DEPARTMENT OF THE ARMY
BUFFALO DISTRICT, CORPS OF ENGINEERS
1776 NIAGARA STREET
BUFFALO, NEW YORK 14207-3199

AUG 30 2010

REPLY TO
ATTENTION OF

Special Projects Branch

SUBJECT: NFSS Interim Waste Containment Structure

[redacted]

Dear [redacted]

Thank you for your continued interest in the Niagara Falls Storage Site (NFSS). The following are the Buffalo District's responses to concerns expressed in your letter of July 29, 2010.

a) Radioactive contamination in the NFSS water lines

Data from the NFSS Remedial Investigation (RI) indicates that radioactive contamination is currently migrating away from the interim waste containment structure (IWCS), along a disused water line. The Corps has yet to investigate the NFSS water lines, and therefore, has no idea how far radioactive contamination has spread. Has the contamination already moved off site?

U.S. Army Corps of Engineers (USACE) Response: It is correct that the Corps did not sample the former LOOW 42-inch water line or the 10-inch water line because both of these lines carried fresh water used as either drinking or process water. The original purpose of the 42-inch line was to draw water from the Niagara River to the freshwater treatment plant to support the manufacturing of trinitrotoluene (TNT), and the 10-inch water line transported potable water under pressure from the City of Niagara Falls to the site. Instead, the Corps focused sampling efforts on pipelines that carried wastewater and posed the greatest potential for contamination. The Corps investigated the former Lake Ontario Ordnance Works (LOOW) sanitary sewer and acid waste subsurface pipelines, which were designed to transport wastewater to the former LOOW Wastewater Treatment Plant.

It is also correct that Figures 5-1 to 5-4 in the NFSS Remedial Investigation Report (RIR) (December 2007) showed potential uranium plumes[1] extending from the southern end of the IWCS (from temporary well point (TWP 833)) along a water line to a manhole in the sanitary sewer line (sample location MH06) approximately 375 feet away. (Please note that the sanitary sewer was plugged where it leaves the site.) However, upon further consideration, the Corps determined that using data from samples collected from within the former sanitary sewer and acid waste pipelines to delineate potential groundwater contamination was not only overly conservative but also an inappropriate use of this data since this data represents contamination contained within the lines and not contamination in the groundwater. The revised uranium groundwater plumes will be presented in the RIR Addendum.

During the RI, temporary well point TWP823 was located at the northern property boundary of the NFSS, near the area where several of the underground utility lines (including the sanitary and water lines) exit the property. The well was screened in the upper water bearing zone and the analytical data indicated that no chemicals or radionuclides exceeded the background upper tolerance limits (UTLs), demonstrating that contaminants are not migrating off-site.

b) No monitoring of the water levels inside the IWCS

The performance of the IWCS (its ability to isolate the high level radioactive waste inside from the surrounding environment) was to be assessed by a performance monitoring program which would run for a minimum of 5 years. The program was based on measuring water levels inside the IWCS and would provide instantaneous data, 24 hours a day. Increasing water levels inside the IWCS would signify water infiltration, in other words, containment failure.

Data from the first year of the program shows increased water levels. Monitoring the water levels inside the IWCS provides an early warning system for IWCS failure. However, the Corps did not review this crucial data for the IWCS and did not undertake monitoring of the water levels inside the IWCS.

USACE Response: In May 1986, Bechtel National, Inc. (BNI) on behalf of the U.S. Department of Energy issued a "Report on the Performance Monitoring System for the Waste Containment at the Niagara Falls Storage Site" that outlined the program that would be used to monitor the integrity of the IWCS. The performance monitoring program would consist in part of the installation of vibrating wire pressure transducers (VWPTs) to measure the water levels inside the IWCS and pneumatic pressure transducers (PPT) to verify the output of the VWPTs. The report indicated that the operation of the VWPTs and PPTs were to "continue for a minimum of 5 years (through 1991) but may be maintained for a longer period depending upon the results

[1] A plume is defined as a line or column of water containing chemicals moving from the source to areas further away.

observed." A total of 13 VWPTs and three PPTs were installed. Once the program was established, the VWPT readings were transmitted automatically on a daily basis and the PPTs were read manually every month.

Between 1987 and 1992, BNI issued annual performance monitoring reports that presented and evaluated the data. Despite numerous operating problems (electrical malfunctions and lightning strikes), the reports generally concluded that the data, which showed seasonal variations in water levels inside the IWCS, indicated no weakness in the clay cap, cutoff wall, cutoff dike, or the gray clay unit and demonstrated a trend toward water-level equalization within the IWCS. The manual readings of the PPTs were discontinued in 1990 because BNI reported that they confirmed that the VWPTs were functioning properly and were no longer needed.

USACE has reviewed the VWPT and PPT data presented in the performance monitoring reports and can draw no conclusions regarding the water levels inside the IWCS based on the data presented. Overall, it appeared that the VWPTs never functioned properly since numerous significant elevation changes were noted in several of the transducers. In addition, the data reported for the PPTs does not appear to correlate well with the VWPT data, yet it was determined that the use of these instruments could be discontinued because the VWPTs were functioning properly.

BNI estimated the life span of the VWPT system to be 25 years, and that assumes a functioning, well maintained system. The VWPTs have remained idle for 15 years and the data over the first five years of operation indicated serious operational issues (e.g., two lightning strikes). Therefore, it would not be feasible to resurrect the VWPT monitoring system.

It remains the Corps' position that the IWCS is performing as designed and will do so through the CERCLA study period. In addition, the Corps does not support penetrating the functional cap to install equipment to measure water levels inside the IWCS. Mr. Paul Giardina, Chief of the Radiation & Indoor Air Branch, U.S. Environmental Protection Agency (EPA) Region 2, expressed the same opinion regarding the performance of the IWCS and maintaining the integrity of the cap at the public workshop held in Lewiston, N.Y. on June 23, 2010.

c) No monitoring of the groundwater below the IWCS

The Department of Energy (DOE) set up a comprehensive groundwater monitoring program for the IWCS to analyze the groundwater near the surface and the groundwater beneath the IWCS to detect contaminants migrating away from the IWCS. The groundwater monitoring program acts as a secondary, albeit delayed method, of detecting IWCS failure. Monitoring of the lower groundwater around the IWCS ceased in 1995 without satisfactory explanation,

following the detection of radioactive contamination in the lower groundwater. From 1997 until 2008, as part of the NFSS Environmental Surveillance Program, the Corps continued to sample and analyze only the upper groundwater around the IWCS.

During the NFSS RI, the Corps sampled and analyzed groundwater from all of the IWCS monitoring wells in the UWBZ, but did not sample three key IWCS monitoring wells in the LWBZ, including the well where DOE had previously detected radioactive contamination.

USACE Response: For six years, between 1986 and 1992, the DOE monitored groundwater in wells screened in both the upper and lower water bearing zones. In 1993, the DOE modified the sampling program by including only one well (OW-15A) screened in the lower water bearing zone. The DOE reported that the wells screened in the upper water bearing zone, which would provide the earliest indication of leakage from the IWCS, had shown no evidence of migration of contaminants from the IWCS. In 1994, the DOE sampled only wells screened in the upper water bearing zone but indicated that sporadic future groundwater sampling events would include wells in the lower zone.

Subsequent environmental monitoring performed by the DOE and later by the USACE continued to include only wells screened in the upper water bearing zone since upper water bearing zone wells would provide the earliest indication of a release from the IWCS. Subsequently, the USACE monitors many upper water bearing zone wells in and around the IWCS as part of the environmental surveillance program. The USACE also measures water levels on a quarterly basis in wells screened in both zones and has found that vertical gradients derived from heads in monitoring well pairs vary with seasonality. During the first and second quarterly measurements, flow from the upper zone to the lower zone is typically dominant; however, during the third and fourth quarters, the majority of elevations in the lower system are greater than those measured in the upper system. This seasonal variation in the direction and magnitude of vertical gradients affects vertical flow between water bearing zones and potential long-term transport of contaminants between water bearing zones, thereby maintaining the upper zone as the primary transport pathway at the NFSS. While groundwater flow is primarily horizontal in these upper and lower zones, the upward vertical gradients help impede the potential for downward migration of contaminants into the lower zone from possible contaminant sources in the upper zone.

Furthermore, DOE's 1994 report entitled "Failure Analysis Report for NFSS" stated that the migration of contaminants from the IWCS to the lower water bearing zone would require significant travel time because of the presence of the glacio-lucustrine clay unit, which limits contaminant transport through low permeability, the ability to absorb contaminants, and lack of appreciable vertical recharge. The report also states that if contamination of the lower water bearing zone did occur, horizontal transport

times would be relatively slow due to the lack of significant permeability and low groundwater gradients across the site.

Between 1999 and 2003, 39 wells screened in the lower water bearing zone were sampled as part of the Remedial Investigation, and 24 of those 39 wells are located around the IWCS (in Exposure Unit (EU) 10). Among these 24 lower water bearing zone wells, the analytical data indicated that only 4 wells exhibited concentrations of total or dissolved radionuclides greater their respective background UTLs. However, the nature and extent of the site-related compounds detected did not warrant the identification of a plume since the hits were isolated and not spatially adjacent.

The latest special sampling event at the site took place on June 24, 2010 following the earthquake on June 23, 2010. The Corps collected samples from five monitoring wells around the IWCS and three of these wells, which included well OW15A, are screened in the lower water bearing zone. The analytical data from this sampling event, presented on the attached table, showed that no isotopic uranium, radium, or thorium concentrations exceeded federal and state regulations/guidelines in any of the wells sampled. Specifically, well OW15A showed that both unfiltered and filtered radium-226 were not detected above laboratory detection limits. Comparing this result to unfiltered radium data for well OW15A collected between 1988 and 1992, which ranged from 0.4 pCi/L to 1 pCi/L, indicates that this latest result is in line with historical data. The 1993 sampling event reported an unfiltered radium-226 concentration of 5.28 pCi/L. When compared to previously collected data, this concentration represents a minor increase and in the context of more recent data, does not represent an increasing trend.

Given the concern that has been expressed regarding the groundwater surveillance program, the USACE is in the process of modifying the program to include several wells screened in the lower water bearing zone. The analytical parameter list is also under review. Although the revised groundwater monitoring program has not been finalized, it will likely include well OW15A, given the proximity of this well to the IWCS and community concerns regarding data collected from this well in 1992 and 1993. However, it is unlikely that gross beta will be included on the analyte list since elevated gross beta data was detected in background samples collected during the RI. Given the distance of these background sampling locations from the NFSS (approximately 3,000 feet upgradient of the NFSS), it would appear that the source(s) of the gross beta activity is naturally occurring. Also, gross beta analysis is primarily a screening tool, so this analysis likely will not be included in the revised groundwater surveillance program. Instead, the revised groundwater surveillance program will include specific beta emitters that are known or suspected to be in the IWCS.

(Also, see response to comment d.)

d) Gross beta contamination in the lower water bearing zone (LWBZ) groundwater around the IWCS.

The Corps identified gross beta contamination in the LWBZ groundwater around the IWCS during the RI, but gave no explanation of the cause of the contamination. DOE previously found elevated levels of gross beta in the LWBZ groundwater, investigated and eliminated radium, thorium, uranium and potassium-40 as being the cause.

