WHC-SA--0173

DE88 013676

On-Line Measurement of Stack-Gas Particulate Radionuclides

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Date Completed
November 1987

DISCLAIMER

Prepared for the U.S. Department of Energy
Assistant Secretary for Defense Programs

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P.O. Box 1970
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Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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ABSTRACT

This paper describes an advanced process-monitioring system for the automatic sample collection, analysis, reporting, and alarming of the concentration of long-lived radionuclides in nuclear process stack-gas effluents. This system, the Moving Filter Radioactive Aerosol Monitor, collects particulates in a conditioned sampling chamber, under computer control, for subsequent and separate alpha and beta counting and analysis. The alpha measurement is performed with a solid-state surface-barrier detector. Beta analysis is performed with a phoswich scintillation detector. A unique alpha-energy analysis program that provides automatic energy calibration and lowered detection limits for plutonium in the presence of high quantities of interfering ^{212}Bi/Po is described. The ^{212}Bi/Po alpha results are further used to monitor system quality and to compensate for the radon daughter beta components in the beta system. Results of the system calibration and detection limits are also presented.

CONTENTS

INTRODUCTION

Stack gases from nuclear facilities can contain both natural and man-made radionuclides. Interferences from natural radon daughters limit the detection capability of common radiation measurement methods. An improved system using on-line multichannel analyzers, alpha energy spectroscopy, gamma-ray analysis, radon compensated gross beta counting, and automated data reduction is presented. This system provides accurate determination of certain stack effluent fine particulates as air concentrations with detection limits at or near the Derived Concentration Guides (DOE 1985).

This system, the Moving Filter Radioactive Aerosol Monitor (MFRAM), is an adaptation of the Transuranic Aerosol Measurement System (Kordas 1980)(TAMS) developed by the Lawrence Livermore Laboratory. The MFRAM comprises two moving filter assemblies (MFA), an operator control console, a multiplexed process signal input subsystem, detector nucleonics, and embedded microcomputers for control and data reduction (fig. 1). Each MFA comprises an in-line heating unit, sample chamber, filter stepping mechanism, counting chamber, and detectors (fig. 2). The MFRAM controls the sample stream temperature to minimize mass formations that degrade alpha spectroscopy results. It features an alpha energy analysis (AEA) algorithm (Troyer 1987) for automatic internal energy calibration with resolution compensation and spectral peak extraction. The AEA program reports ^{239}Pu, ^{212}Bi, ^{212}Po as concentrations; ^{212}Po concentration is used in an algorithm that compensates for the radon contribution to gross beta measurements. The resultant beta determination is the long-lived radionuclide gross beta concentration.

DISCUSSION

The MFRAM periodically collects the sample in one chamber and transports it to a separate evacuated chamber for analysis. This separation allows the sample stream to be heated to 105 °C, minimizing most overburden mass. Such a configuration is desirable where the effluent contains high moisture and NO_x concentrations, which destroy any detectors placed in the sample stream. The arrangement also allows the use of laboratory equipment and methods for on-line process analysis.

High-resolution alpha spectroscopy is obtained from a large area surface barrier detector and a computer algorithm to yield accurate analyses for ^{239}Pu, ^{212}Bi, and ^{212}Po. A special CaF(Eu)/NaI(Tl) phoswich scintillation beta-gamma anticoincidence detector is used to minimize radon influence on the beta channel. Additional radon daughter compensation is provided by the ^{212}Po AEA. The gamma-ray analysis from the phoswich detector is useful for both detector stabilization and analysis.

