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Underestimation of Activity from DU Oxide Measurements Due to Self/Media Absorption & Attenuation

Comparative Measurements for Electroplated Source Versus Field Samples
AMUG Workshop,
October, 2016

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Purpose

- Over-arching Purpose:

  Evaluate the significance of self-absorption & attenuation on the quantification of alpha activity for heavy metal particulate contamination.

- Focused Purpose:

  Consider the affect of self-absorption & attenuation, overall, within the particular context of quantifying particulate contamination at variable source to detector distances.

HP Relevance

Radiological Surveillance Relative to Surface Contamination Issues:

- Identify the need for posting
- Assess the need for and level of monitoring
- Control material and equipment
- Control areas

Airborne Radioactivity Monitoring Data Analysis Relevance

- Determination of DAC for radiological controls
- Internal dose estimates

Equipment/material Clearance and D&D

- Per DOE O458.1

Recognition of Alpha Self/Media Absorption & Attenuation Issues

Fuel manufacturing facility

- Waste water and sewer samples
- Various air sampling considerations

SNL RMWMF contamination control/air monitoring

- LSC results on known DU contaminated surfaces/subsequent investigation

Earlier DU Related Studies

- Results of comparative analysis of DU samples using field and laboratory instrumentation

Relevant General Facts

- Depleted uranium refers to uranium with lower than natural isotopic ratios of ^{235}U
- DU, as a metal/oxide, is very dense (19/11 g/cm^3 respectively)
- Greater the depletion, the greater the alpha activity is due to ^{238}U.
- Beta/alpha ratios for DU *typically* run between 1.2 and 1.7
- The primary beta emitters are ^{234}Th (low E) and ^{234}Pa (high E)

Study Materials

- 10 Swipes of DU oxide were used for this study (~ 2K-55K dpm).
- An electroplated DU source (~1100 dpm) was used as a "control".
- All samples and the DU source were counted using a RadEye SX with a SHP380AB probe.
- All analyses were carried out on a jig and spacers to maintain uniformity of measurements
- Spacers used to yield set source to detector distances.

Jig Assembly

Methodology

- The electroplated source and each swipe counted 3 times, 5 minutes each, in each study position.
- A thin spacer was used for “nominal” contact to avoid actual physical contact with source or samples.
- Additional spacers used to achieve ~ 3.3, 6.6 and 9.9 mm source to detector distances for sample analysis.
- Backgrounds collected before and after sample analysis.

Data Analysis Basis

- Instrument counting efficiencies were determined by our instrumentation group, specific to the probe center position.

  alpha: 0.178 c/d   beta (^{36}Cl): 0.187 c/d

- All swipes quantified in advance, using gamma spectroscopy, beta activity based on known parent-daughter stoichiometric relationships.
- Electroplated source is NIST traceable, activity as per manufacturer.
- Data for each sample in each position was averaged and converted to dpm.

Note: From an earlier study, a swipe from this DU material was evaluated for particle size, yielding a bi-modal distribution with most of the mass between 1-5 microns, and between 0.1 and 0.3 microns.

True Alpha Counting Efficiency & Activity Correction Factor

True Count Eff.	Contact %	@ 3.3 mm %	@ 6.6 mm %	@ 9.9 mm %	Correct. Factor Contact	Correct. Factor @ 3.3 mm	Correct. Factor @ 6.6 mm	Correct. Factor @ 9.9 mm
RS06023	**14.3**	**11.5**	**7.76**	**2.48**	**1.2**	**1.6**	**2.3**	**7.2**
Samples Mean	**2.54**	**1.35**	**0.76**	**0.2**	**8.2**	**15.5**	**27.8**	**108**
Samples Min.	**1.22**	**0.66**	**0.35**	**0.08**	**4.4**	**8.5**	**13.1**	**43.4**
Samples Max.	**4.06**	**2.10**	**1.36**	**0.41**	**14.6**	**26.9**	**51.5**	**213**