Note that there are even higher detections of gross beta in some of the background (upgradient) wells, specifically MW-17 (236 pCi/L), SP-2M (497 pCi/L), and W-11 (2,340 pCi/L) (total beta) and PZ-8D (372 pCi/L) (dissolved beta). Contributions from uranium, radium, thorium, and potassium in those background wells also do not account for the elevated beta measurements. Although our data do not provide an explanation for these elevated gross beta measurements in groundwater, we note that previous investigators have also observed anomalously elevated gross beta activity in environmental groundwater samples (Welch, A.H., Szabo, Zoltan, Parkhurst, D.L., VanMetre, P.C., and Mullin, A.H., 1995, *Gross-beta activity in ground water: natural sources and artifacts of sampling and laboratory analysis*: Applied Geochemistry, v. 10, p. 491-503).

DOE did not analyze for other beta emitting radioisotopes which are likely to be present on the NFSS, such as strontium-90. Soils contaminated with nuclear reprocessing waste from the Knolls Atomic Power Laboratory (KAPL) have been placed inside the IWCS; the KAPL radioactive wastes contain cesium-137, strontium-90 and plutonium.

Please see the accompanying memorandum from SAIC to USACE, discussing quantities of cesium-137 (Cs-137) likely in the IWCS. As further discussed below, the relative quantity and mobility of Cs-137 in the IWCS (relative to other better characterized constituents of the IWCS such as radium and uranium) indicate that Cs-137 would not be the best constituent to use as an indicator of IWCS breach.

Has the Corps analyzed the contaminated LWBZ groundwater for strontium-90, a beta emitting contaminant contained in KAPL waste? Strontium-90 may be a useful tracer for IWCS leakage.

The USACE has not analyzed lower water bearing zone groundwater samples specifically for strontium-90 (Sr-90); however, the environmental surveillance program is currently under review and future sampling will likely include groundwater samples collected from lower water bearing zone wells and analyzed for specific beta emitters that are known or suspected to be in the IWCS.

The USACE has some more recent environmental monitoring data for Sr-90 and other potential contaminants associated with KAPL waste. Three upper water bearing zone wells near Building 401 and the IWCS (201A, BH49A, and 0W-11B) were sampled in the fall and spring of 2009. No H-3, Sr-90, Tc-99, Cs-137, Pu-238 or Pu-239 were detected during those sampling events. Please see Tables 9-1 and 9-2, which will be incorporated into the 2009 Environmental Surveillance Technical Memorandum.

The Corps has not discussed its findings of gross beta contamination in the LWBZ groundwater with the public.

The USACE has been relying on isotopic analysis of groundwater samples, rather than gross alpha and beta measurements, to monitor potential movement of specific nuclides present in the IWCS to groundwater. Although there is a possibility of Sr-90 contamination in the IWCS, the amount of Sr-90 is likely smaller relative to the radium-226 and uranium present. In addition, uranium is at least as, if not more, mobile than strontium is in the subsurface. Given the confirmed higher presence of uranium in the IWCS (relative to Sr-90) and its similar or higher relative mobility, uranium-specific analyses which we have been performing in both the upper water bearing zone (UWBZ) and LWBZ make uranium a better indicator for IWCS breach. Note that uranium was not detected above background in the groundwater samples obtained from the LWBZ wells during the Remedial Investigation.

Section 4.3.2.1 of the Groundwater Flow Model http://www.lrb.usace.army.mil/fusrap/nfss/nfss-groundwatermodel-narrative-2007-12.pdf contains a discussion of which constituents would be the best indicators of leakage from IWCS (and hence included in the groundwater flow modeling effort for NFSS). Table 4-2 is not available on-line and is attached.

e) Uranium contamination in groundwater south of the IWCS

The Corps believes high levels of uranium contamination in wells south of the IWCS are from past storage practices and therefore discounts leakage from the IWCS. The Corps bases its opinion solely on a review of historical aerial photographs which show mounded material around Building 409. However, historical surveys of the area south of the IWCS contradict the belief that the mounded material contaminated the area. This is illustrated in one of the Corps' own reference documents (provided with the July 9, 2010 letter): Fig 2-3, from

the 1981 Bechtel Characterization and Hazard Assessment, shows no contamination around Building 409.

USACE Response: In Figure 2-3 of the *Chemical Characterization Report*, Bechtel National (DOE Contractor) highlights "areas of known contamination" in 1981, prior to the construction of the IWCS and also prior to interim remedial action activities undertaken on NFSS by DOE. It is correct that contamination in this figure appears well south of Building 409.

Figure 3-2 of the Post *Remedial Action Report for the Niagara Falls Storage Site*, (Bechtel, 1996) documents what portions of NFSS ultimately required remedial action due to the presence of contamination. This figure shows the "West Drainage Area," which includes areas surrounding Building 409 itself, was remediated by DOE between 1982 and 1983, prior to the final construction of the IWCS. It appears that the estimated extent of contamination in 1981 was an underestimate of the actual extent surrounding Building 409 (as identified during remedial action in this area between 1982 and 1983).

The Corps is committed to ensuring that the IWCS is functioning as designed and will continue to monitor radon flux from the IWCS surface, radon and external gamma radiation around the perimeter of the IWCS and site, and sample groundwater and streambed surface water and sediment.

f) Uranium contamination east of the IWCS

The Corps asserts that long term trends in the environmental surveillance groundwater data show steady-state to declining contaminant concentration levels. This is incorrect. Analysis of the contaminant trend for uranium in well OW-11B shows uranium has increased to 254pCi/L in 2008 compared to 216pCi/L in 2003, 133pCi/L in 2000 and 32pCi/L in 1992. The Corps does not combine environmental surveillance data with data from the RI, so has overlooked the steadily increasing levels of uranium in well OW-11B, which is an indication of IWCS leakage.

USACE Response: The Corps does acknowledge that the long term uranium levels in groundwater samples collected from UWBZ monitoring well OW11B show a slightly increasing trend; however, the Corps disagrees that this data is indicative of a breach in the IWCS. Well OW11B is not used to monitor IWCS integrity but was added by the Corps to the environmental surveillance program in 2008 to monitor elevated uranium concentrations detected during the Remedial Investigation (NFSS Remedial Investigation Report, December 2007).

Upper water bearing zone wells MW862, A50, A 51, and MW860 are located within approximately 30 feet of the eastern IWCS boundary and as such, are a much better

indicator of IWCS integrity than well OW11B, which is located over 180 feet east (upgradient) and across the Central Drainage Ditch from the IWCS. Well A50 is part of the environmental surveillance program and data from this well has shown no increase in radionuclide concentrations. The remaining wells (MW862, A51, and MW860) were sampled during the RI and exhibited much lower total uranium concentrations (less than 17 pCi/L) than OW11B, indicating that the IWCS is functioning as designed.

The Corps will continue to closely monitor the quality of groundwater surrounding the IWCS as part of the ongoing environmental surveillance program sampling. As previously stated, the Corps is in the process of modifying the environmental surveillance program to include several wells screened in the lower water bearing zone and to include additional analytical parameters.

g) Radium-226 Detection in the LWBZ

The Corps states that the 1992 radium-226 detection in the LWBZ would have been accompanied by uranium if the source was IWCS leakage, since uranium is much more mobile than radium in the groundwater at the NFSS. This is incorrect.

Although uranium is generally more mobile than radium, the presence of iron in groundwater has been shown to immobilize uranium in the NFSS groundwater. The LWBZ groundwater, including well OW-15A, is high in iron so uranium would not be expected to be present with radium.

The Corps states that I requested Well OW-15A be sampled in order to verify whether the 1993 radium detect was a result of turbidity, but this is not correct. I accept that turbidity may be an issue in the UWBZ wells where contamination in the subsurface soil is common owing to past impacts, but do not believe that turbidity is an issue in the LWBZ wells, since significant contamination of the soil should be absent. The Corps attempt to speculate on and dismiss published groundwater data is an inappropriate response. I find it disturbing that both DOE and the Corps do not monitor the LWBZ groundwater around the IWCS and that the Corps has not investigated the source of gross beta and radium contamination in the LWBZ around the IWCS.

USACE Response: Please refer to the response to question c.

h) Cesium-137 Detections in Groundwater ([redacted])

The Corps asserts that cesium-137 detections are likely due to interference from soil floating in the groundwater and that the source of the cesium-137 in soil may

be past fall out. This is nonsense. There is no history of impacts involving cesium-137 immediately east of the IWCS, which would lead to cesium contamination in the soil and cesium-137 fall out is far too low to account for the high level of cesium (57.1pCi/L) detected in groundwater. The Corps should review its follow-up sampling to determine why the Corps initial sample results were not replicated in subsequent rounds of sampling. By dismissing its own analytical data, the Corps is again attempting to make the data fit in with the Corps preconceived opinion that the IWCS is not leaking.

USACE Response: Please also see the response to question (d). In addition, analyses of water samples obtained from OW11B (the well that had the cesium-137 result of 57.1 pCi/L) in 2008 and 2009 did not result in positive detections of Cs-137. The previous elevated detection could not be replicated.

(i) Investigation of the LWBZ Groundwater

The Corps states that 39 of 42 (or 93%) of the LWBZ wells were sampled across the site, but does not compare that with the 160 wells and TWPs sampled in the UWBZ groundwater. Reviewing the distribution of the LWBZ wells, shows that 24 (or 62%) of the LWBZ wells sampled were located in a single EU, leaving only 15 LWBZ groundwater wells to investigate the groundwater under 165 acres of the 191 acre NFSS site.

USACE Response: Please refer to the response to question c.

It remains our position that the integrity of the IWCS is performing as designed. We firmly believe that the results of the Environmental Surveillance Program support that conclusion. We are modifying the program in response to community concerns. We would like to schedule a conference call with you once you have reviewed our response to verify that your concerns have been addressed. We look forward to working with you and the community during the Feasibility Study development for the IWCS. Please contact [redacted] at your convenience to schedule a conference call.

Sincerely,

[redacted]