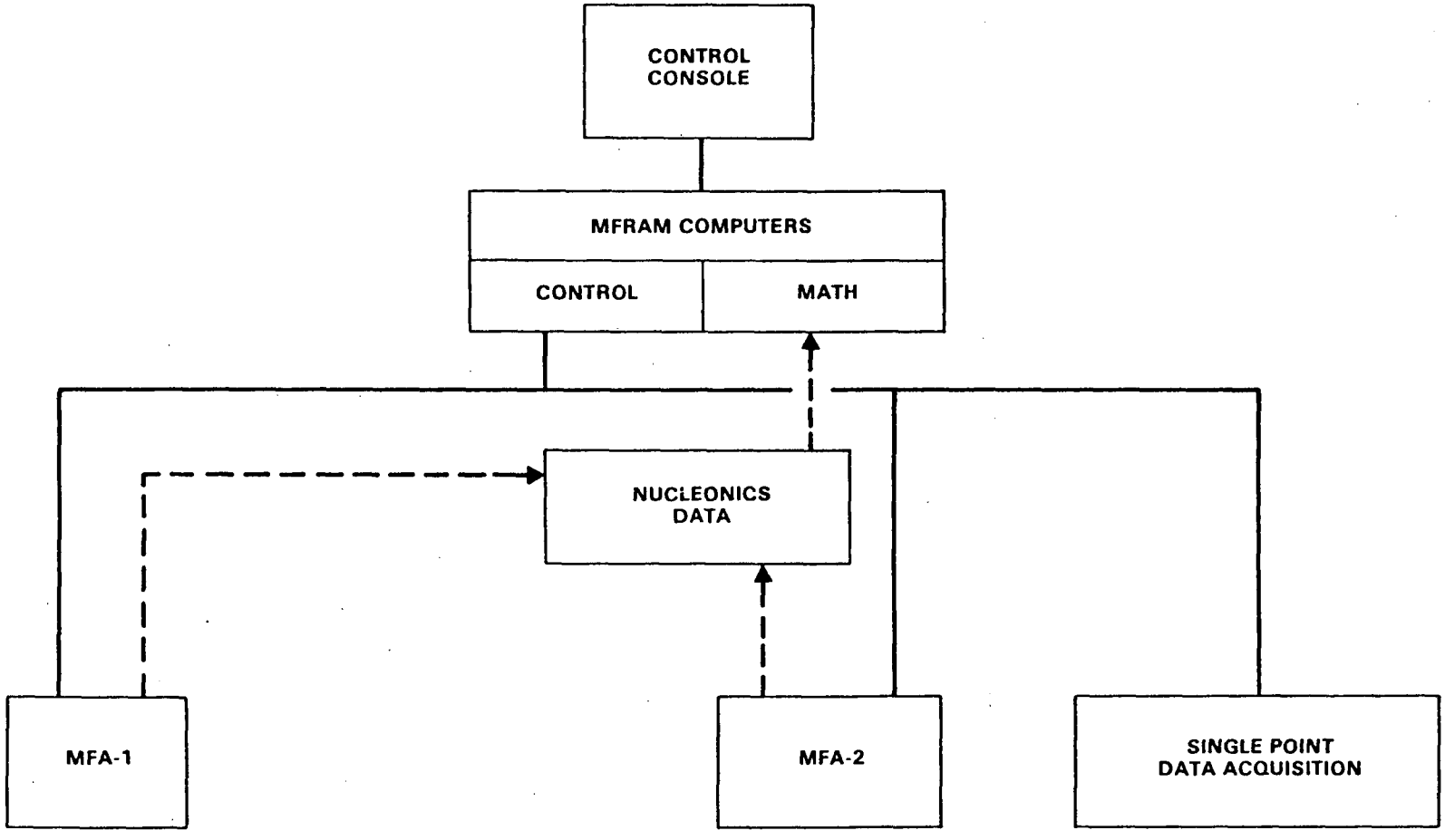

Figure 1. Moving Filter Radioactive Aerosol Monitor System Block Diagram.

