt mg cm^{-2}	Efficiency, % (4π)			
	^{239}Pu (5.1 MeV)		^{238}Pu (5.5 MeV)	
	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

*Efficiencies are relative to a 10% no-load 5.1 MeV efficiency.

salt between the source and detector. The spectral effects of interposed salt are more pronounced when less energetic ^{238}Pu (5.5 MeV) and ^{239}Pu (5.1 MeV) alpha particles are the source. The theoretical efficiency of the Alpha-6 was calculated as shown in Table 1.(4) Changing the lower ROI-1 discriminator setting from 92 to 65 improves theoretical plutonium efficiency, but the overall CAM performance may be affected by other factors such as false alarm rates or background subtraction.

IV. Particle Collection Mechanisms

The EEG collects fixed-air sampling filters each day at the above ground sampling site, Station A. Salt particles ranging in physical size up to 7 μm in diameter were observed using a scanning electron microscope. Microscopic analyses also revealed numerous dendritic structures on the sampling-filter surface. If exposed to high humidity the dendritic structures tend to collapse and form a confluence.(2) Sputtering the sampling-filter surface, as a preparation for electron microscopic analysis, also disrupted the dendritic structures, as did viewing with a scanning electron microscope for long periods. In general, the sampling-filter salt deposits appeared loosely formed and porous. If observed with a light microscope, the dendritic structures remained stable.

The microscopic observations strongly indicated that the aerosol particles are electrostatically bound. The dry, hygroscopic nature of the WIPP salt repository favors electrostatic buildup, and particle-to-particle interactions were evident in our observations. There are no pressure drop measurements across the sampling filter, but air flow is maintained relatively constant by flow control devices. Data from 1993 indicated only 2 days during the year when air-flow rate decreased by more than 10% during a 24-hour sampling period. The lack of filter clogging suggested that the particle packing fraction was low and that air easily passed through the sampling-filter salt deposit.

Because of these observations, it was hypothesized that the sampling-filter salt deposits may behave more like a fibrous filter than a membrane filter. If so, aerosol would penetrate differentially into the salt deposit. As the salt layer becomes thicker, more particles penetrate deeper into the salt deposit. The deeper the aerosol penetrates into the sampling-filter salt deposit, the greater the potential exists for reduced alpha particle energy and poor alpha spectra.

It was 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,(12) and particle penetration was described by a simple differential equation with the following solution:

$$\mathbf{N(x) = N(o)\, e^{-\alpha x}} \qquad (2)$$

where
$N(x)$ = particle concentration at depth x
$N(o)$ = particle concentration at surface, $x = 0$
α = layer efficiency ($cm^2\ mg^{-1}$)
x = layer thickness ($mg\ cm^{-2}$)

and

$$\mathbf{P = N(x) / N(o) = e^{-\alpha x}} \qquad (3)$$

where P = penetration fraction

A plot of a hypothetical, monodisperse aerosol penetrating a fibrous matrix appears in Figure 5. A polydisperse aerosol was described as having a more complex penetration pattern and for a bimodal distribution was characterized as follows:(12)

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

where
P = penetration fraction
$\alpha 1$ = layer efficiency of first aerosol fraction ($cm^2\ mg^{-1}$)
$\alpha 2$ = layer efficiency of second aerosol fraction ($cm^2\ mg^{-1}$)
β = fraction of the $\alpha 2$ aerosol particles
x = layer thickness ($mg\ cm^{-2}$)

A plot of a typical monodisperse aerosol penetrating a matrix is represented by either line 1 or 2 in Figure 6. Line 1 shows a highly penetrating fraction while line 2 shows a less penetrating fraction. Line 3 is a combination of two monodisperse aerosol fractions and is characteristic of a bimodal or polydisperse aerosol.