NFSS Project Manager

Enclosures

Niagara Falls Storage Site

Groundwater Wells Sampled on 24JUN2010

After 23JUN2010 Ontario-Quebec Border 5.0 Mag. Earth Quake

Validated Radiological Data

Well ID	Filtered	Analysis	Detected	Result	Units		Uncertainity	Minimum Detectable Activity	Lab Qualifiers	Validated Qualifiers	Usability	NY State- Unrestricted Use**	NY State- Restricted Use -Industrial**	DOE Cleanup Criteria**
862	No	Radium-226	No	0.149	pCi/l	±	0.163	0.319	U	U	YES	5[a]	5[a]	100[a]
862	No	Radium-228	YES	0.940	pCi/l	±	0.074	0.475			YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.940*	*pCi/l*							5[a]	5[a]	100[a]
862	No	Thorium-228	No	0.097	pCi/l	±	0.089	0.133	U	U	YES	15[b]	NE	400
862	No	Thorium-230	No	0.161	pCi/l	±	0.175	0.334	U	U	YES	15[b]	NE	300
862	No	Thorium-232	YES	0.162	pCi/l	±	0.094	0.127			YES	15[b]	NE	50
		Total Thorium[b]		*0.162*	*pCi/l*							15[b]	NE	NE
862	No	Uranium-234	YES	4.360	pCi/l	±	0.562	0.432	B		YES	27[c]	NE	600[c]
862	No	Uranium-235	No	0.000	pCi/l	±	0.196	0.35	U	U	YES	27[c]	NE	600[c]
862	No	Uranium-238	YES	2.550	pCi/l	±	0.432	0.341			YES	27[c]	NE	600[c]
		Total Uranium[c]		*6.910*	*pCi/l*	=	*7.678*	*µg/L*				27[c]	NE	600[c]
862F	Yes	Radium-226	YES	0.463	pCi/l	±	0.283	0.284			YES	5[a]	5[a]	100[a]
862F	Yes	Radium-228	No	0.142	pCi/l	±	0.054	0.361	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.463*	*pCi/l*							5[a]	5[a]	100[a]
862F	Yes	Thorium-228	YES	0.233	pCi/l	±	0.114	0.114			YES	15[b]	NE	400
862F	Yes	Thorium-230	YES	0.442	pCi/l	±	0.202	0.349			YES	15[b]	NE	300
862F	Yes	Thorium-232	YES	0.144	pCi/l	±	0.086	0.079			YES	15[b]	NE	50
		Total Thorium[b]		*0.819*	*pCi/l*							15[b]	NE	NE
862F	Yes	Uranium-234	YES	4.460	pCi/l	±	0.492	0.311	B		YES	27[c]	NE	600[c]
862F	Yes	Uranium-235	No	0.180	pCi/l	±	0.136	0.181	U	U	YES	27[c]	NE	600[c]
862F	Yes	Uranium-238	YES	3.530	pCi/l	±	0.419	0.167			YES	27[c]	NE	600[c]
		Total Uranium[c]		*7.990*	*pCi/l*	=	*8.878*	*µg/L*				27[c]	NE	600[c]
863	No	Radium-226	No	0.189	pCi/l	±	0.171	0.331	U	U	YES	5[a]	5[a]	100[a]
863	No	Radium-228	YES	1.580	pCi/l	±	0.075	0.415			YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.189*	*pCi/l*							5[a]	5[a]	100[a]
863	No	Thorium-228	YES	0.495	pCi/l	±	0.163	0.19			YES	15[b]	NE	400
863	No	Thorium-230	YES	0.709	pCi/l	±	0.216	0.341			YES	15[b]	NE	300
863	No	Thorium-232	YES	0.221	pCi/l	±	0.126	0.168			YES	15[b]	NE	50
		Total Thorium[b]		*1.425*	*pCi/l*							15[b]	NE	NE
863	No	Uranium-234	YES	1.200	pCi/l	±	0.304	0.096	B	J	YES	27[c]	NE	600[c]
863	No	Uranium-235	YES	0.153	pCi/l	±	0.11	0.056			YES	27[c]	NE	600[c]
863	No	Uranium-238	YES	0.803	pCi/l	±	0.252	0.106		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*2.156*	*pCi/l*	=	*2.396*	*µg/L*				27[c]	NE	600[c]
A42	No	Radium-226	YES	0.341	pCi/l	±	0.21	0.321			YES	5[a]	5[a]	100[a]
A42	No	Radium-228	No	0.000	pCi/l	±	0.066	0.409	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.341*	*pCi/l*							5[a]	5[a]	100[a]
A42	No	Thorium-228	YES	0.181	pCi/l	±	0.078	0.084			YES	15[b]	NE	400
A42	No	Thorium-230	No	0.057	pCi/l	±	0.136	0.286	U	U	YES	15[b]	NE	300
A42	No	Thorium-232	No	0.066	pCi/l	±	0.064	0.092	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.181*	*pCi/l*							15[b]	NE	NE

Niagara Falls Storage Site

Groundwater Wells Sampled on 24JUN2010

After 23JUN2010 Ontario-Quebec Border 5.0 Mag. Earth Quake

Validated Radiological Data

Well ID	Filtered	Analysis	Detected	Result	Units		Uncertainity	Minimum Detectable Activity	Lab Qualifiers	Validated Qualifiers	Usability	NY State- Unrestricted Use**	NY State- Restricted Use -Industrial**	DOE Cleanup Criteria**
A42	No	Uranium-234	YES	18.600	pCi/l	±	0.956	0.202	B		YES	27[c]	NE	600[c]
A42	No	Uranium-235	No	0.242	pCi/l	±	0.179	0.255	U	U	YES	27[c]	NE	600[c]
A42	No	Uranium-238	YES	18.900	pCi/l	±	0.967	0.242			YES	27[c]	NE	600[c]
		Total Uranium[c]		*37.500*	*pCi/l*	=	*41.667*	*µg/L*				27[c]	NE	600[c]
A42-F	Yes	Radium-226	No	0.000	pCi/l	±	0.314	0.798	U	U	YES	5[a]	5[a]	100[a]
A42-F	Yes	Radium-228	No	0.000	pCi/l	±	0.059	0.296	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
A42-F	Yes	Thorium-228	No	0.064	pCi/l	±	0.078	0.117	U	U	YES	15[b]	NE	400
A42-F	Yes	Thorium-230	No	0.050	pCi/l	±	0.125	0.279	U	U	YES	15[b]	NE	300
A42-F	Yes	Thorium-232	YES	0.132	pCi/l	±	0.078	0.092			YES	15[b]	NE	50
		Total Thorium[b]		*0.132*	*pCi/l*							15[b]	NE	NE
A42-F	Yes	Uranium-234	YES	17.200	pCi/l	±	1.3	0.249	B		YES	27[c]	NE	600[c]
A42-F	Yes	Uranium-235	YES	0.437	pCi/l	±	0.224	0.163			YES	27[c]	NE	600[c]
A42-F	Yes	Uranium-238	YES	15.700	pCi/l	±	1.24	0.243			YES	27[c]	NE	600[c]
		Total Uranium[c]		*33.337*	*pCi/l*	=	*37.041*	*µg/L*				27[c]	NE	600[c]
OW04A	No	Radium-226	No	0.281	pCi/l	±	0.203	0.32	U	U	YES	5[a]	5[a]	100[a]
OW04A	No	Radium-228	No	0.000	pCi/l	±	0.065	0.358	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
OW04A	No	Thorium-228	No	0.086	pCi/l	±	0.124	0.196	U	U	YES	15[b]	NE	400
OW04A	No	Thorium-230	YES	0.343	pCi/l	±	0.163	0.298			YES	15[b]	NE	300
OW04A	No	Thorium-232	No	0.000	pCi/l	±	0.109	0.191	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.343*	*pCi/l*							15[b]	NE	NE
OW04A	No	Uranium-234	YES	1.060	pCi/l	±	0.219	0.178	B		YES	27[c]	NE	600[c]
OW04A	No	Uranium-235	YES	0.269	pCi/l	±	0.11	0.096			YES	27[c]	NE	600[c]
OW04A	No	Uranium-238	YES	0.498	pCi/l	±	0.151	0.131		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*1.827*	*pCi/l*	=	*2.030*	*µg/L*				27[c]	NE	600[c]
OW04A-F	Yes	Radium-226	No	0.081	pCi/l	±	0.149	0.327	U	U	YES	5[a]	5[a]	100[a]
OW04A-F	Yes	Radium-228	No	0.143	pCi/l	±	0.06	0.329	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
OW04A-F	Yes	Thorium-228	No	0.049	pCi/l	±	0.101	0.163	U	U	YES	15[b]	NE	400
OW04A-F	Yes	Thorium-230	YES	0.348	pCi/l	±	0.171	0.31			YES	15[b]	NE	300
OW04A-F	Yes	Thorium-232	No	0.106	pCi/l	±	0.089	0.12	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.348*	*pCi/l*							15[b]	NE	NE
OW04A-F	Yes	Uranium-234	YES	0.914	pCi/l	±	0.21	0.168	B	J	YES	27[c]	NE	600[c]
OW04A-F	Yes	Uranium-235	YES	0.133	pCi/l	±	0.092	0.11			YES	27[c]	NE	600[c]
OW04A-F	Yes	Uranium-238	YES	0.490	pCi/l	±	0.157	0.141		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*1.537*	*pCi/l*	=	*1.708*	*µg/L*				27[c]	NE	600[c]
OW04B	No	Radium-226	No	0.051	pCi/l	±	0.145	0.346	U	U	YES	5[a]	5[a]	100[a]
OW04B	No	Radium-228	No	0.424	pCi/l	±	0.066	0.473	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]

Niagara Falls Storage Site

Groundwater Wells Sampled on 24JUN2010

After 23JUN2010 Ontario-Quebec Border 5.0 Mag. Earth Quake

Validated Radiological Data

Well ID	Filtered	Analysis	Detected	Result	Units		Uncertainity	Minimum Detectable Activity	Lab Qualifiers	Validated Qualifiers	Usability	NY State- Unrestricted Use**	NY State- Restricted Use -Industrial**	DOE Cleanup Criteria**
OW04B	No	Thorium-228	No	0.000	pCi/l	±	0.152	0.276	U	U	YES	15[b]	NE	400
OW04B	No	Thorium-230	No	0.108	pCi/l	±	0.2	0.38	U	U	YES	15[b]	NE	300
OW04B	No	Thorium-232	YES	0.212	pCi/l	±	0.118	0.164			YES	15[b]	NE	50
		Total Thorium[b]		*0.212*	*pCi/l*							15[b]	NE	NE
OW04B	No	Uranium-234	YES	9.270	pCi/l	±	0.709	0.131	B		YES	27[c]	NE	600[c]
OW04B	No	Uranium-235	YES	0.246	pCi/l	±	0.118	0.056			YES	27[c]	NE	600[c]
OW04B	No	Uranium-238	YES	8.890	pCi/l	±	0.693	0.092			YES	27[c]	NE	600[c]
		Total Uranium[c]		*18.406*	*pCi/l*	=	*20.451*	*µg/L*				27[c]	NE	600[c]
OW04B-F	Yes	Radium-226	YES	0.406	pCi/l	±	0.178	0.154			YES	5[a]	5[a]	100[a]
OW04B-F	Yes	Radium-228	No	0.000	pCi/l	±	0.058	0.34	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.406*	*pCi/l*							5[a]	5[a]	100[a]
OW04B-F	Yes	Thorium-228	YES	0.181	pCi/l	±	0.089	0.094			YES	15[b]	NE	400
OW04B-F	Yes	Thorium-230	YES	0.366	pCi/l	±	0.16	0.288			YES	15[b]	NE	300
OW04B-F	Yes	Thorium-232	YES	0.104	pCi/l	±	0.065	0.066			YES	15[b]	NE	50
		Total Thorium[b]		*0.651*	*pCi/l*							15[b]	NE	NE
OW04B-F	Yes	Uranium-234	YES	9.450	pCi/l	±	0.783	0.166	B		YES	27[c]	NE	600[c]
OW04B-F	Yes	Uranium-235	YES	0.294	pCi/l	±	0.148	0.107			YES	27[c]	NE	600[c]
OW04B-F	Yes	Uranium-238	YES	9.360	pCi/l	±	0.779	0.149			YES	27[c]	NE	600[c]
		Total Uranium[c]		*19.104*	*pCi/l*	=	*21.227*	*µg/L*				27[c]	NE	600[c]
OW06A	No	Radium-226	No	0.091	pCi/l	±	0.119	0.231	U	U	YES	5[a]	5[a]	100[a]
OW06A	No	Radium-228	No	0.048	pCi/l	±	0.06	0.342	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
OW06A	No	Thorium-228	YES	0.249	pCi/l	±	0.119	0.15			YES	15[b]	NE	400
OW06A	No	Thorium-230	No	0.102	pCi/l	±	0.162	0.327	U	U	YES	15[b]	NE	300
OW06A	No	Thorium-232	YES	0.162	pCi/l	±	0.091	0.109		J	YES	15[b]	NE	50
		Total Thorium[b]		*0.513*	*pCi/l*							15[b]	NE	NE
OW06A	No	Uranium-234	YES	0.379	pCi/l	±	0.162	0.137	B	R	NO	27[c]	NE	600[c]
OW06A	No	Uranium-235	YES	0.088	pCi/l	±	0.078	0.074			YES	27[c]	NE	600[c]
OW06A	No	Uranium-238	YES	0.111	pCi/l	±	0.089	0.088		R	NO	27[c]	NE	600[c]
		Total Uranium[c]		*0.578*	*pCi/l*	=	*0.642*	*µg/L*				27[c]	NE	600[c]
OW06A-F	Yes	Radium-226	No	0.000	pCi/l	±	0.147	0.585	U	U	YES	5[a]	5[a]	100[a]
OW06A-F	Yes	Radium-228	No	0.052	pCi/l	±	0.07	0.386	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
OW06A-F	Yes	Thorium-228	YES	0.117	pCi/l	±	0.084	0.115			YES	15[b]	NE	400
OW06A-F	Yes	Thorium-230	No	0.197	pCi/l	±	0.171	0.326	U	U	YES	15[b]	NE	300
OW06A-F	Yes	Thorium-232	No	0.091	pCi/l	±	0.08	0.115	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.117*	*pCi/l*							15[b]	NE	NE
OW06A-F	Yes	Uranium-234	YES	0.767	pCi/l	±	0.19	0.191	B	J	YES	27[c]	NE	600[c]
OW06A-F	Yes	Uranium-235	YES	0.148	pCi/l	±	0.092	0.117			YES	27[c]	NE	600[c]
OW06A-F	Yes	Uranium-238	YES	0.354	pCi/l	±	0.129	0.136		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*1.269*	*pCi/l*	=	*1.410*	*µg/L*				27[c]	NE	600[c]