Comparative Measurements
True Beta Ct. Efficiency & Activity C. Factor

True Count Eff.	Contact %	@ 3.3 mm %	@ 6.6 mm %	@ 9.9 mm %	Correct. Factor	Contact	@ 3.3 mm	@ 6.6 mm	@ 9.9 mm
RS06023	**19.0**	**18.2**	**18.1**	**15.7**		**1.0**	**1.0**	**1.0**	**1.2**
Samples Mean	**12.5**	**12.1**	**11.6**	**10.6**		**1.5**	**1.5**	**1.6**	**1.8**
Samples Min.	**9.32**	**9.04**	**8.90**	**7.83**		**1.2**	**1.2**	**1.2**	**1.3**
Samples Max.	**14.7**	**14.3**	**13.9**	**12.9**		**2.0**	**2.1**	**2.1**	**2.4**

Comparative Measurements Beta/Alpha Ratios

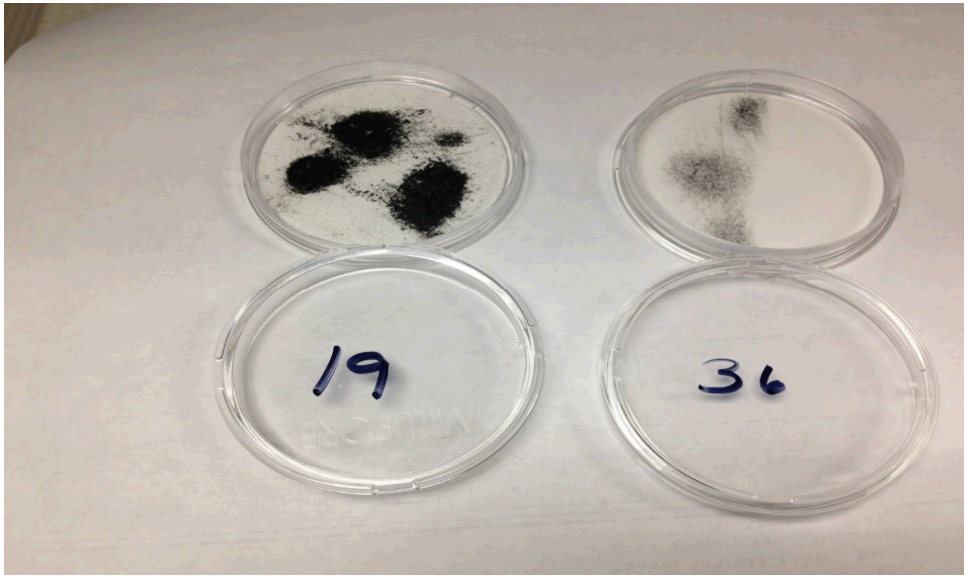
Sample	Contact	@ 3.3 mm	@ 6.6 mm	@ 9.9 mm
RS6023	2.2	2.7	3.9	10.6
Samples Mean	9.0	16.5	28.4	102
Samples Min.	4.2	7.5	12.0	36.7
Samples Max.	17.9	33.6	56.6	215