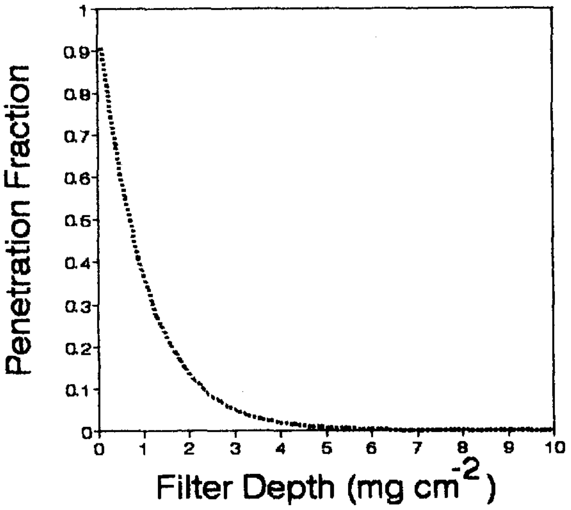

Figure 5 Monodisperse penetration of aerosol into a filter.

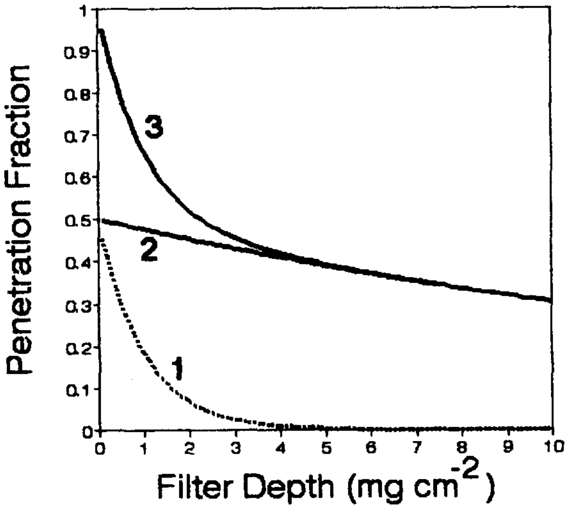

Figure 6 Polydisperse penetration of aerosol into a filter.

IV. Operational Data

Early in 1991, the maximum ^{214}Po peak height at the end of a 12-hour sampling period was evaluated as a function of sampling-filter salt loading, and a declining relationship was found (Figure 7). Following extensive CAM modifications in 1991 and 1992, a similar analysis was performed for a 24-hour sampling period, and a declining relationship was again found (Figure 8). These simple analyses indicated that sampling-filter salt loading was affecting alpha spectra and suggested the need for additional analyses.

CAM sampling filters are changed each morning before underground activities begin, and with a clean filter in place, well resolved spectra begin accumulating. Spectra typically become degraded mid-morning when underground operations begin. The poor spectra persist until filters are changed.

An example of the effects of sampling-filter salt loading occurred on January 25, 1994 when backfill demonstrations were conducted in the alcove rooms (Figure 3). Hourly spectra from 9:00 a.m. to 1:00 p.m. are shown for the underground CAM 129 and Station A CAMs (153, 157, and in-line) in Figure 9. The Station A CAMs accumulated 11 mg cm^{-2} during this sampling period, whereas CAM 129 was out of the salt aerosol air flow and accumulated very little salt deposit. The Station A CAM spectra became severely degraded after 9:00 a.m and continued to be degraded until the sampling filters were changed the next day. CAM 129 spectra remained well resolved.

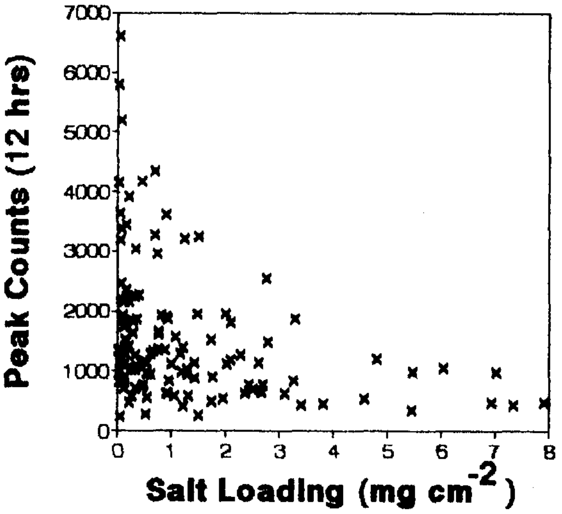

Figure 7 Maximum peak height of ^{214}Po, CAM 153, 1991, 12-hr sampling period.