Niagara Falls Storage Site

Groundwater Wells Sampled on 24JUN2010

After 23JUN2010 Ontario-Quebec Border 5.0 Mag. Earth Quake

Validated Radiological Data

Well ID	Filtered	Analysis	Detected	Result	Units		Uncertainity	Minimum Detectable Activity	Lab Qualifiers	Validated Qualifiers	Usability	NY State- Unrestricted Use**	NY State- Restricted Use -Industrial**	DOE Cleanup Criteria**
OW06B	No	Radium-226	No	0.163	pCi/l	±	0.193	0.383	U	U	YES	5[a]	5[a]	100[a]
OW06B	No	Radium-228	YES	0.792	pCi/l	±	0.066	0.412			YES	5[a]	5[a]	100[a]
		Total Radium[a]		*0.792*	*pCi/l*							5[a]	5[a]	100[a]
OW06B	No	Thorium-228	YES	0.244	pCi/l	±	0.113	0.143			YES	15[b]	NE	400
OW06B	No	Thorium-230	YES	0.696	pCi/l	±	0.188	0.295			YES	15[b]	NE	300
OW06B	No	Thorium-232	No	0.105	pCi/l	±	0.095	0.139	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.940*	*pCi/l*							15[b]	NE	NE
OW06B	No	Uranium-234	YES	4.310	pCi/l	±	0.519	0.281	B		YES	27[c]	NE	600[c]
OW06B	No	Uranium-235	No	0.201	pCi/l	±	0.154	0.202	U	U	YES	27[c]	NE	600[c]
OW06B	No	Uranium-238	YES	3.480	pCi/l	±	0.47	0.27			YES	27[c]	NE	600[c]
		Total Uranium[c]		*7.790*	*pCi/l*	=	*8.656*	*µg/L*				27[c]	NE	600[c]
OW06B-F	Yes	Radium-226	YES	0.401	pCi/l	±	0.22	0.384			YES	5[a]	5[a]	100[a]
OW06B-F	Yes	Radium-228	YES	0.706	pCi/l	±	0.071	0.506			YES	5[a]	5[a]	100[a]
		Total Radium[a]		*1.412*	*pCi/l*							5[a]	5[a]	100[a]
OW06B-F	Yes	Thorium-228	No	0.000	pCi/l	±	0.11	0.201	U	U	YES	15[b]	NE	400
OW06B-F	Yes	Thorium-230	No	0.112	pCi/l	±	0.135	0.293	U	U	YES	15[b]	NE	300
OW06B-F	Yes	Thorium-232	YES	0.070	pCi/l	±	0.055	0.053			YES	15[b]	NE	50
		Total Thorium[b]		*0.070*	*pCi/l*							15[b]	NE	NE
OW06B-F	Yes	Uranium-234	YES	4.420	pCi/l	±	0.529	0.091	B		YES	27[c]	NE	600[c]
OW06B-F	Yes	Uranium-235	No	0.049	pCi/l	±	0.096	0.147	U	U	YES	27[c]	NE	600[c]
OW06B-F	Yes	Uranium-238	YES	3.160	pCi/l	±	0.452	0.138			YES	27[c]	NE	600[c]
		Total Uranium[c]		*7.580*	*pCi/l*	=	*8.422*	*µg/L*				27[c]	NE	600[c]
OW15A	No	Radium-226	No	0.257	pCi/l	±	0.191	0.321	U	U	YES	5[a]	5[a]	100[a]
OW15A	No	Radium-228	YES	1.430	pCi/l	±	0.063	0.415			YES	5[a]	5[a]	100[a]
		Total Radium[a]		*1.430*	*pCi/l*							5[a]	5[a]	100[a]
OW15A	No	Thorium-228	YES	0.293	pCi/l	±	0.132	0.169			YES	15[b]	NE	400
OW15A	No	Thorium-230	YES	0.432	pCi/l	±	0.213	0.37			YES	15[b]	NE	300
OW15A	No	Thorium-232	YES	0.304	pCi/l	±	0.131	0.164			YES	15[b]	NE	50
		Total Thorium[b]		*1.029*	*pCi/l*							15[b]	NE	NE
OW15A	No	Uranium-234	YES	0.836	pCi/l	±	0.215	0.195	B	J	YES	27[c]	NE	600[c]
OW15A	No	Uranium-235	YES	0.145	pCi/l	±	0.105	0.134			YES	27[c]	NE	600[c]
OW15A	No	Uranium-238	YES	0.249	pCi/l	±	0.136	0.166		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*1.230*	*pCi/l*	=	*1.367*	*µg/L*				27[c]	NE	600[c]
OW15A-F	Yes	Radium-226	No	0.141	pCi/l	±	0.135	0.241	U	U	YES	5[a]	5[a]	100[a]
OW15A-F	Yes	Radium-228	No	0.066	pCi/l	±	0.052	0.29	U	U	YES	5[a]	5[a]	100[a]
		Total Radium[a]		*Non-detect*	*pCi/l*							5[a]	5[a]	100[a]
OW15A-F	Yes	Thorium-228	YES	0.189	pCi/l	±	0.095	0.115			YES	15[b]	NE	400
OW15A-F	Yes	Thorium-230	YES	0.481	pCi/l	±	0.197	0.337			YES	15[b]	NE	300
OW15A-F	Yes	Thorium-232	No	0.101	pCi/l	±	0.085	0.119	U	U	YES	15[b]	NE	50
		Total Thorium[b]		*0.670*	*pCi/l*							15[b]	NE	NE
OW15A-F	Yes	Uranium-234	YES	0.494	pCi/l	±	0.189	0.232	B	J	YES	27[c]	NE	600[c]
OW15A-F	Yes	Uranium-235	YES	0.115	pCi/l	±	0.088	0.112			YES	27[c]	NE	600[c]
OW15A-F	Yes	Uranium-238	YES	0.161	pCi/l	±	0.102	0.126		J	YES	27[c]	NE	600[c]
		Total Uranium[c]		*0.770*	*pCi/l*	=	*0.856*	*µg/L*				27[c]	NE	600[c]

Niagara Falls Storage Site

Groundwater Wells Sampled on 24JUN2010
After 23JUN2010 Ontario-Quebec Border 5.0 Mag. Earth Quake
Validated Radiological Data

Well ID	Filtered	Analysis	Detected	Result	Units		Uncertainity	Minimum Detectable Activity	Lab Qualifiers	Validated Qualifiers	Usability	NY State- Unrestricted Use**	NY State- Restricted Use -Industrial**	DOE Cleanup Criteria**

Column Headings:

Well ID -NFSS Well Identifier
"-F" denotes sample was field filtered

Filtered - Yes =Field filtered with in-line 0.45 micron filter
No = Not filtered
*Well #: 863 purged dry before obtaining a filtered sample

Analysis - radiological isotope analysis:

Radiological Isotope	Method
Radium-226	SM 7500 Ra B M
Radium-228	EPA 904
Thorium-228, 230 and 232	LANL ER 200 M
Uranium-234, 235 and 238	ASTM D 3972/DOE U-02

Detected - YES =radiological isotope detected (Result) above Minimum Detection Activity
No = radiological isotope not detected above Minimum Detectable Activity

Units: pCi/l -Pico curies per liter
Uncertainty -Calculated +/- of radiological result
Minimum Detectable Activity - Minimum detectable activity for that radiological isotope.

Lab Qualifiers - Blank/Empty field - finding above minimum detectable activity
U - non-detect
B - method blank result exceeds minimum detectable activity

Validated Qualifiers -
Blank/Empty field - finding above minimum detectable activity
U - non-detect
J - estimated value for finding above minimum detectable activity
R- finding rejected due to possible bias from laboratory contamination

Usability: YES – data finding is usable
NO – data finding is not usable

Niagara Falls Storage Site

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**** Groundwater at NFSS is not a drinking water source.**

The above federal and state regulation concentrations are for comparative purposes only.

Federal Regulations:

National Primary Drinking Water Regulations 40CFR141.62&63

US Dept of Energy:

USDOE derived concentration guide (USDOE Order 5400.5) for drinking water.

NE - Not Established

New York State:

New York State Standards -Water Quality Criteria (class GA) per 6 NYCRR, Part 703.

NE - Not Established

a. Applies to the sum of Ra-226 and Ra-228

b. "Adjusted" gross alpha MCL of 15 pCi/, including Thorium isotopes, excluding radon and uranium
-National Primary Drinking Water Regulations; Radionuclide; Final Rule (Federal Register -December 7, 2000)

c. Sum of Uranium Isotopes (27 pCi/l or 30 µg/L).

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Gross-beta activity in ground water: natural sources and artifacts of sampling and laboratory analysis

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Abstract—Gross-beta activity has been used as an indicator of beta-emitting isotopes in water since at least the early 1950s. Originally designed for detection of radioactive releases from nuclear facilities and weapons tests, analysis of gross-beta activity is widely used in studies of naturally occurring radioactivity in ground water. Analyses of about 800 samples from 5 ground-water regions of the United States provide a basis for evaluating the utility of this measurement. The data suggest that measured gross-beta activities are due to (1) long-lived radionuclides in ground water, and (2) ingrowth of beta-emitting radionuclides during holding times between collection of samples and laboratory measurements.

Although ^{40}K and ^{228}Ra appear to be the primary sources of beta activity in ground water, the sum of ^{40}K plus ^{228}Ra appears to be less than the measured gross-beta activity in most ground-water samples. The difference between the contribution from these radionuclides and gross-beta activity is most pronounced in ground water with gross-beta activities > 10 pCi/L, where these 2 radionuclides account for less than one-half the measured gross-beta activity. One exception is ground water from the Coastal Plain of New Jersey, where ^{40}K plus ^{228}Ra generally contribute most of the gross-beta activity. In contrast, ^{40}K and ^{228}Ra generally contribute most of beta activity in ground water with gross-beta activities < 1 pCi/L.