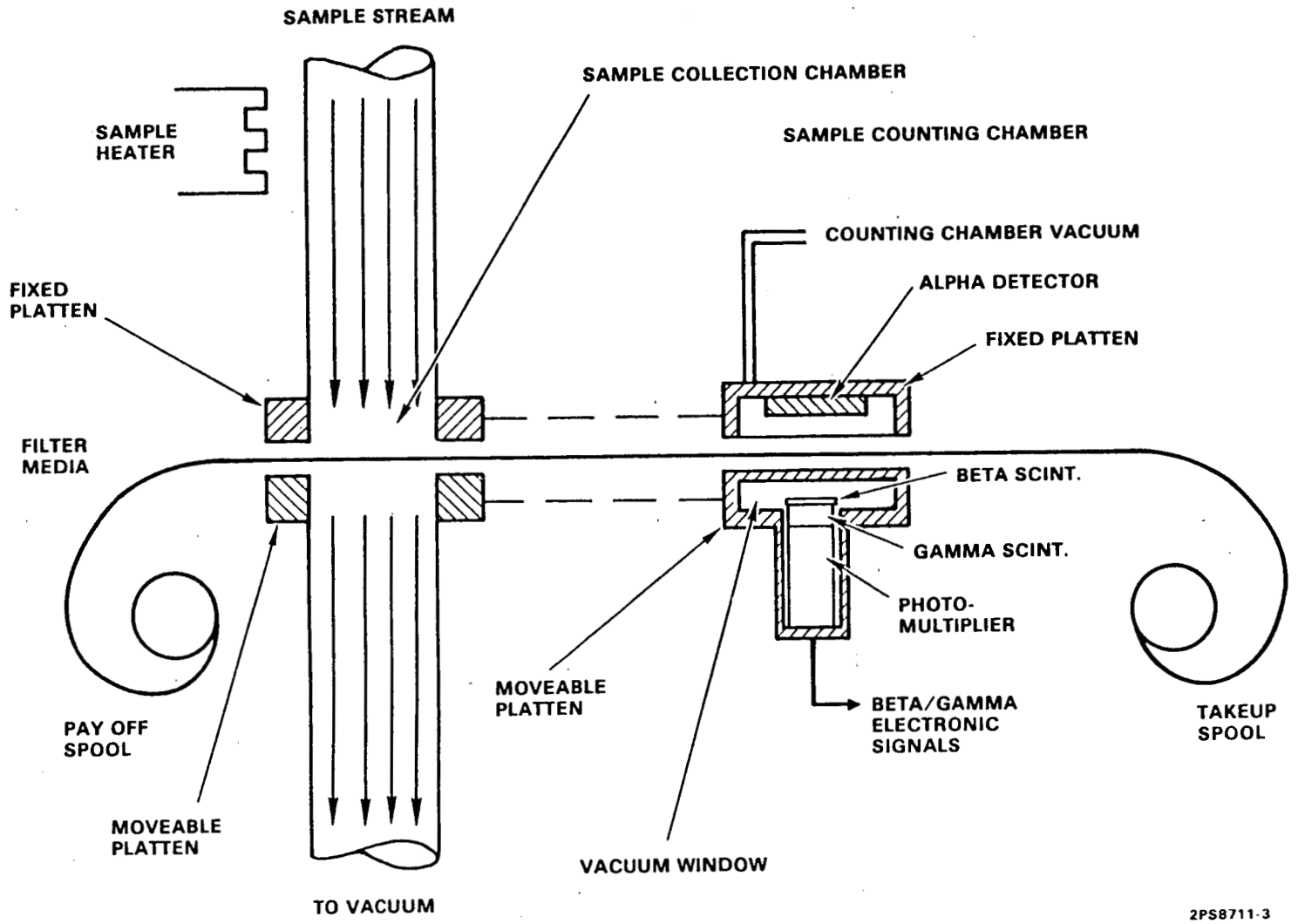

Figure 2. Moving Filter Assembly Configuration.