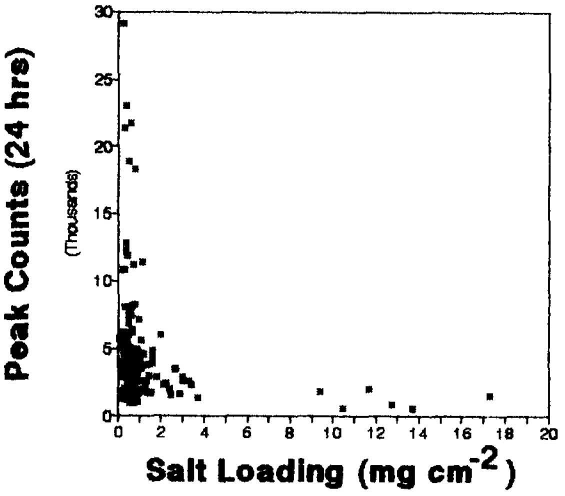

Figure 8 Maximum peak height of ^{214}Po, CAM 157, 1992-3, 24-hr sampling period.

In addition to maximum peak height, the full-width-at-half-maximum (FWHM) of ^{214}Po alpha peaks was used as a performance indicator for alpha spectra. ^{214}Po peak resolutions were calculated for days in January through March 1994 when salt loading varied from near zero to as high as 17 mg cm^{-2}. The average FWHM during a 5-hour afternoon period was calculated and graphed as shown in Figure 10. The FWHM is normally in the range of 14 to 20 channels. The data indicate that as little as 1 to 2 mg cm^{-2} salt loading causes the FWHM to be greater than 20. The increase in FWHM and loss in ^{214}Po resolution at relatively low sampling-filter salt loading (1 to 2 mg cm^{-2}) is consistent with theoretical calculations in Figure 4.

The net counts in the plutonium ROI (Pu_{net} from Equation 1), were also plotted in Figure 10 as a function of filter salt loading. The data indicated periods of very negative Pu_{net} counts (< -100 CPM) for 3 to 5-hour periods. Compared to an effluent alarm setting of 40 CPM, these negative excursions are significant. At relatively low salt loading (0 to 2 mg cm^{-2}), the background subtraction appears reasonably good. At salt loading above 2 mg cm^{-2}, there was consistent oversubtraction of plutonium region counts. At very high salt loading (18 mg cm^{-2}), the oversubtraction was not as pronounced, but alpha peaks were essentially non-existent. Pu_{net} was not a consistent CAM performance indicator, but at times, the oversubtraction of plutonium background counts was so extreme that the monitor could not be considered operational.

The total spectrum counts in each of the Station-A CAMs were compared to total counts from CAM 129 during the same time period. The results were graphed as a ratio shown in Figure 11. When salt loading was low, all CAMs had similar total counts. As salt load increased, efficiency dropped quickly. These data are indicative of the quantitative reduction of CAM efficiency as salt