Comparative Measurements DU Alpha

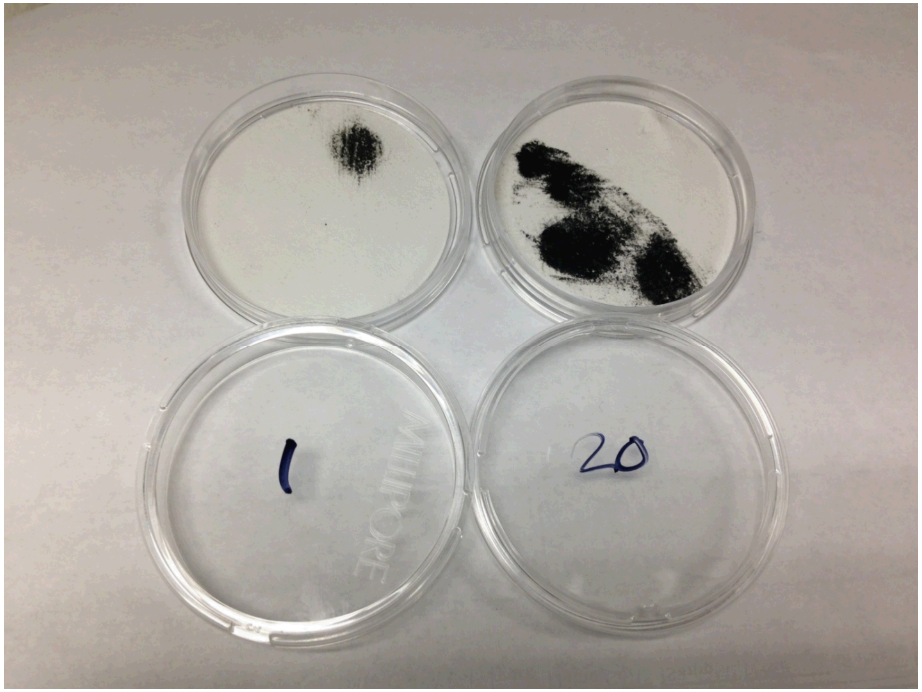
Sample	Contact dpm	@ 3.3 mm dpm	@ 6.6 mm dpm	@ 9.9 mm dpm	^{tot}U (ɣ-spec) dpm
RS06023	910	732	496	158	1136
4	439	245	127	39	2906
7	378	212	125	34	2428
14	1252	692	362	94	7544
15	1700	875	502	135	21861
19	3800	2063	1076	261	55459
20	3277	1699	983	240	42826
26	2044	980	565	122	13260
29	1859	978	536	143	11289
33	483	249	161	49	2114
37	569	310	165	43	3055

Build-up Variability

- For #19: $\alpha_\gamma/\alpha_g = 14.6$ β/α ratio = 16.1
- For #36: $\alpha_\gamma/\alpha_g = 4.2$ β/α ratio = 3.8



Variability in Swipe Collection Area



Results Summary

- For this study beta/alpha ratios averaged close to 10 on contact and over 100 at 1 cm, significantly higher than the expected 1.2-1.7 for DU. Elevated beta/alpha ratios are a good indicator of potential absorption & attenuation issues for DU
- Based on counting data, true counting efficiencies for alpha particulate drop in a non-linear manner with increasing detector-source distance, resulting in order of magnitude correction factors (averaging 10 for contact, > 100 @ 1 cm).

Results Summary (Cont.)

- Based on the significant reduction of true counting efficiencies, correction factors on nominal source/detector contact exceeded a factor of 10 in some cases, averaging > 8. This strongly suggests that absorption within dense particulate and through a layer of contamination can lead to significant reduction in true counting efficiencies.
- For DU, there appears to be some level of absorption/attenuation effects that occur for beta, although to a much lesser degree than alpha. It is suggested that this is due mainly to the effect on the relatively low energy spectrum of the ^{234}Th betas. This is not expected to exceed a factor of 2 for contact readings .

Results Summary (Cont.)

- The affect of self/media absorption and attenuation was greatest for swipes with the greatest build-up of contamination (based on visual inspection). The correction factor increased 6 fold between contact and 1 cm for the DU Source, between 10 and 17 for swipes.
- While this study focused on these phenomenon as related to measurements with a specific instrument/probe, other studies clearly indicate that other gross activity counting field instruments as well as laboratory instruments significantly under-quantify true activity for dense alpha emitting particulate.

Final Conclusion

This study has shown significant differences in the parametric values obtained using a laboratory source (basically "ideal" world) compared to actual field samples of contamination (representing "real" world). Conventional wisdom regarding the significance of self/media absorption and attenuation for quantification of alpha emitting nuclides has often been molded based on experience with laboratory sources.