The gross-beta technique does not measure all beta activity in ground water. Although ^{3}H contributes beta activity to some ground water, it is driven from the sample before counting and therefore is not detected by gross-beta measurements. Beta-emitting radionuclides with half-lives shorter than a few days can decay to low values between sampling and counting. Although little is known about concentrations of most short-lived beta-emitting radionuclides in environmental ground water (water unaffected by direct releases from nuclear facilities and weapons tests), their activities are expected to be low.

Ingrowth of beta-emitting radionuclides during sample holding times can contribute to gross-beta activity, particularly in ground water with gross-beta activities > 10 pCi/L. Ingrowth of beta-emitting progeny of ^{238}U, specifically ^{234}Pa and ^{234}Th, contributes much of the measured gross-beta activity in ground water from 4 of the 5 areas studied. Consequently, gross-beta activity measurements commonly overestimate the abundance of beta-emitting radionuclides actually present in ground water. Differing sample holding times before analysis lead to differing amounts of ingrowth of the two progeny. Therefore, holding times can affect observed gross-beta measurements, particularly in ground water with ^{238}U activities that are moderate to high compared with the activity of ^{40}K plus ^{228}Ra. Uncertainties associated with counting efficiencies for beta particles with different energies further complicate the interpretation of gross-beta measurements.

INTRODUCTION

The gross-beta activity has been used as an indicator of beta-emitting radionuclides in water since at least the early 1950s (Wheler *et al.*, 1954). The gross-beta technique, originally designed for detection of radioactive releases from nuclear facilities and weapons tests, is widely used as a measurement of beta activity in water and for regulatory monitoring of public drinking water (Blanchard *et al.*, 1985; U.S. Environmental Protection Agency, 1986, 1991). Despite widespread use, the limitations of gross-beta activity as an

indicator of the presence of beta-emitting radionuclides present in environmental ground water have not been critically evaluated except briefly by Thomas *et al.* (1993). For the purposes of discussion, ground water unaffected by direct release from, or contact with, material associated with nuclear facilities and weapons testing is referred to as environmental ground water. Water containing ^{3}H derived from atmospheric precipitation since above-ground nuclear weapons testing began in the early 1950s is considered 'environmental'.

The objective of this paper is to evaluate gross-beta activity as an indicator of beta-emitting radionuclides in environmental ground water. Much of this evaluation is related to the effects of sample collection and preparation on gross-beta measurements. The scope is limited to an evaluation of data collected during field investigations as distinct from a laboratory exercise. Gross-beta analyses of about 800 ground-water samples from 5 ground-water regions across the United States have been compiled for this evaluation (Fig. 1). The data for the 5 regions provide an opportunity to evaluate the utility of gross-beta measurements in different geochemical environments. In particular, the assembled data allow a comparison of ground water with low and high ^{238}U activities. This comparison is desirable because the ingrowth of ^{238}U progeny after sample collection has been suggested as a source of gross-beta activity (Thomas *et al.*, 1993).

METHODS OF SAMPLE COLLECTION AND ANALYSIS

Data compiled for this evaluation include measurements of gross-beta activity, K concentrations, and activities of ^{238}U and ^{228}Ra in ground water collected from 1985 through 1992. Ground-water samples for these constituents were collected using positive-displacement pumps in monitoring, domestic, public-supply, and irrigation wells. Samples were collected from points in the distribution system as near to the well as possible and before the water entered a pressure tank. Water samples were obtained at wells after water temperature, specific conductance, and pH were constant and at least 3 well-bore volumes of water were extracted. Water samples were filtered in the field through a 0.45-μm or 0.1 μm pore-size nitrocellulose membrane filter into 1-L acid-cleaned polyethylene bottles. The pH was adjusted to ~ 2 by addition of HNO_3 immediately after filtration.

Most analytical work was done by the U.S. Geological Survey's National Water Quality Laboratory (NWQL) or its contractors following quality control and quality assurance procedures described by Pritt and Raese (1992). Some ground-water samples from New Jersey were analyzed for gross-beta activity and ^{228}Ra by the New Jersey Bureau of Environmental Laboratories Radiation Protection Laboratory using the same procedures as those used by the NWQL.

Gross-beta activities were determined by evaporating to dryness an aliquot of a sample containing ~ 100 mg of dissolved solids and counting the emitted-beta particles using a widely employed method (Barker and Robinson, 1963; Thatcher *et al.*, 1977). Measured activities are reported relative to the $^{90}Sr/^{90}Y$ standard. The analytical method described in detail by Thatcher *et al.* (1977) was used, with the exception that samples were filtered at the time of sampling rather than in the laboratory. Although ^{3}H contributes beta activity to some ground water, it is driven from the sample before counting and therefore is not detected by gross-beta measurements. The method (Thatcher *et al.*, 1977) does not specify a sample holding time. However, the activity of short-lived nuclides can be affected by sample holding times. Holding times for samples discussed here were commonly a few days, but sometimes exceeded 100 days. A long holding time can produce a measured beta activity that does not represent activity at the time of sampling.

Potassium was analyzed by atomic absorption as described by Fishman and Friedman (1989). Potassium-40 activities were not measured directly, but were estimated from K concentrations. Beta activity (in pCi/L) contributed by ^{40}K was estimated by multiplying K concentration (in mg/L) by

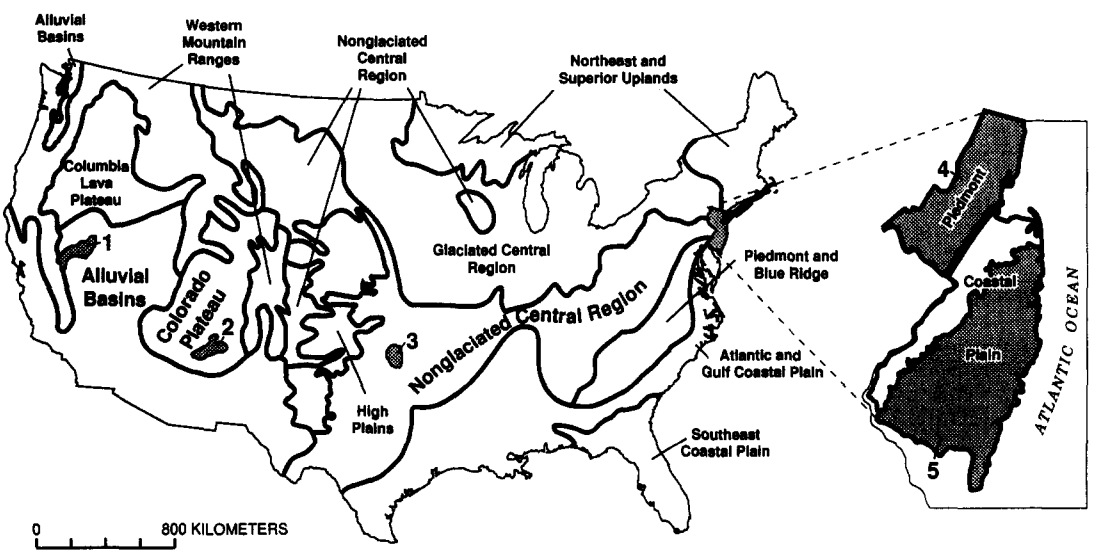

Fig. 1. Location of five areas sampled for gross-beta activity. Numbers and shaded areas indicate (1) Carson River Basin, (2) Puerco River Basin, (3) Central Oklahoma aquifer, (4) Piedmont Province in New Jersey, and (5) eastern Coastal Plain of New Jersey. Ground-water regions modified from Heath (1984).

0.82. This conversion factor is based on a ^{40}K natural abundance of 0.0118% (Beckinsale and Gale, 1969) and a half-life of 1.26×10^9y. Natural isotopic variations of K have been estimated to be <5% (Beckinsale and Gale, 1969), which is consistent with measurements of Morozova and Alferovsky (1974) and Verbeck and Schreiner (1967).

Uranium-238 activities were determined by alpha spectrometry on electroplated samples after separation of U from Th by ion exchange (Thatcher *et al.*, 1977). Uranium in samples containing more than 10,000 mg/L dissolved solids was first precipitated with $Al_2(PO_4)_3$.

Radium-228 activities were determined by chemical separation and beta counting of ^{228}Ac-the progeny of ^{228}Ra in the precipitate (Thatcher *et al.*, 1977).

HYDROGEOLOGY OF AREAS STUDIED

Most ground-water samples discussed in this paper were collected in parts of 5 ground-water regions of the conterminous United States described by Heath (1984). These regions represent various hydrologic and geochemical settings, including the Alluvial Basins, the Colorado Plateau and Wyoming Basin, the Nonglaciated Central region, the Piedmont and Blue Ridge, and the Atlantic and Gulf Coastal Plain (Fig. 1). For brevity, the last 4 regions are called the Colorado Plateau, the Central region, the Piedmont, and the Coastal Plain, respectively.

The Carson River Basin lies largely in the Alluvial Basins ground-water region of the southwestern United States (Welch *et al.*, 1989). A few samples from the Western Mountain Ranges in the Carson River Basin are also included. Most ground-water samples were collected from basin-fill sediments derived primarily from igneous rocks that range in composition from granitic to basaltic. Alluvial, colluvial, and lacustrine processes have formed the sedimentary deposits. Ground water in the upper valleys of the basin generally has dissolved-solids concentrations <500 mg/L. Because the basin is hydrologically closed, dissolved-solids concentrations in the lower part of the basin commonly exceed 1000 mg/L.

In the Puerco River Basin in the southeastern part of the Colorado Plateau, most of the ground-water samples were pumped from alluvial sediments beneath the Puerco River channel (Van Metre and Gray, 1992). The alluvial sediments are predominantly sand with interbedded clay. Sandstone, siltstone, and mudstone of the Chinle Formation underlie the alluvial sediments. Under natural conditions, the river is ephemeral with long dry periods. Since the 1950s, flow in some reaches has been continuous because of discharge from U-mines and a sewage-treatment facility. Alluvial sediments are recharged by infiltration of precipitation and water from the river. Although the U content of the Puerco River has been affected by releases from mining activities, the U in the sampled ground water has largely been removed from solution (Van Metre and Gray, 1992). The dissolved-solids concentration of the sampled ground water ranged from 270 to 3,700 mg/L, with a median concentration of 800 mg/L.

The Central Oklahoma aquifer, as described by Parkhurst *et al.* (1994), is in the Alluvial Valleys and Central region ground-water regions of Heath (1984). Samples from the Alluvial Valleys region (Heath, 1984; not shown in Fig. 1) in central Oklahoma are included with the discussion of the Central region because of geochemical similarity to the underlying ground water. The Central Oklahoma aquifer consists of westward-dipping bedrock units overlain by alluvial and terrace deposits in the stream and river valleys. The bedrock units of the aquifer consist of red or reddish-brown sandstone, mudstone, and siltstones. The alluvial and terrace deposits consist of lenticular beds of unconsolidated sediments ranging in thickness from 0 to 30 m. The western one-third of the aquifer is overlain by the Hennessey Group, a low-permeability confining unit. The entire aquifer is underlain by the Vanoss Formation, which is also a confining unit. Freshwater in the aquifer is underlain by saline water with dissolved-solids concentrations >5000 mg/L at depths ranging from ~30 to ~300 m within the various bedrock units.