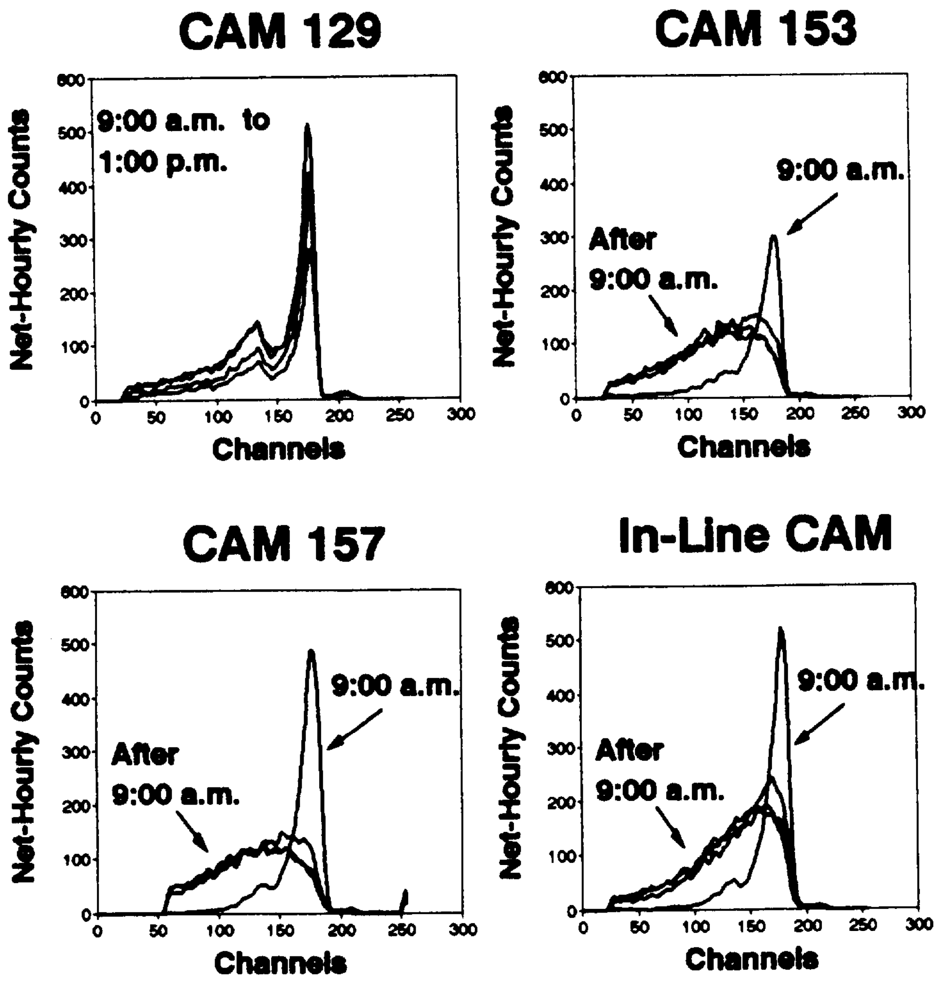

Figure 9 CAM alpha spectra on January 25, 1994.

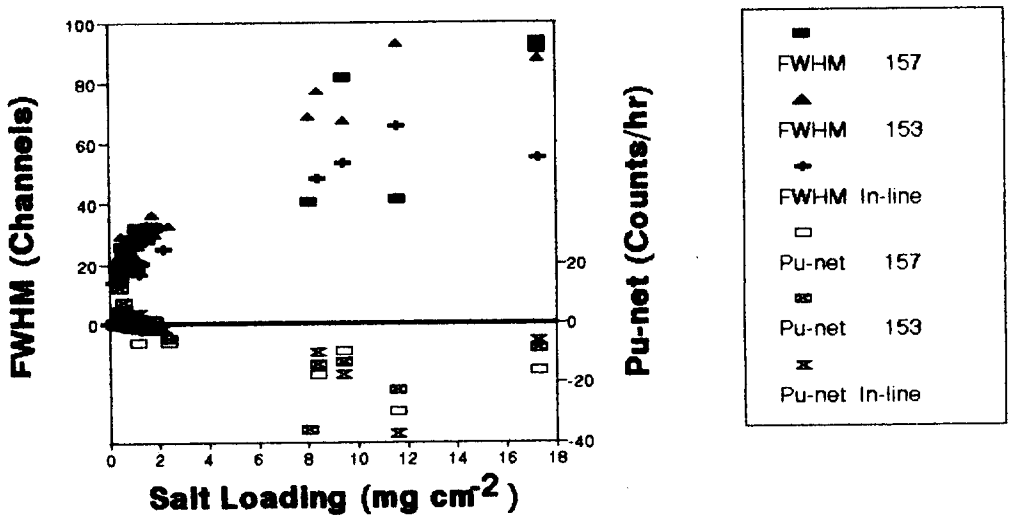

Figure 10 FWHM and Pu_{net} as sampling-filter salt deposits increase.

deposits become greater. The most straightforward explanation is that aerosol particles are penetrating the sampling-filter salt deposit, and alpha particle energy is reduced by interaction with the salt deposits.

If aerosol penetrates a fibrous filter and the sampling-filter salt deposit in a similar manner, then it would be expected that alpha detection efficiency would decrease exponentially as salt deposits increase. The observed relative efficiency data was fit to the exponential equation, e^{-bx} (Figure 12). At the 95% confidence level, data up to 17 mg cm^{-2} yielded a goodness of fit of 0.97. Because previous empirical data(9) and information(8) suggest that the WIPP mine aerosol is a bimodal distribution, data below and above 2 mg cm^{-2} were fit independently with the same exponential equation. The data below 2 mg cm^{-2} yielded a goodness of fit of 0.89, the data above 2 mg cm^{-2} were fit at 0.99. Each of the analyses indicates an exponential loss of counts on CAMs with salt-laden filters. The data suggest that a bimodal distribution is probable, but additional work is needed to confirm this hypothesis.