Final Conclusion

Failure to account for these phenomena may lead to gross underestimation of alpha activity and negatively impact HP decisions. These include clearing items, equipment as well as facilities from all regulatory controls, determining the types and levels of controls needed in operational work areas regarding surface contamination issues and identification of air monitoring issues.

Supplement for Data Summary Details

Comparative Measurements DU Beta

Sample	Contact dpm	@ 3.3 mm dpm	@ 6.6 mm dpm	@ 9.9 mm dpm	Stoich. beta dpm
RS6023	2029	1942	1937	1683	1994
4	2688	2706	2563	2363	4606
7	2517	2517	2388	2230	3853
14	8954	8518	8177	7406	11971
15	22911	21912	21021	19559	34691
19	61264	59375	57449	53171	88006
20	58653	56986	55633	51515	67960
26	12679	11930	11395	10183	21042
29	14098	13635	13100	11745	17914
33	2158	2121	1925	1787	3355
37	2416	2343	2306	2030	4848

True Alpha Counting Efficiency & Activity Correction Factor

Sample True Count Eff.	Contact %	@ 3.3 mm %	@ 6.6 mm %	@ 9.9 mm %	Correct. Factor	Contact	@ 3.3 mm	@ 6.6 mm	@ 9.9 mm
RS06023	14.3	11.5	7.76	2.48		1.2	1.6	2.3	7.2
4	2.69	1.50	0.78	0.24		6.6	11.8	22.9	75.3
7	2.77	1.55	0.92	0.25		6.4	11.5	19.4	72.1
14	2.95	1.63	0.86	0.22		6.0	10.9	20.8	80.0
15	1.38	0.71	0.41	0.11		12.9	25.0	43.5	161.7
19	1.22	0.66	0.35	0.08		14.6	26.9	51.5	212.8
20	1.36	0.71	0.41	0.10		13.1	25.2	43.6	178.7
26	2.74	1.32	0.76	0.16		6.5	13.5	23.5	108.9
29	2.93	1.54	0.85	0.23		6.1	11.5	21.0	78.9
33	4.06	2.10	1.36	0.41		4.4	8.5	13.1	43.4
37	3.31	1.81	0.96	0.25		5.4	9.8	18.5	71.0

True Beta Counting Efficiency & Activity Correction Factor

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Sample True Count Eff.	Contact %	@ 3.3 mm %	@ 6.6 mm %	@ 9.9 mm %	Correct. Factor	Contact	@ 3.3 mm	@ 6.6 mm	@ 9.9 mm
RS06023	19.0	18.2	18.1	15.7		1.0	1.0	1.0	1.2
4	10.9	11.0	10.4	9.59		1.7	1.7	1.8	1.9
7	12.2	12.2	11.6	10.8		1.5	1.5	1.6	1.7
14	14.0	13.3	12.8	11.6		1.3	1.4	1.5	1.6
15	12.4	11.8	11.3	10.5		1.5	1.6	1.7	1.8
19	13.0	12.6	12.2	11.3		1.4	1.5	1.5	1.7
20	14.7	14.2	13.9	12.9		1.2	1.2	1.2	1.3
26	11.3	10.6	10.1	9.05		1.7	1.8	1.8	2.1
29	14.7	14.2	13.7	12.3		1.3	1.3	1.4	1.5
33	12.0	11.8	10.7	9.96		1.6	1.6	1.7	1.9
37	9.32	9.04	8.90	7.83		2.0	2.1	2.1	2.4

Comparative Measurements Beta/Alpha Ratios

Sample	Contact	@ 3.3 mm	@ 6.6 mm	@ 9.9 mm
RS6023	2.2	2.7	3.9	10.6
4	6.1	11.0	20.2	61.3
7	6.7	11.9	19.0	66.2
14	7.2	12.3	22.6	78.6
15	13.5	25.0	41.9	145
19	16.1	28.8	53.4	204
20	17.9	33.5	56.6	215
26	6.2	12.2	20.2	83.6
29	7.6	13.9	24.4	82.1
33	4.5	8.5	12.0	36.7
37	4.2	7.5	14.0	47.2