Ground water of central and northeastern New Jersey sampled in the Piedmont region (Fig. 1) is in interbedded arkosic sandstones and red and black mudstones of the Newark Supergroup. Uranium-enriched strata are in permeable zones adjacent to organic-rich horizons in both arkosic sandstones (Turner-Peterson, 1980) and black mudstones (Muessig and Houghton, 1988).

The Kirkwood-Cohansey aquifer system (Zapecza, 1989) is the most important unconfined aquifer in the Coastal Plain of southern New Jersey (Fig. 1) in terms of water supply. This aquifer system consists of a wedge of unconsolidated sands and gravels composed primarily of quartz grains (Zapecza, 1989). Trace quantities of minerals containing U, such as zircon and monazite, are present; however, no U-enriched strata are present.

DISCUSSION

Gross-beta activity would be of greatest use in studies of environmental ground water if the results provided an unequivocal, quantitative measure of a known set of beta-emitting radionuclides in ground water at the time of sampling. Gross-beta measurements provide useful information on the presence of radionuclides in environmental water; however, the results can be misleading. Four factors that can affect interpretation of gross-beta measurements are: (1) as mentioned previously, ^3H is not measured by the gross-beta-activity technique; (2) the beta-particle energy is related to the probability of detection. For instance, detection of low-energy beta particles emitted by ^{228}Ra (0.01 MeV, Table 1) is much less likely than the relatively energetic beta particles from ^{40}K (0.51 MeV); (3) the decay of radionuclides during

Table 1. Energy of beta particles emitted by selected radionuclides. Includes radionuclides commonly found in ground water and radionuclides present after sampling

Isotope	Intensity (%)	Average beta energy (MeV)
Gross-beta activity standards		
^{90}Sr	0.20	100
^{90}Y	0.93	100
^{137}Cs	0.16	94.6
Beta-emitting radionuclides commonly found in ground water		
^{40}K	0.51	89
^{228}Ra	0.01	100
Beta-emitting progeny of ^{220}Rn and ^{222}Rn		
^{210}Pb	0.0065	100
^{210}Bi	0.004	80.2
	0.016	19.8
^{212}Pb	0.099 (average of 6 betas)	100
^{212}Bi	0.717 (average of 3 betas)	64.0
Beta-emitting progeny of ^{228}Ra		
^{228}Ac	0.380 (average of 31 betas)	Sum = 97.0
Beta-emitting progeny of ^{238}U		
^{234}Th	0.050	72.5
	0.025	18.5
^{234}Pa	0.22 (average of 18 betas)	Sum = 99.7

sample holding times (the period between sampling and counting) can decrease the beta activity. Because ^{40}K and ^{228}Ra have relatively long half-lives (1.3×10^9 and 6.7 a respectively), they are not materially affected by most holding times; and (4) ingrowth of radionuclides not present in ground water at the time of sampling can contribute to the gross-beta activity. This ingrowth can lead to an overestimate of the beta activity in ground water.

The effects of factors 2–4 on gross-beta measurements are discussed in this section. The first factor is not discussed further because the effect is obvious.

Effects of beta energy on counting

The energy of a beta particle affects the probability that the particle will be counted. Generally, the likelihood of detection increases as the energy of the particle increases. A comparison of two beta-activity standards provides insight into the effect of different beta energies on counting efficiency. Within the recommended range of residue (< 10 mg/cm^2), counting of the ^{137}Cs standard with a beta energy of 0.16 MeV is as much as 25% less efficient than the $^{90}Sr/^{90}Y$ standard (Janzer, 1980), which has an average beta energy of 0.56 MeV. Because a laboratory study of the relative counting efficiency for the beta particles listed in Table 1 is not available, we have made arbitrary assumptions regarding the relationship between beta energy and counting efficiency. Our initial assumptions are (1) beta particles with energies of less than or equal to 0.01 MeV (or 5% of the ^{90}Sr-beta energy) are arbitrarily considered to be too weak to contribute to the gross-beta measurement, and (2) beta particles with energies between the energies of the 2 beta particles emitted by the $^{90}Sr/^{90}Y$ standard (0.2 and 0.93 MeV) have the same probability of being detected as beta particles from the standard. The possible contribution to gross beta by radionuclides that emit beta particles with energies between 0.01 and 0.2 MeV (^{228}Ra and ^{234}Th) is discussed later in this section.

Naturally occurring beta-emitting radionuclides in ground water

Among the radionuclides shown in Table 1, ^{40}K and ^{228}Ra are sufficiently soluble and abundant to contribute beta activities > 1 pCi/L to some ground water. The energies of beta particles emitted by ^{40}K are high enough to be measured by the gross-beta technique. In contrast, ^{228}Ra emits a relatively weak beta particle (Table 1) that may not be detected by gross-beta measurements.

Although it is not expected to be present at the time of sampling, ingrowth of ^{228}Ac from ^{228}Ra (Fig. 2) approaches 95% of secular equilibrium with ^{228}Ra within about a day (Table 2). Although not well studied, Ac probably is sparingly soluble in most ground water (Ferronsky and Polyakov, 1982). Because few samples are analyzed within a day after collection, ^{228}Ac is expected to contribute beta activity equal to that from ^{228}Ra. This conclusion is based on the assumption that ^{228}Ac either remains in solution at a pH of ~ 2 or the time between evaporating a sample to dryness and counting is at least one day. Thus, gross-beta activity from these 2 radionuclides is approximately equal to the activity of ^{228}Ra in ground water at the time of sampling. This conclusion is consistent with the method described earlier for determination of ^{228}Ra activity. Because the ^{228}Ac activity is produced by ingrowth from ^{228}Ra, the contribution is discussed as if it were directly from ^{228}Ra.

In 4 of the 5 regions, at least 75% of the estimated ^{40}K activities are < 5 pCi/L (Fig. 3). Potassium-40

Table 2. Time for ingrowth of ^{238}U and ^{228}Ra progeny to approach secular equilibrium

Radionuclide	Half-life	Time to reach 95% of secular equilibrium	Time to reach 99% of secular equilibrium
^{228}Ac	6.13 h	26.5 h	40.7 h
^{234}Th	24.1 days	104 days	160 days
^{234}Pa	1.2 min	5.1 min	7.8 m

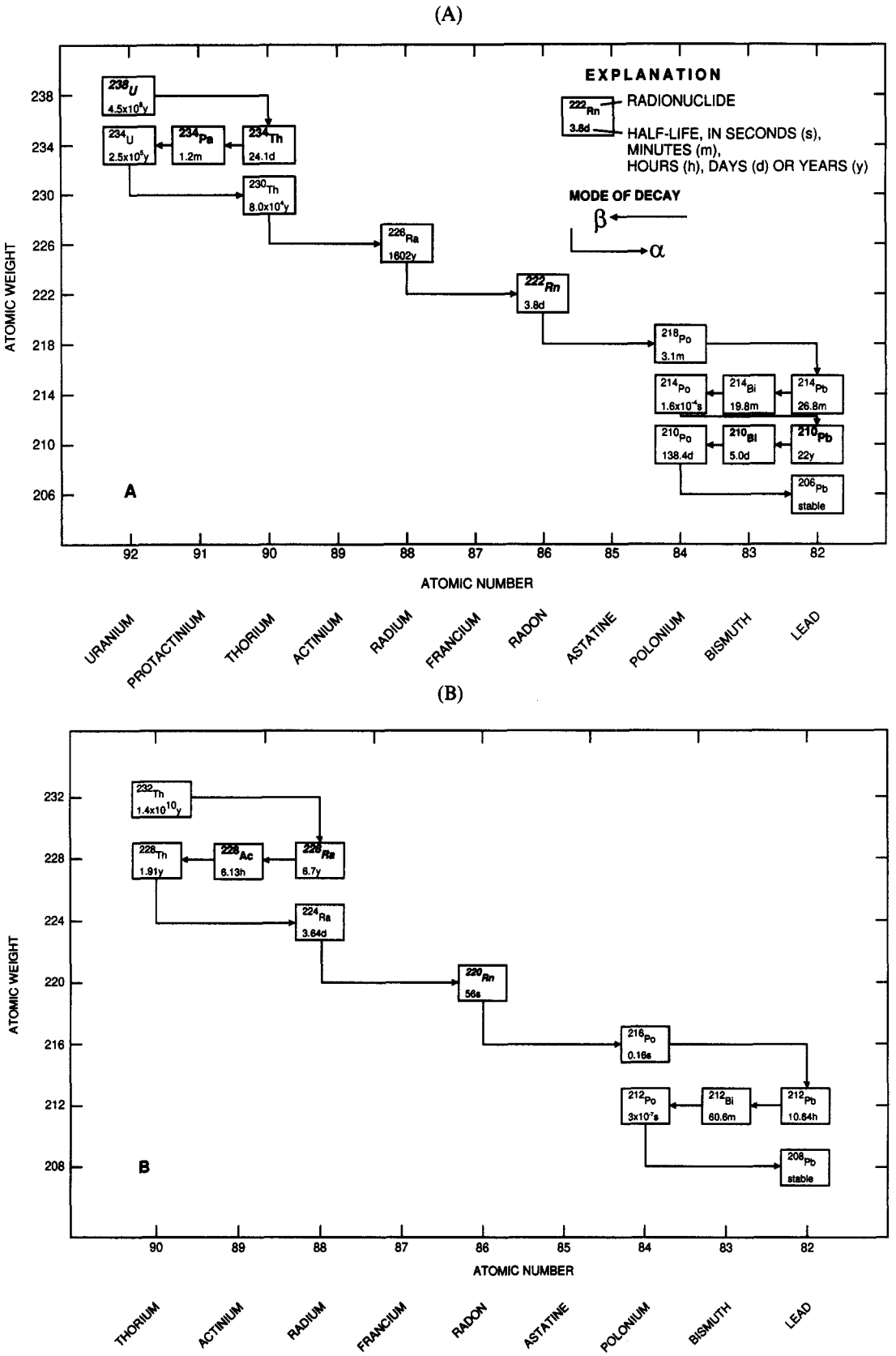

Fig. 2. Potential sources of gross-beta activity in ground-water samples from radionuclides in the (A) ^{238}U and (B) ^{232}Th decay series. Beta-emitting radionuclides shown in bold letters can grow in a sample and contribute 1 pCi/L or more to the total activity. Radionuclides shown in italics can be present with activities greater than 1 pCi/L and produce beta-emitting progeny.

