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Department of Energy

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NOV 20 1997

DOE-0141-98

Mr. Tom Schneider, Project Manager
Ohio Environmental Protection Agency
401 East 5th Street
Dayton, Ohio 45402-2911

Dear Mr. Schneider:

MEASUREMENT AND CALCULATION OF THORIUM-232

In your October 17, 1997, letter commenting on the issues surrounding the measurement and calculation of thorium-232, the Ohio Environmental Protection Agency (OEPA) offered a proposal with respect to determining the concentrations of thorium-232, radium-228, and thorium-228. The Department of Energy (DOE) accepts the central tenet of that proposal: That only gamma emitting radionuclides below (in the decay chain) the analyte of interest and in secular equilibrium with the analyte of interest should be used in the activity concentration calculations. The gamma emitting radionuclides and the gamma photon energies to be used in the computations are shown in the table below:

Isotope to be Calculated	Isotope of Emission	Energy of Emitted Gamma Photon (keV)
Thorium-232	Actinium-228	911.1
	Actinium-228	969.1
	Thallium-208	583.1
	Lead-212	238.6
Radium-228	Actinium-228	911.1
	Actinium-228	969.1
	Thallium-208	583.1
	Lead-212	238.6
Thorium-228	Thallium-208	583.1
	Lead-212	238.6

Recycled and Recyclable

Other gamma photons from other thorium daughter isotopes could have been included as a basis of calculation. Reasons for omitting those gamma photons also are discussed below:

- Bismuth-212 (727.2 keV). Even though the Fernald Environmental Management Project (FEMP) had originally recommended using this gamma photon, a review of a considerable amount of data resulted in the decision to omit it from the above table. The low abundance of 727.2 keV photons emitted by bismuth-212 often results in very few, or no, photons detected and a corresponding large relative measurement error.

- Thallium-208 (511 keV). This gamma photon was proposed by the OEPA in the above referenced letter from Schneider to Reising. However, FEMP radiochemists and radiochemistry consultants to the FEMP unanimously agreed that the 511 keV energy is prone to significant interference from photons originating from positron annihilation. Thus, even in the absence of any of the above isotopes of interest, gamma spectra most likely will contain prominent 511 keV energy peaks.

- Thallium-208 (2.615 MeV). This gamma photon was also suggested by the OEPA. The FEMP felt that many of its contract laboratories might not have standards for calibrating to that high an energy and/or their calibration software might not extend to such high energies.

- Lead-212 (238.6 keV). The OEPA was concerned about using this photon due to potential interferences from radium-224 (241 MeV) and lead-214 (242 MeV). High resolution germanium detectors are easily able to resolve peaks differing by as little as 2 keV. Such resolution, combined with the peak quantification/deconvolution software available with modern gamma spectrometry systems, results in a very low likelihood of interferences for lead-212. A review of a number of recently acquired gamma spectra from the FEMP laboratories indicated that radium-224 and/or lead-214 could easily be differentiated from lead-212. Therefore, the FEMP believes this isotope provides valid information pertaining to the abundance of lead-212, in that this peak gives results which are consistent with those from other thorium daughters. However, even in the unlikely event that some interference counts were not subtracted or removed, then the result would be higher than the true value and would thereby provide a conservative concentration.

The OEPA concurred with the FEMP's proposal to use an error weighted average. A survey of radiochemistry labs under contract to the FEMP as well as discussions with other DOE laboratories indicated that a more standard approach is to use the inverse of the counting error squared rather than the inverse of the counting error as a weighting factor. The equation below is therefore written using the inverse of the counting error squared.

In the equation below X_1 is the activity concentration of an analyte determined by using the lead-212 gamma photon; X_2 is the activity concentration of an analyte determined by using the thallium-208 gamma photon; X_3 is the activity concentration of an analyte determined

by using the actinium-228 gamma photon (911.1 keV); and X_4 is the activity concentration of an analyte determined by using the actinium-228 gamma photon (969.1 keV). CE_1, CE_2, ... CE_4 are the one sigma counting errors associated with those calculated concentrations.

The error weighted average concentration is then computed as follows:

$$ERROR\ WEIGHTED\ AVERAGE = \frac{X_1\left(\frac{1}{CE_1^2}\right) + X_2\left(\frac{1}{CE_2^2}\right) + X_3\left(\frac{1}{CE_3^2}\right) + X_4\left(\frac{1}{CE_4^2}\right)}{\frac{1}{CE_1^2} + \frac{1}{CE_2^2} + \frac{1}{CD_2^3} + \frac{1}{CE_4^2}}$$

If one or more gamma photopeaks are missing from a particular sample spectrum, the corresponding term is omitted from both the numerator and denominator of the above equation.