ALPHA SPECTRA PROCESSING

An alpha energy spectral peak is characterized by a statistical distribution with a low-energy tail. The general peak shape is comparable to that found for gamma energy analysis (GEA). The principal differences are that resolution and peak tailing are directly dependent on the sample mass. Experimentation has shown that a common GEA shape model (Routti 1969; Schick 1974) can be adapted for AEA use. The model is a modified Gaussian error function with a quadratic tailing factor folded into the low-energy side.

The variability of AEA spectra due to mass, low count rates (0.57 Bq ^{239}Pu in 240 min), and low relative resolution require the spectra be conditioned to improve the shape quality for peak search and processing. Beginning with the highest energy, a portion of each full peak, sufficient to describe the peak but limit adjacent contribution, is introduced to a simplex (Fiori and Myklebust 1979; Caceci and Cacheria 1984) fitting algorithm. After completion of the fit, the modeled peak is subtracted from the original spectrum. The process is reapplied until all peaks are analyzed. The ever present radon daughters, ^{212}Bi and ^{212}Po, are used to internally energy calibrate each spectrum so that the plutonium region of interest (ROI) is exactly known. Spectral parameters are used to test the integrity of the alpha analysis. Figure 3 shows the results of this computer processing for the various peaks of an MFRAM plutonium standard sample with a fresh accumulation of radon daughters.

A proper fit of the tailing factor for the ^{212}Bi ROI requires the inclusion of the multiple ^{212}Bi peaks (fig. 3). Additionally, a low broad peak, assumed to be detector coincidence summing, is fit between the ^{212}Bi and ^{212}Po peaks. These additions provide improvements for low-level ^{239}Pu estimation. The responses to ^{239}Pu standards are shown in figure 4. When no peak is detectable in the plutonium ROI, a procedure (Currie 1968) is used that fits residual data with other spectral peak parameters to estimate the maximum possible concentration of plutonium.

BETA ANALYSIS PROCESSING

The beta channel is a quasi-gross-counting analytical instrument. While offering some immunity to short-lived radon, it sums, proportional to efficiency, the contributions of the long-lived beta emitting nuclides. The efficiency value is that value determined for the most prevalent radionuclide in the stack effluent having the lowest efficiency due to a coincident gamma. Alarms are based on and corrected by the ratio of the nuclide having the lowest concentration guide and the highest efficiency. The composite results yield good worst-case accuracy for both alarms and output concentrations.