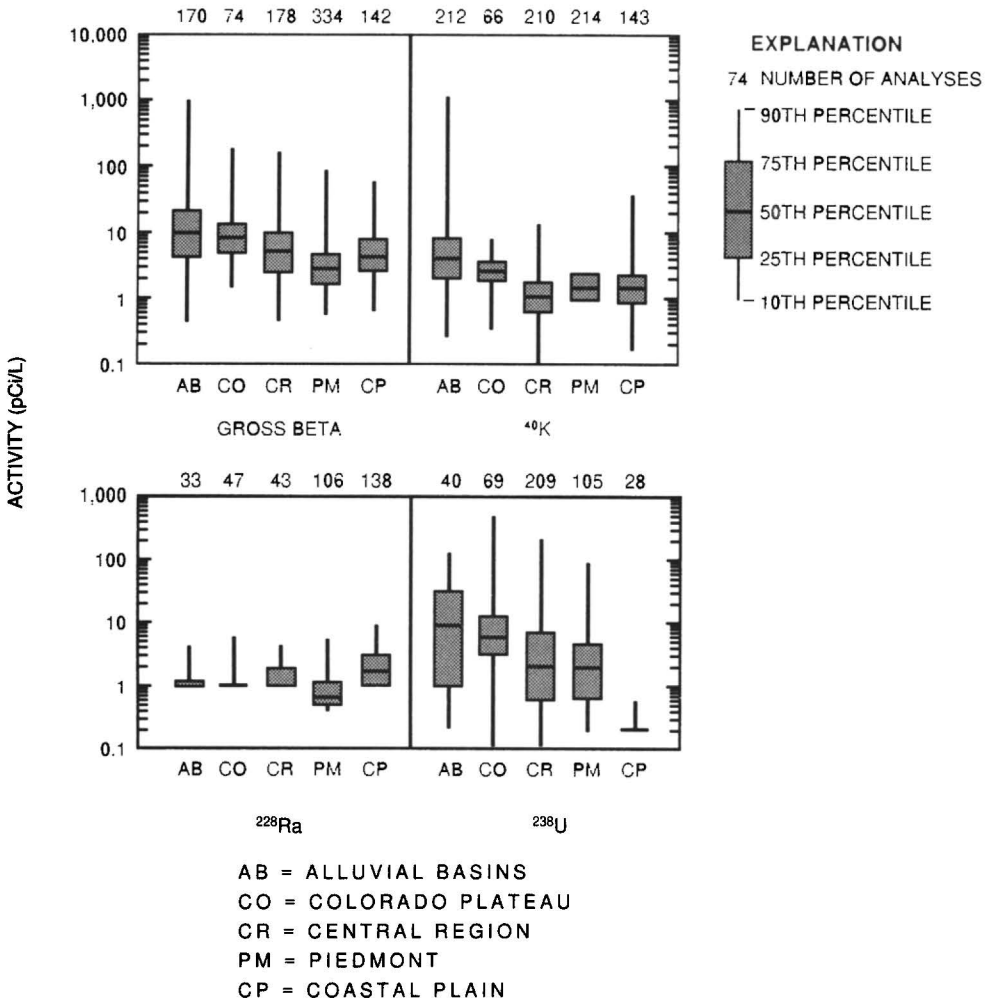

Fig. 3. Summary statistics for gross beta, ^{40}K, ^{228}Ra, and ^{238}U activities in ground water from five regions in the U.S.A.

activities in the Alluvial Basins are commonly greater than those found in the other 4 regions; some samples from this region have ^{40}K activities > 100 pCi/L and 25% are > 8 pCi/L. The higher ^{40}K activities in the Alluvial Basins ground water largely are due to higher K concentrations caused by evaporative concentration in this semi-arid environment.

The median ratios of ^{40}K and ^{228}Ra activities to gross beta provide a measure of the proportion of the gross beta commonly contributed by these 2 radionuclides (Fig. 4). The median ratio of ^{40}K to gross beta is about one-half (0.5 on Fig. 4) or less. Potassium-40 generally contributes a much smaller portion of the gross-beta activity in water with gross-beta activities > 10 pCi/L compared with water with lower gross-beta activities (Fig. 4), except in the Coastal Plain. Most gross-beta activities are greater than the estimated ^{40}K activities (Fig. 5), suggesting an additional source or sources of beta activity.

Radium-228 in ground water is also a source of beta activity (Fig. 6), although the activity is generally less than that from ^{40}K (Figs 3 and 4). In four of the five regions, 75% of the measured ^{228}Ra activities are < 2.5 pCi/L (Fig. 3). Radium-228 contributes a much smaller portion of the gross-beta activity in water with gross-beta activities > 10 pCi/L compared with water with lower gross-beta activities (Fig. 4).

Radium-228 activities in the Coastal Plain samples are generally higher than those in ground water from the other 4 regions (Fig. 3). About 25% of the ^{228}Ra activities exceed 2.5 pCi/L and some are as high as 9 pCi/L in this one region. Unlike in the other 4 regions, ^{228}Ra is the dominant beta-emitting radionuclide in ground water with gross-beta activities < 10 pCi/L from the Coastal Plain. A few samples with ^{228}Ra activities equal to 1 pCi/L have gross-beta activities < 1 pCi/L. Because the analytical uncertainty is relatively great for samples with low activities, the ^{228}Ra activities are within the analytical uncertainty of the gross-beta activities.

In general, the sum of ^{40}K plus ^{228}Ra does not account for all of the gross-beta activity, particularly

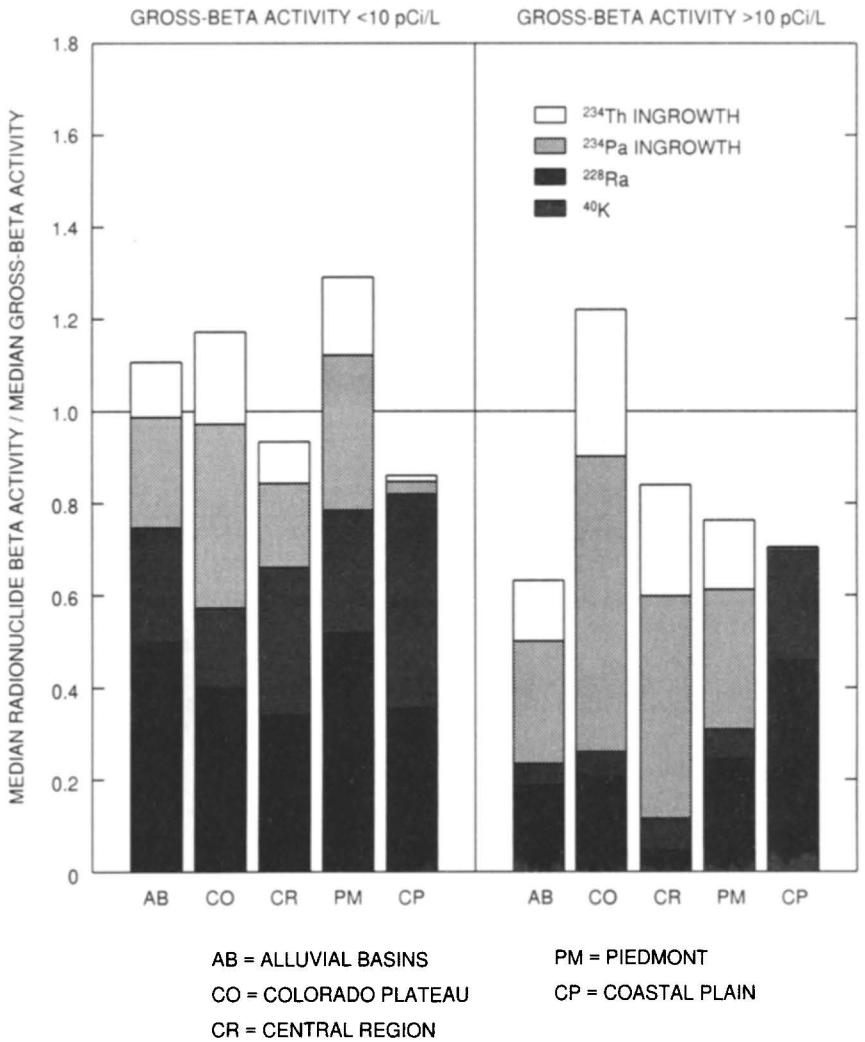

Fig. 4. Median ratio of gross-beta activity from different sources relative to measured gross-beta activities.

in samples with a gross-beta activity > 10 pCi/L. The presence of other beta-emitting radionuclides in ground water at the time of sampling, ingrowth of beta-emitting radionuclides, or both, could explain this lack of correspondence. An evaluation of beta-emitting radionuclides that could be present in ground water is given below, based on known abundances and solubilities.

Radionuclides in the ^{235}U decay series probably are not a quantitatively important source of beta-emitting radionuclides because of low natural abundance of the parent and low solubility of U in most ground water. Uranium-235 constitutes < 5% of naturally occurring U, on an activity basis (Ferronsky and Polyakov, 1982).

Thorium-234, ^{210}Pb, and ^{210}Bi emit beta particles and have half-lives greater than a few days (Fig. 2A). Although these radionuclides could contribute to the gross-beta activity, they are not believed to be quantitatively important contributors to the gross-beta activity of ground water because their solubility is low. Thorium-234 produced by the decay of ^{238}U in the ground water is rapidly adsorbed onto aquifer materials (Krishnaswami *et al.*, 1982; Landa, 1980, 1982; Langmuir and Herman, 1980; Laflamme and Murray, 1987). Thus, ^{234}Th activities should be very low in ground water (Wanty and Nordstrom, 1993). Low ^{230}Th activities in 2 samples from the Alluvial Basins (0.15 and 0.20 pCi/L, Thomas *et al.*, 1993) and 16 samples from the Colorado Plateau (all < 1.0 pCi/L) are consistent with the expectation that ^{234}Th activities are low in ground water.

Lead-210 activities are also expected to be low in ground water because of rapid adsorption (Hussain and Krishnaswami, 1982; Krishnaswami *et al.*, 1982). The low energy of the emitted beta particles (Table 1) would limit detection due to low counting efficiency.

No information on ^{210}Bi activities in ground water is available. Nonetheless, the short half-life (5 days) combined with sample holding times exceeding a few

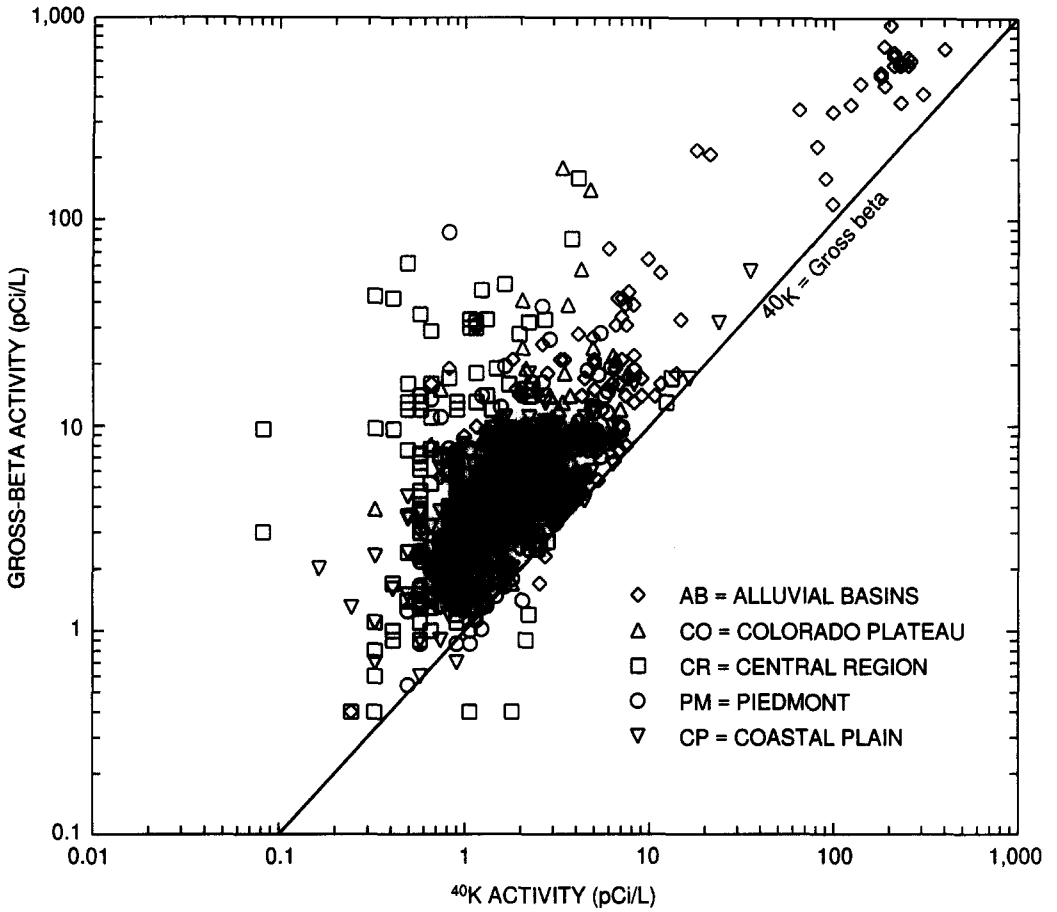

Fig. 5. Relation between gross-beta activity and ^{40}K. Data for 765 analyses shown.

weeks would result in decay of most or essentially all ^{210}Bi before beta counting. Lead-212, ^{212}Bi, and ^{234}Pa also emit beta particles, but their half-lives (10.64 h, 60.6 min, and 1.2 min, respectively, Fig. 2) are short compared with typical sample holding times for gross-beta analysis. Accordingly, these 3 radionuclides are not expected to contribute beta activity from their presence in ground water at the time of sampling. Possible contribution to gross beta by Pb, Bi, Th, and Pa isotopes produced from U and Rn in ground water is discussed in the following section.