Table 1 shows the results of using the above methodology for A2P1 data. Thorium-228, thorium-232, and radium-228 are calculated using both the inverse of the counting error and the inverse of the counting error squared as weighting factors. The data show very little difference in the calculated results irrespective of which weighting factor is used. Further, the data show very little difference, particularly on an average basis, between activity concentrations of thorium-228 calculated using two gamma photons and activity concentrations of thorium-232 calculated using four gamma photons. Such equivalence is expected for isotopes in secular equilibrium.

If you or your staff should have any questions, please contact Robert Janke at (513) 648-3124.

Sincerely,

Johnny Reising

FEMP:R.J. Janke

Johnny W. Reising
Fernald Remedial Action
Project Manager

Enclosure: As Stated

cc w/enc:

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TABLE 1
Isotopic Thorium Summary Results for A2P1

FDF ID	LAB ID	CU	Th228 2 lines pCi/g*	Cnt Err 1 sig pCi/g*	Th232 4 lines pCi/g*	Cnt Err 1 sig pCi/g*	Ra228 4 lines pCi/g*	Cnt Err 1 sig pCi/g*	Th228 2 lines pCi/g**	Cnt Err 1 sig pCi/g**	Th232 4 lines pCi/g**	Cnt Err 1 sig pCi/g**	Ra228 4 lines pCi/g**	Cnt Err 1 sig pCi/g**
200304612	ONSITE	SF-B	0.76	0.02	0.77	0.02	0.77	0.02	0.77	0.02	0.77	0.02	0.77	0.02
200304613	ONSITE	SF-B	0.95	0.03	0.99	0.03	0.99	0.03	0.96	0.03	0.98	0.02	0.98	0.02
200304614	ONSITE	SF-B	0.51	0.02	0.54	0.02	0.54	0.02	0.50	0.01	0.51	0.01	0.51	0.01
200304615	ONSITE	SF-B	0.98	0.03	1.01	0.03	1.01	0.03	0.98	0.03	1.00	0.02	1.00	0.02
200304616	ONSITE	SF-B	0.66	0.02	0.68	0.02	0.68	0.02	0.65	0.02	0.67	0.02	0.67	0.02
200304617	ONSITE	SF-B	0.31	0.02	0.33	0.02	0.33	0.02	0.31	0.01	0.32	0.01	0.32	0.01
200304618	ONSITE	SF-B	0.32	0.02	0.32	0.02	0.32	0.02	0.31	0.01	0.31	0.01	0.31	0.01
200304619	ONSITE	SF-B	0.25	0.01	0.27	0.01	0.27	0.01	0.25	0.01	0.25	0.01	0.25	0.01
200304620	ONSITE	SF-B	0.22	0.01	0.24	0.01	0.24	0.01	0.22	0.01	0.23	0.01	0.23	0.01
200304621	ONSITE	SF-B	0.25	0.01	0.25	0.01	0.25	0.01	0.25	0.01	0.25	0.01	0.25	0.01
200304622	ONSITE	SF-B	0.84	0.02	0.87	0.02	0.87	0.02	0.83	0.02	0.85	0.02	0.85	0.02
200304623	ONSITE	SF-B	1.78	0.03	1.80	0.03	1.80	0.03	1.78	0.03	1.79	0.03	1.79	0.03
200304624	ONSITE	SF-B	0.65	0.02	0.68	0.02	0.68	0.02	0.63	0.02	0.65	0.02	0.65	0.02
200304625	ONSITE	SF-B	0.60	0.02	0.60	0.02	0.60	0.02	0.60	0.02	0.60	0.02	0.60	0.02