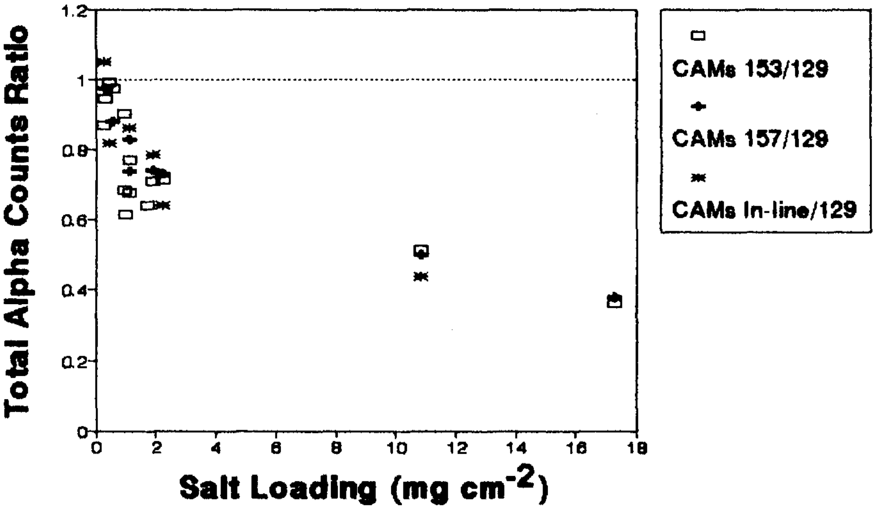

Figure 11 Relative efficiency of station-A CAMs to CAM 129 as a function of salt loading at Station A.

V. Discussion and Conclusions

The operational data and fibrous-filter collection theories suggest that degraded alpha spectra and poor background subtraction are attributed to penetration of aerosol particles into preestablished sampling-filter salt deposits. A number of variables were correlated with sampling-filter salt loading, and each case, there were direct correlations with salt loading. Electron micrographs revealed porous salt deposits and dendritic structures on the surface of the sampling filters. It appeared that once a significant level of salt deposit built up on the surface of a membrane filter, then the filter performed like a fibrous filter, rather than a membrane filter.

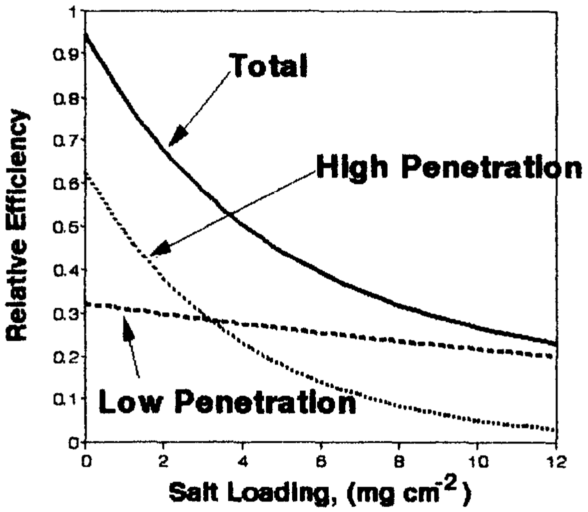

Figure 12 Best fit analysis of relative efficiency data.

If sampled aerosol collected strictly as layers on the surface of the sampling filter, then acutely released radioactive aerosol could be measured, regardless of the sampling-filter salt mass. Neither the reviewed data nor the fibrous-filter collection theories support such a limited mechanism for particle collection. In fact, more questions are raised about the complexity of the aerosol collection mechanism than are resolved. 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. It is not known whether radioactive progeny will preferentially attach to small or large dust particles. If radioactivity were found to attach only to the small particle fraction, then the collection mechanism of small particles would need to be studied. Although it is suspected that aerosol particles are predominantly collected by an electrostatic mechamism, other mechanisms can not be ruled out. And most importantly, it is not known whether transuranic aerosols would behave similarly to those of radon-thoron progeny.

