Determination of Gross Alpha Radioactivity in Sewage Sludge Samples

Abdul Khalique, Bernard Sawyer, David R. Zenz,
and Cecil Lue-Hing
Metropolitan Water Reclamation District of Greater Chicago
100 East Erie Street
Chicago, IL 60611

INTRODUCTION

The Metropolitan Water Reclamation District of Greater Chicago (District) is located within the boundaries of Cook County Illinois and serves an area of 872 square miles. The District treats, on the average, 1500 million gallons per day of wastewater. This wastewater flow is treated at the District's seven water reclamation plants (WRPs).

As part of its regular monitoring program, the District's radiochemistry laboratory routinely analyzes raw sewage, final effluent, and sludge samples for gross alpha and gross beta activity using a gas flow proportional counter. The Standard Methods for the Examination of Water and Wastewater (Standard Methods) (1) procedure is used to measure gross alpha activity in all of these samples.

Based upon routine monitoring data from the District's seven WRPs, it has been observed that the gross alpha activity of Lemont WRP sludge is from five to ten times higher than the gross alpha activity of sludges from the other six WRPs. This has been attributed to the fact that the groundwater in the Lemont area is known to contain relatively high concentrations of naturally occurring radium-226 (2, 3).

Recently, in the course of conducting a specialized sludge sampling and analysis program it was observed that the gross alpha activity of Lemont WRP sludge samples was increasing as a function of the elapsed time between ashing the sample and counting the sample in the gas flow proportional counter. For example, in one sludge sample from the Lemont WRP, gross alpha activity increased from 30.5 pCi/g dry weight when counted on the day of ashing to over 100 pCi/g when counted 31 days after ashing.

This report describes the results of an evaluation of the alternate analytical method used in an attempt to eliminate the problem of alpha activity increasing with holding time.

MATERIAL AND METHODS

Sewage Sludge Samples

The sewage sludge samples used in this study were collected from various stages of the wastewater and sludge treatment processes at the

different District WRPs, to represent a cross-section of sludge types. The sludge ranged from 1% total solids by weight to 60% total solids by weight, with at least one sample from each WRP.

Analytical Methodology

METHOD 1 (ALTERNATE PROCEDURE)

A thoroughly mixed sludge sample (25 to 50g) was transferred to a tared evaporating dish. The sludge sample was dried to constant weight at 103-105°C. The increase in weight over the empty dish represents the total solids. The dried residue was finely powdered in a grinder and sifted through a 200-mesh sieve. An accurately weighed portion (90 to 100 mg) of the fine powder was transferred to a tared stainless steel planchet. The powdered sample was uniformly distributed with a few drops of 0.5% w/v acrylic solution in acetone. The acetone was evaporated at room temperature. The residue was oven dried to constant weight at 103 to 105°C, and counted on a Tennelec LB5100 gas flow proportional counter. The counter was calibrated with americum-241.

METHOD 2 (STANDARD METHODS PROCEDURE)

The finely ground powdered sample from Method 1 was accurately weighed in a tared evaporating dish and ignited to a constant weight at 550°C. The residue in the dish represents the fixed solids. The fixed solids were reduced to fine powder with a pestle in the evaporating dish. An accurately weighed portion (90 to 100 mg) of the fixed solids was transferred to a tared stainless steel planchet. It was distributed to a uniform thickness by spreading with a few drops of 0.5% w/v acrylic solution in acetone. The acetone was evaporated at room temperature and the residue was oven dried to constant weight at 103 to 105°C. It was then counted on a Tennelec LB5100 gas flow proportional counter. The counter was calibrated with americum-241.

Calculations

METHOD 1

$$\text{Gross alpha activity (pCi/g dry sludge)} = \frac{\text{net alpha counts per minute}}{2.22 \times E \times WD}$$

Where E = counter efficiency
WD = weight of dried sample counted

METHOD 2

$$\text{Gross alpha activity (pCi/g dry sludge)} = \frac{\text{net alpha counts per minute}}{2.22 \times E \times WA} \times \frac{WF}{WT}$$

Where E = counter efficiency
WA = weight of ashed sample counted
WF = weight of fixed solids
WT = weight of total solids

ALPHA INGROWTH FACTOR

Alpha ingrowth factor = $1 + 3(1-e^{-\lambda T})$

Where $\lambda = \frac{0.693}{\text{half-life of radon-222}} = \frac{0.693}{3.824 \text{ days}}$

T = elapsed time from the time of ashing the sample to the midpoint of the counting period.