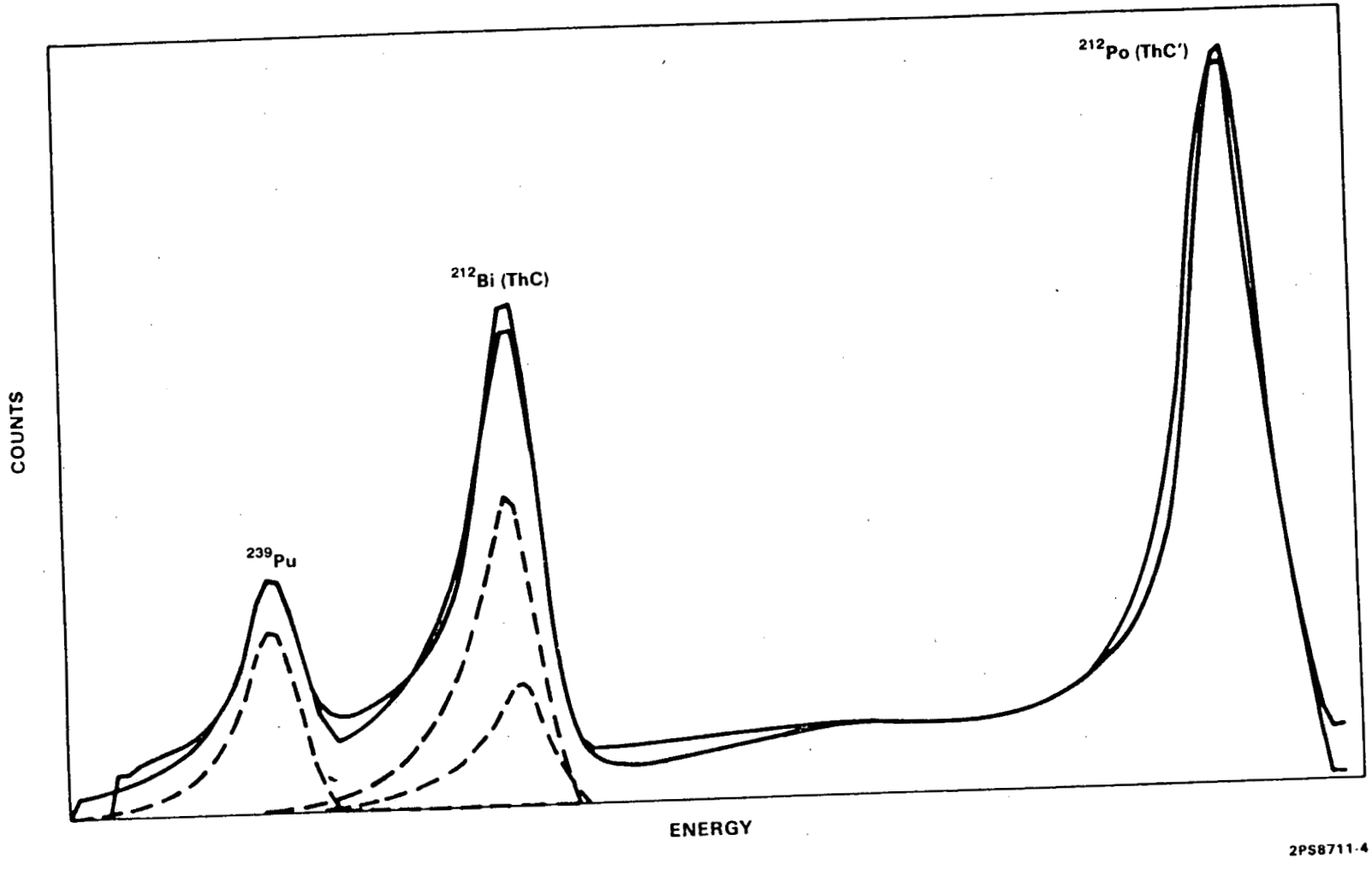

Figure 3. Moving Filter Radioactive Aerosol Monitor Alpha Energy Analysis Spectrum.