200304626	ONSITE	SF-B	1.12	0.03	1.13	0.03	1.13	0.03	1.09	0.03	1.10	0.02	1.10	0.02
200304627	ONSITE	SF-B	0.83	0.02	0.87	0.02	0.87	0.02	0.81	0.02	0.84	0.02	0.84	0.02
200304628	ONSITE	SF-B	0.96	0.03	1.00	0.03	1.00	0.03	0.96	0.02	0.97	0.02	0.97	0.02
200303738	ONSITE	IFP-B1	0.98	0.03	0.98	0.03	0.98	0.03	0.95	0.02	0.96	0.02	0.96	0.02
200303739	ONSITE	IFP-B1	0.75	0.03	0.76	0.03	0.76	0.03	0.73	0.02	0.74	0.02	0.74	0.02
200303740	ONSITE	IFP-B1	0.87	0.03	0.89	0.03	0.89	0.03	0.87	0.02	0.88	0.02	0.88	0.02
200303732	ONSITE	IFP-B1	1.06	0.03	1.11	0.03	1.11	0.03	1.07	0.03	1.09	0.02	1.09	0.02
200303733	ONSITE	IFP-B1	0.70	0.02	0.76	0.02	0.76	0.02	0.71	0.02	0.74	0.02	0.74	0.02
200303734	ONSITE	IFP-B1	1.15	0.03	1.09	0.03	1.09	0.03	1.12	0.03	1.10	0.03	1.10	0.03
200303735	ONSITE	IFP-B1	0.40	0.02	0.43	0.02	0.43	0.02	0.40	0.02	0.41	0.02	0.41	0.02
200303736	ONSITE	IFP-B1	0.91	0.03	0.92	0.02	0.92	0.02	0.90	0.02	0.91	0.02	0.91	0.02
200303737	ONSITE	IFP-B1	0.84	0.03	0.88	0.03	0.88	0.03	0.85	0.02	0.86	0.02	0.86	0.02
200303741	ONSITE	IFP-B1	1.08	0.03	1.06	0.03	1.06	0.03	1.05	0.03	1.05	0.02	1.05	0.02
200303742	ONSITE	IFP-B1	1.28	0.03	1.31	0.03	1.31	0.03	1.29	0.03	1.30	0.03	1.30	0.03
200303743	ONSITE	IFP-B1	0.76	0.02	0.78	0.02	0.78	0.02	0.73	0.02	0.75	0.02	0.75	0.02
200303727	ONSITE	IFP-B1	0.84	0.03	0.89	0.03	0.89	0.03	0.84	0.02	0.86	0.02	0.86	0.02
200303728	ONSITE	IFP-B1	0.95	0.03	0.96	0.03	0.96	0.03	0.94	0.03	0.95	0.03	0.95	0.03
200303729	ONSITE	IFP-B1	1.08	0.03	1.12	0.03	1.12	0.03	1.08	0.03	1.10	0.03	1.10	0.03
200303730	ONSITE	IFP-B1	0.86	0.02	0.86	0.02	0.86	0.02	0.86	0.02	0.86	0.02	0.86	0.02
200303731	ONSITE	IFP-B1	0.82	0.03	0.86	0.02	0.86	0.02	0.81	0.02	0.83	0.02	0.83	0.02
200304147	ONSITE	IFP-B2	0.99	0.03	0.98	0.03	0.98	0.03	1.00	0.02	0.99	0.02	0.99	0.02
200304148	ONSITE	IFP-B2	1.04	0.03	1.08	0.03	1.08	0.03	1.01	0.03	1.03	0.02	1.03	0.02
200304149	ONSITE	IFP-B2	1.03	0.03	1.02	0.03	1.02	0.03	1.04	0.03	1.04	0.03	1.04	0.03
200304150	ONSITE	IFP-B2	0.95	0.03	0.97	0.03	0.97	0.03	0.92	0.03	0.93	0.02	0.93	0.02
200304151	ONSITE	IFP-B2	0.96	0.03	0.95	0.03	0.95	0.03	0.96	0.02	0.96	0.02	0.96	0.02
200304152	ONSITE	IFP-B2	0.98	0.03	0.99	0.03	0.99	0.03	0.97	0.02	0.97	0.02	0.97	0.02
200304153	ONSITE	IFP-B2	0.95	0.03	0.97	0.03	0.97	0.03	0.94	0.02	0.96	0.02	0.96	0.02
200304154	ONSITE	IFP-B2	1.03	0.03	1.05	0.03	1.05	0.03	1.01	0.03	1.03	0.02	1.03	0.02
200304155	ONSITE	IFP-B2	1.01	0.03	1.07	0.03	1.07	0.03	1.00	0.03	1.03	0.02	1.03	0.02