INGROWTH CORRECTED ALPHA ACTIVITY

Ingrowth corrected alpha activity = $\frac{\text{gross alpha activity}}{\text{alpha ingrowth factor}}$

RESULTS AND DISCUSSION

Alternate Procedure

The Standard Methods procedure for analysis of gross alpha activity in sludge samples specifies drying the sample at 103 to 105°C, then ashing the sample at 550°C. The ashing removes organic matter, as organic matter can sometimes interfere with alpha particle counting in the gas flow proportional counter.

As an experiment, it was decided to determine exactly what would happen to the gross alpha activity of Lemont WRP, and other District sludges, if samples were held for various lengths of time between drying and counting using Method 1, versus ashing and counting using the Standard Methods procedure. The results are presented in Table 1. For the sludge samples from WRPs other than the Lemont WRP, there was close agreement in the gross alpha activities determined using either Method 1 or Method 2. In addition, for these samples, the gross alpha activity did not change appreciably with time between drying/ashing and counting.

For the Lemont WRP sample, a large increase in gross alpha activity with time between ashing/drying and counting was observed using Method 2 (41.6 pCi/g increasing to 103.2 pCi/g after 40 days) versus Method 1 (25.6 pCi/g increasing to 38.2 pCi/g after 40 days).

Alpha Ingrowth Factor

As mentioned previously, Lemont WRP sludge is known to contain radium-226. The United States Environmental Protection Agency (USEPA) has published analytical procedures for the measurement of radioactivity in

Table 1. Comparison of Gross Alpha Acitivty (pCi/g) in Sewage Sludge Samples Using Method and Method 2

Sludge Type	Method*	Day 1	Day 2	Day 4	Day 9	Day 13	Day 16	Day 20	Day 25	Day 30	Day 40
		(Gross Alpha Activity, pCi/g dry weight)									
Lemont WRP	1	25.6	29.0	35.3	34.5	33.2	33.6	40.2	33.9	34.3	38.2
Thickened	2	41.6	53.3	63.9	84.2	88.2	93.5	98.5	96.5	99.5	103.2
Calumet WRP	1	NA**	4.0	4.5	6.1	4.6	4.5	4.9	5.8	4.6	5.3
Centirfuge Cake	2	4.5	5.1	5.9	5.9	5.7	6.3	6.2	6.4	6.7	6.9
Stickney WRP	1	NA	4.4	5.2	4.8	5.0	5.0	4.8	4.6	4.8	6.2
Centrifuge Cake	2	5.4	6.0	6.1	5.6	7.2	6.9	7.6	6.4	7.5	6.0
Stickney WRP	1	NA	3.7	3.7	2.7	4.5	2.7	2.9	3.3	2.8	2.8
Air-dried	2	5.0	5.8	6.1	6.2	6.1	6.0	6.4	6.1	5.6	6.1
Calumet WRP	1	NA	9.6	7.2	9.2	8.5	9.2	7.9	7.2	7.6	8.4
Air-dried	2	6.0	5.0	5.4	6.7	6.5	5.9	5.6	5.6	5.3	6.5
Stickney WRP	1	NA	NA	6.9	6.9	7.3	7.8	7.8	7.2	8.2	5.3
Lagoon	2	5.7	NA	7.6	7.1	6.2	6.9	6.9	7.0	7.2	NA
Calumet WRP	1	5.7	6.7	6.9	6.2	5.2	5.9	7.0	7.5	7.4	NA
Lagoon	2	4.9	6.5	7.4	5.3	7.5	5.9	6.9	7.4	NA	NA
Kirie WRP	1	NA	4.2	4.8	5.5	3.4	2.5	3.3	2.9	4.3	NA
Thickened	2	NA	5.6	5.3	7.7	6.4	6.2	6.6	6.5	7.3	6.8
Egan WRP	1	NA	5.1	5.2	4.2	3.5	5.1	4.3	3.6	3.8	NA
Digested	2	NA	5.5	7.3	7.2	7.4	8.4	7.0	6.2	7.5	7.9
Hanover Park	1	NA	3.1	4.0	3.6	4.0	4.3	2.9	3.0	3.0	NA
WRP Digested	2	NA	5.0	5.4	6.7	5.9	6.8	6.1	6.4	6.0	6.7
North Side WRP	1	NA	NA	5.2	3.8	3.8	4.4	5.5	3.4	4.1	3.3
Thickened	2	4.1	4.4	4.5	4.2	5.1	4.4	4.7	4.3	NA	NA