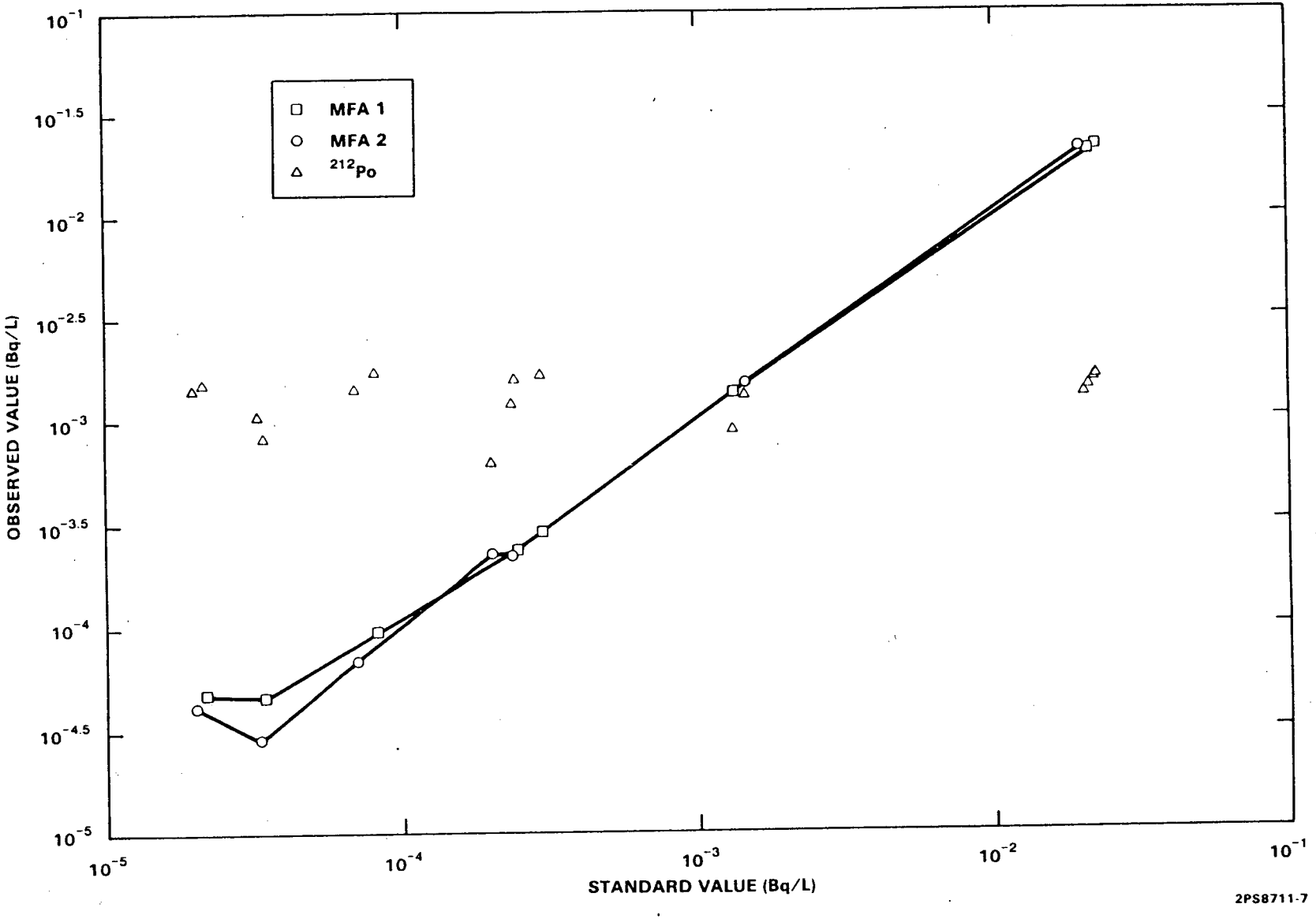

Figure 4. Moving Filter Radioactive Aerosol Monitor Plutonium Response.

The short-lived radon contribution to the beta analysis channel that escapes the anticoincidence gamma-ray analysis is compensated by the determination of a relationship between total beta and ^{212}Po concentrations. Figure 5 shows results from ^{90}Sr standards measurements in the presence of the radon progeny beta component. Using a linear regression, the beta and ^{212}Po data are reduced to a set of correction constants. The effect of this correction is shown in figure 6 in which the detection limit for ^{90}Sr is significantly reduced.

RESULTS

Standards used for primary calibration testing have demonstrated that plutonium air concentrations of 3.7×10^{-5} Bq/L can be measured with an error of less than ±20% from a 4-h collection period (near real-time) in the presence of particulate overburden and high concentrations of radon progeny. The detection limit for plutonium based on the mathematical modeling of the ^{212}Bi tail is calculated to be 3.7×10^{-6} Bq/L. This value was determined from samples with an average ^{212}Bi concentration of 2.6×10^{-3} Bq/L. The beta channel detection capability was determined by the foregoing testing matrix to be 3.7×10^{-4} Bq/L ±20% with a typical gross beta concentration of 7.4×10^{-3} Bq/L.