The amount of salt on a sampling filter appears to be a much more important variable than the average-salt-aerosol concentration. Because of this finding, the mass of sampling-filter deposits should be carefully documented, and CAMs ideally should alarm when sampling-filter 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^{-2} (Table 1, Figures 11).

Effluent alpha CAMs are no longer a part of the limiting conditions of operations or the dynamic confinement system at the WIPP. Instead, WIPP will reduce potential radioactive uptake risks to workers and the environment by eliminating the operational backfill, restricting radioactive content of waste drums, using underground barriers to control potential releases and fires, modifying ventilation, relocating critical CAM monitors, and changing sampling filters when significant salt buildup is likely.

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DISCUSSION

BRESSON: I thought I heard you say that when you were making measurements and mining operations were being conducted there was a period of enhanced salt concentration in the air. Is that correct?

BARTLETT: That is correct.

BRESSON: Do you have any data that show how the alpha CAM spectrometer system functions when there isn't that kind of activity going on? I would expect such activity would be typical of operations at WIPP. The question is, how do alpha CAMs function when you are not disturbing the environment and creating the particles?

BARTLETT: That's a good question. Figure 2 shows a typical alpha spectrum that you would see at Station A. When there is no salt, the instrument has good resolution, as shown in Figure 2. Your question is interesting because we have been asked this before. Some people think we can not do alpha spectroscopy in a mining environment, but we have five years' worth of electronic data to the contrary. The monitors work well when there is no salt aerosol.

BRESSON: So it is possible to come up with an operational scheme whereby you can rely on the air monitoring system to do its job except during periods when you are deliberately adding contaminants.

BARTLETT: That is very true. In fact, that has been one of our recommendations. The problem is determining when you are going to have salt aerosol collecting on the filter. Rather than manually determining when spectra are poor, it would be advisable to have the monitor automatically recognize poor spectra. Ventilation air could then be diverted to HEPA filters, or other operational options could be considered.

BRESSON: Does the particle size of the salt seem to be several micrometers in diameter?

BARTLETT: Yes, in electron micrographs the largest sized particle we have seen is about 7μm in diameter. There have been other studies by ITRI Research in Albuquerque, NM, in the mid-80's, and they reported particle sizes in the 3-5μm range.

BRESSON: Would a prefilter on the detector system filter out the larger salt particles but allow passage of PuO_2 particles?

BARTLETT: It is not known whether the Pu particles would be attached to the salt.

ENGLEMANN: What is the size distribution of the salt, and of the anticipated plutonium? The geometry you show suggests that salt may deposit around the periphery of the detector, on the filter, and the smaller plutonium makes it hard to get under the detector. Have you considered or tried a cascade impactor?

BARTLETT: There could be a wide range of Pu particle sizes. As mentioned previously, salt particle sizes range up to about 3-5μm.

ENGLEMANN: And larger for the salt?

BARTLETT: That is for the salt.

ENGLEMANN: Now it occurs to me to ask if you have tried the cascade impactor where you would get the activity in the final stage.

BARTLETT: I think that is a good recommendation. That is one of our recommendations, too.

ENGLEMANN: Another question is concerned with the geometry in which the detector is close to the filter. One would expect the salt and the larger particles to be peripheral to the detector. Are they able to make the turn and head toward the center of the filter? I wonder to what extent you are sure that you have a problem, and whether it can be corrected with the geometry.

BARTLETT: Perhaps I have given the wrong impression. Let me go back a bit. The question of geometry and uniform deposition of particles across the filter has been the subject of other studies. As a result of the studies, the collection chamber was redesigned to preclude this problem. I showed you two CAM spectra from station A. One was an older design. Another one was a new design that gives a much more uniform distribution of the particles across the face of the filter. Am I addressing your question?

ENGLEMANN: Perhaps if the fix isn't to correct uneven distribution across the filter, you can collect your large particles around the outside and let the rest go to the center.

BARTLETT: That is difficult.

ENGLEMANN: At any rate, I assume you have studied it?

BARTLETT: Those are questions that have been considered. We have not studied it *per se*, because others have looked at it. I believe these concerns have been addressed by the WIPP project.