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200304156	ONSITE	IFP-B2	1.05	0.03	1.06	0.03	1.06	0.03	1.04	0.03	1.05	0.02	1.05	0.02
200304157	ONSITE	IFP-B2	0.98	0.03	1.01	0.03	1.01	0.03	0.95	0.02	0.98	0.02	0.98	0.02
200304158	ONSITE	IFP-B2	0.96	0.03	0.98	0.03	0.98	0.03	0.96	0.03	0.97	0.02	0.97	0.02
200304159	ONSITE	IFP-B2	0.87	0.03	0.92	0.03	0.92	0.03	0.87	0.02	0.89	0.02	0.89	0.02
200304160	ONSITE	IFP-B2	0.88	0.03	0.92	0.03	0.92	0.03	0.86	0.02	0.89	0.02	0.89	0.02
200304558	ONSITE	AFP-B	0.67	0.02	0.64	0.02	0.64	0.02	0.67	0.02	0.65	0.02	0.65	0.02
200304559	ONSITE	AFP-B	0.72	0.02	0.72	0.02	0.72	0.02	0.70	0.02	0.71	0.02	0.71	0.02
200304560	ONSITE	AFP-B	0.67	0.02	0.66	0.02	0.66	0.02	0.66	0.02	0.66	0.02	0.66	0.02
200304561	ONSITE	AFP-B	0.84	0.02	0.87	0.02	0.87	0.02	0.83	0.02	0.84	0.02	0.84	0.02
200304562	ONSITE	AFP-B	0.36	0.02	0.35	0.02	0.35	0.02	0.35	0.02	0.35	0.01	0.35	0.01
200304563	ONSITE	AFP-B	0.88	0.02	0.93	0.02	0.93	0.02	0.87	0.02	0.90	0.02	0.90	0.02
200304564	ONSITE	AFP-B	0.56	0.02	0.60	0.02	0.60	0.02	0.57	0.02	0.59	0.02	0.59	0.02
200304565	ONSITE	AFP-B	1.79	0.05	1.77	0.04	1.77	0.04	1.76	0.04	1.75	0.04	1.75	0.04
200304566	ONSITE	AFP-B	0.79	0.02	0.84	0.02	0.84	0.02	0.77	0.02	0.80	0.02	0.80	0.02
200304567	ONSITE	AFP-B	0.82	0.03	0.79	0.02	0.79	0.02	0.79	0.02	0.78	0.02	0.78	0.02
200304568	ONSITE	AFP-B	0.49	0.02	0.48	0.02	0.48	0.02	0.49	0.02	0.49	0.02	0.49	0.02
200304569	ONSITE	AFP-B	0.50	0.02	0.48	0.02	0.48	0.02	0.50	0.02	0.49	0.02	0.49	0.02
200304570	ONSITE	AFP-B	1.05	0.03	1.09	0.03	1.09	0.03	1.05	0.03	1.06	0.03	1.06	0.03
200304571	ONSITE	AFP-B	1.04	0.03	1.01	0.03	1.01	0.03	1.02	0.03	1.01	0.03	1.01	0.03
200305472	ONSITE	AFP-B	1.07	0.04	1.03	0.03	1.03	0.03	1.06	0.03	1.04	0.03	1.04	0.03
200304573	ONSITE	AFP-B	1.06	0.03	1.05	0.03	1.05	0.03	1.01	0.03	1.02	0.03	1.02	0.03
200304574	ONSITE	AFP-B	0.96	0.03	0.94	0.03	0.94	0.03	0.95	0.03	0.94	0.03	0.94	0.03
Total	Avg		0.85		0.87		0.87		0.84		0.85		0.85	
	std dev		0.29		0.29		0.29		0.29		0.29		0.29	
SF-B	Avg		0.71		0.73		0.73		0.70		0.71		0.71	
	std dev		0.39		0.39		0.39		0.39		0.39		0.39	
IFP-B1	Avg		0.90		0.92		0.92		0.89		0.90		0.90	
	std dev		0.20		0.19		0.19		0.19		0.19		0.19	
IFP-B2	Avg		0.98		1.00		1.00		0.97		0.98		0.98	
	std dev		0.05		0.05		0.05		0.05		0.05		0.05	
AFP-B	Avg		0.84		0.84		0.84		0.83		0.83		0.83	
	std dev		0.32		0.32		0.32		0.31		0.31		0.31	

* Calculations use the inverse of the counting error as a weighting factor
** Calculations use the inverse of the counting error squared as a weighting factor.