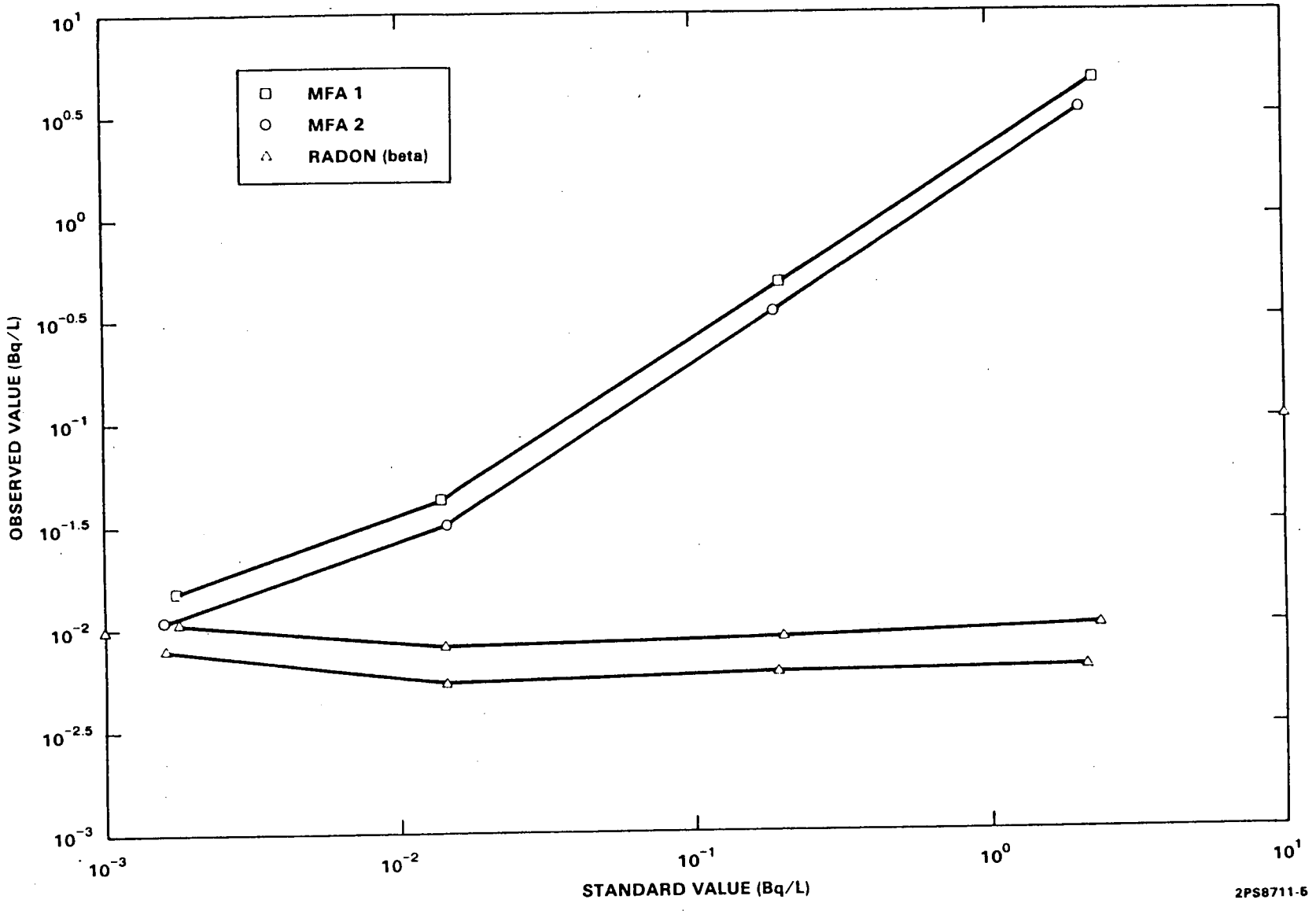

Figure 5. Strontium-90 Uncorrected for Radon.