*Method 1 - drying at 103 to 105°C only.
Method 2 - drying and ashing.
**NA = no analysis.

drinking water (4). The procedure for gross radium analysis states that "...the sample should be counted for alpha activity within one hour of preparation to minimize the effect of the ingrowing radium-226 daughters." Radium-226 decays to radon-222 by emitting alpha particles. The radon-222 subsequently continues to decay to various progeny (daughter compounds) by emitting additional alpha particles. As described in the USEPA radium procedure, if the samples are not counted within one hour, an alpha ingrowth factor must be calculated to correct the measured alpha count for the build-up of radon-222 and its progency in the sample matrix.

The Standard Methods procedure for gross alpha activity does not call for the use of an Alpha Ingrowth Factor in calculating a final gross alpha result. However, if a correction for alpha ingrowth is made on the Lemont WRP sludge gross alpha data presented in Table 1, an interesting result is obtained (Table 2). As can be seen, the corrected gross alpha activities for the Standard Methods procedure do not increase with time. Thus, it can be concluded that the observed increase in gross alpha activity with time

Table 2. Gross Alpha Activity of Lemont WRP Sludge

Time From Sample Ashing/Drying to Counting (Days)	Gross Alpha Activity (pCi/g dry weight)		
	Standard Methods Procedure*	Standard Methods Corrected for Alpha Ingrowth	Method 1*
1	41.6	27.8	25.6
2	53.3	27.8	29.0
4	63.9	25.1	35.3
9	84.2	24.7	34.5
13	88.2	24.1	33.2
16	93.5	24.6	33.6
20	98.5	25.2	40.2
25	96.5	24.4	33.9
30	99.5	24.9	34.3
40	103.2	25.8	38.2

*Same data as shown in Table 1.

between ashing/drying and counting in the Lemont WRP sludge samples is caused by ingrowth of alpha particles in the ashed sludge sample. This alpha ingrowth does not manifest itself to the same extent when the sludge sample is analyzed using the alternate sample preparation procedure (Method 1). The reason alpha ingrowth is not observed using Method 1, but is observed using the Standard Methods procedure, has not been firmly established. However, it appears that ashing the sample causes a change in the physical structure of the sample which traps the radon-222 gas in the sample matrix, whereas radon is driven off when the sample is dried using Method 1.

CONCLUSIONS

This study of two gross alpha activity analytical techniques for sewage sludge samples indicates the following:

For sewage sludge samples not containing significant amounts of radium-226, the gross alpha activity results were similar using either method, and the activity did not vary with holding time between ashing and counting. For the sewage sludge sample known to contain appreciable quantities of radium-226, gross alpha activity increased with holding time using the Standard Methods procedure, but increased only slightly using the alternate procedure. When gross alpha activity using the Standard Methods procedure was corrected using an Alpha Ingrowth Factor to take into account the ingrowth of radon-222 and its progeny from radium-226 decay, the results for both procedures were comparable, verifying that alpha ingrowth is responsible for the apparent increase in gross alpha activity with time.

The experimental data indicates that gross alpha activity in sewage sludge can be accurately determined after drying the sample at 103 to 105°C as opposed to ashing the sample at 550°C. The study also indicates that sludges suspected of containing radium-226 should be counted immediately after ashing, if the Standard Methods procedure is used, or an alpha ingrowth correction factor should be applied. Otherwise, incorrectly high gross alpha results will be obtained.

REFERENCES

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