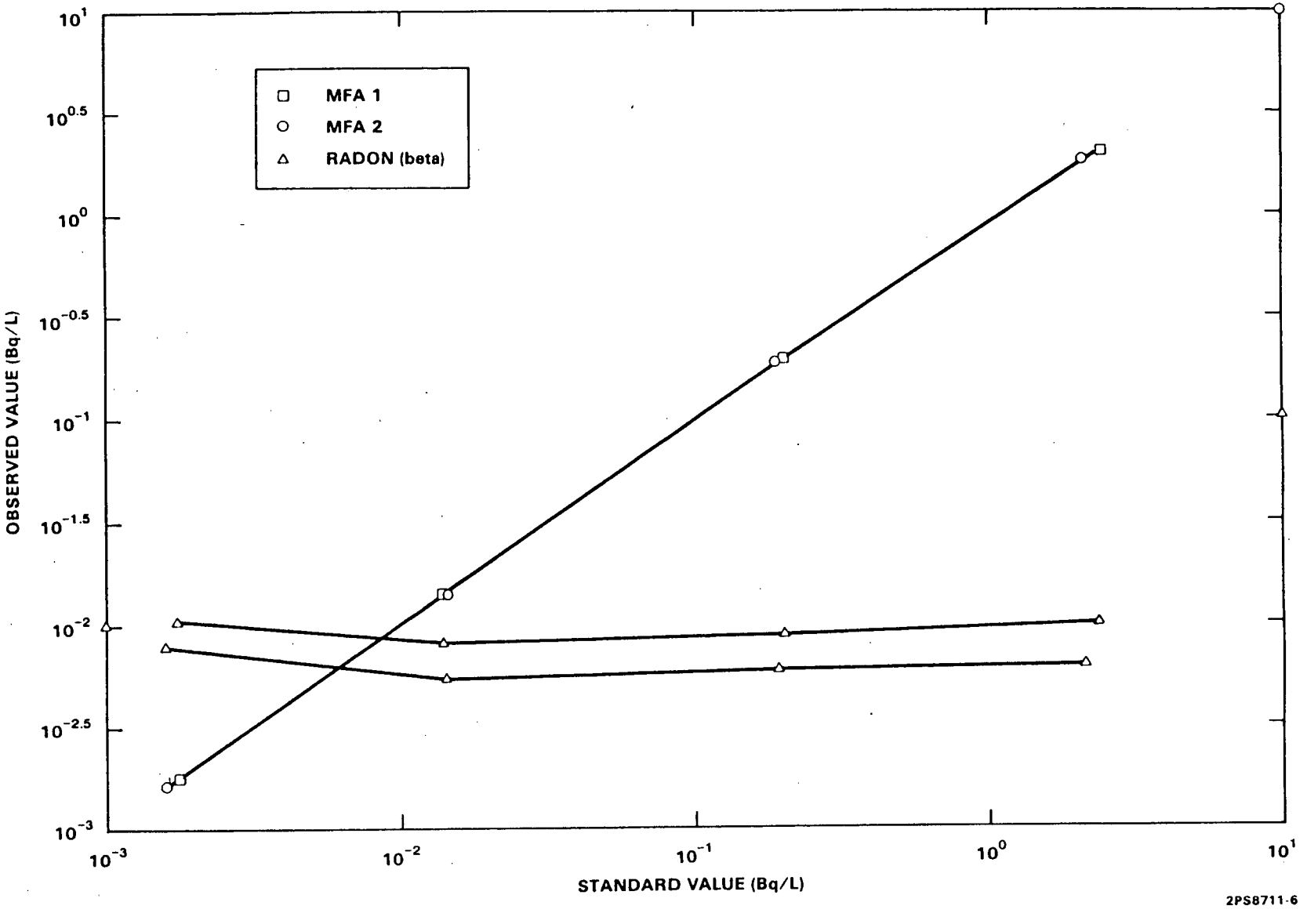

Figure 6. Strontium-90 Corrected for Radon.

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