In summary, ^{40}K and ^{228}Ra appear to be the primary sources of beta activity in ground water. The sum of ^{40}K plus ^{228}Ra appears to be less than the measured gross-beta activity in most ground water samples. The difference between the contribution from these isotopes and gross beta is most pronounced in ground water with gross-beta activities > 10 pCi/L. Low solubility and short half-lives of beta-emitting isotopes of Th, Pb, and Bi suggest that these isotopes are not important contributors to the gross-beta activity. Accordingly, variations in gross-beta activity from these short-lived radionuclides are expected to be insignificant.

Additional sources of beta-emitting radionuclides

Although the effects of sample holding times (the period between sample collection and counting) on gross-alpha activity have been recognized (Oural *et al.*, 1988), the effects on gross-beta activity seem to have received little consideration. One exception is the ingrowth of ^{228}Ac from ^{228}Ra, as discussed above. Sample holding times, which commonly are a few tens to several hundred days, allow ingrowth of several radionuclides. Ingrowth can occur either before or after the sample is dried. However, the time for ingrowth after drying generally is short because samples commonly are counted within a few days after drying. Uranium-238, ^{220}Rn, and ^{222}Rn are sufficiently abundant in some ground water to produce beta-emitting progeny at levels > 1 pCi/L at some time after sampling. These parent radionuclides and their beta-emitting progeny are discussed in this section.

Many short-lived beta-emitting radionuclides can be produced between sample collection and analysis. The cations Th and Pa are sparingly soluble in most ground-water systems, but can remain in a sample

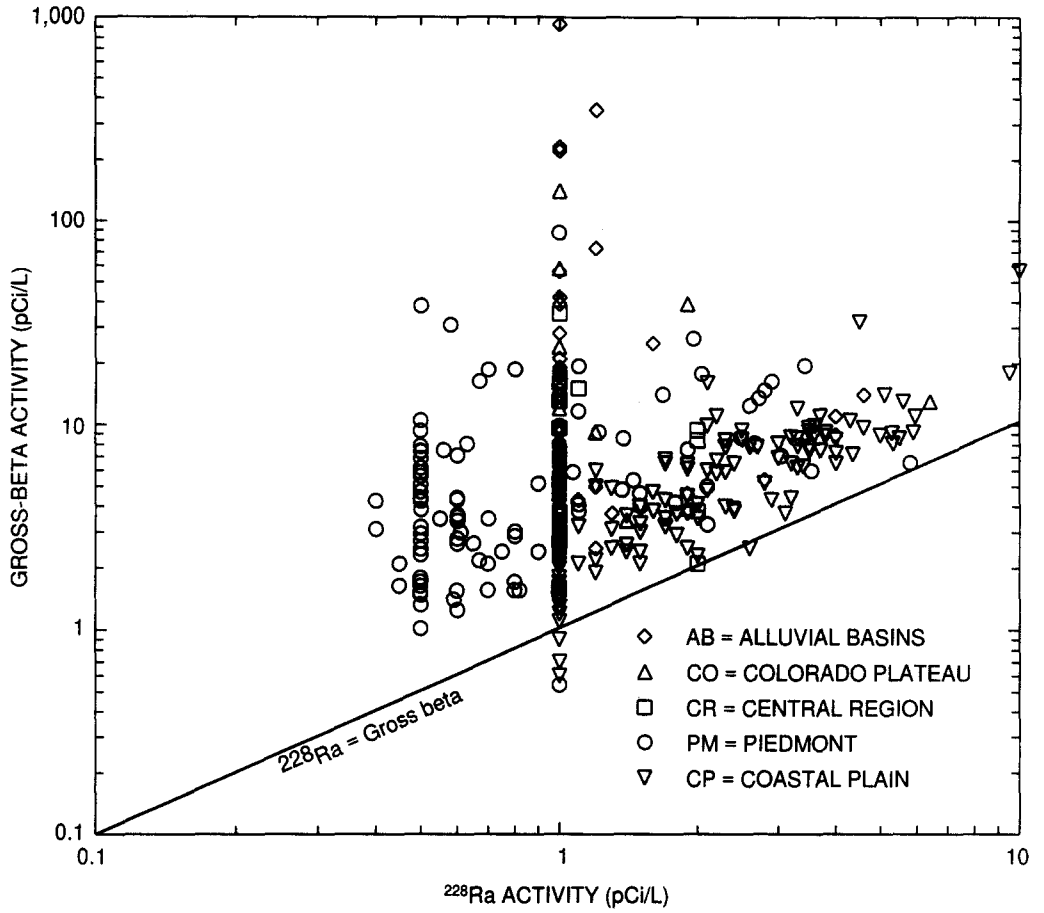

Fig. 6. Relation between gross-beta activity and ^{228}Ra. Data for 336 analyses shown.

after ingrowth because of the pH adjustment to ~2. This pH adjustment inhibits adsorption of cations on the walls of sample bottles, preventing loss from solution before analysis.

Ingrowth of ^{238}U progeny during sample holding time can account for much of the observed gross-beta activity. Thorium-234 and ^{234}Pa, which are radioactive progeny of ^{238}U, both can approach secular equilibrium with ^{238}U during the sample holding time (Table 2). Protactinium-234 is the progeny of ^{234}Th (Fig. 2A); therefore, ^{234}Pa activity depends upon ^{234}Th activity. Rapid approach of ^{234}Pa to equilibrium compared with that of ^{234}Th means that the activities of these two radionuclides will be nearly equal during beta counting. Most ^{234}Th and ^{234}Pa growing in an acidified sample probably remain in solution. Ingrowth of these progeny to secular equilibrium would produce beta activity equal to twice the alpha activity of ^{238}U. Thus, twice the alpha activity of ^{238}U represents the upper limit of beta activity that can be produced by ingrowth of these two beta-emitting progeny. The actual contribution to gross-beta measurements probably is less than twice the ^{238}U activity because the energies of the two beta particles emitted by ^{234}Th is low compared with the $^{90}Sr/^{90}Y$ standard (Table 1).

Uranium-238 activity is generally greater in Alluvial Basins and Colorado Plateau samples compared with samples from the Central Region and from both regions studied in New Jersey (Fig. 3). Uranium-238 activities in the Coastal Plain are lower than in the other 4 regions, with all samples containing < 1 pCi/L. Many samples contain sufficient ^{238}U to account for 50% or more of the gross-beta activity (Fig. 4), assuming complete ingrowth of the ^{238}U progeny, and counting efficiencies for the progeny equal to that of the $^{90}Sr/^{90}Y$ standard. Potassium-40 and ^{228}Ra generally account for less than 50% of the gross-beta activity in samples with gross-beta activities > 10 pCi/L, except for sampled ground water from the Coastal Plain (Fig. 4). Ingrowth of ^{238}U progeny (^{234}Th and ^{234}Pa) seems an important source of the measured gross-beta activity in samples from all of the regions, again with the exception of the Coastal Plain. Consequently, measured gross-beta activities suggest greater radionuclide activities than are present in ground water at the time of sampling.

Uranium-238 activities tend to correspond with

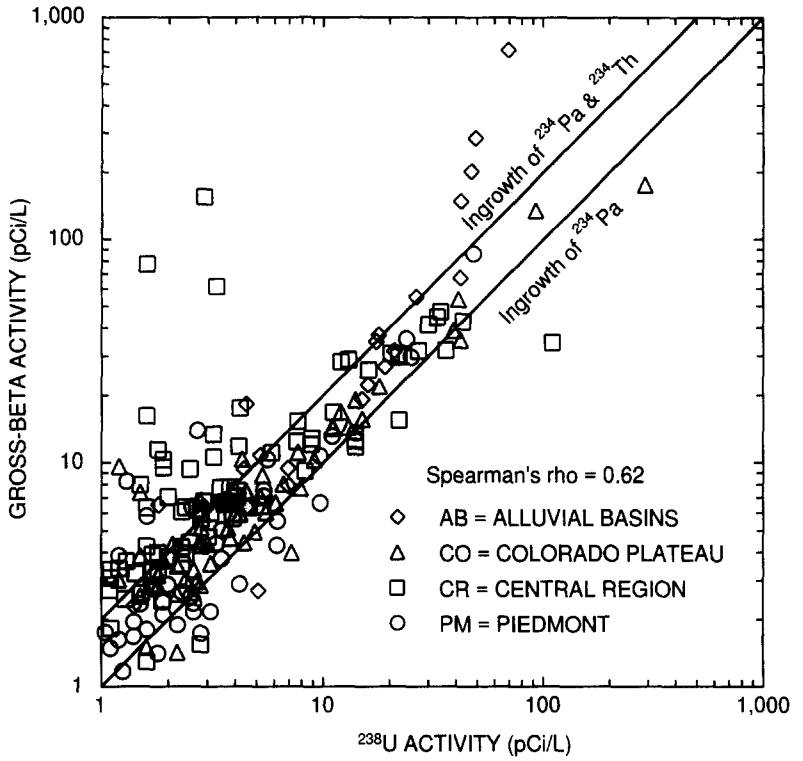

Fig. 7. Relation between ^{238}U and gross-beta activity corrected for ^{40}K activity. Sloping lines represent beta activity contributed by ^{234}Th and ^{234}Pa in secular equilibrium with ^{238}U. Data for 202 analyses shown. Spearman's rho = 0.79 (Iman and Conover, 1983) with a confidence level of >99%.

gross-beta activities, as indicated by a positive correlation (Spearman's rho = 0.79, Fig. 7) and a greater potential contribution from ^{238}U beta-emitting progeny at higher gross-beta activities (Fig. 4). These relations suggest that ^{238}U progeny are important sources of beta activity, particularly in ground water with gross-beta activity > 10 pCi/L.

Many of the corrected gross-beta activities are within the range estimated by assuming a contribution between 1 and 2 times the ^{238}U activity (Fig. 7). The upper line on Fig. 7 represents the upper limit for the beta contribution from both ^{234}Th and ^{234}Pa, the progeny of ^{238}U. The contribution from ^{228}Ra was not subtracted from the gross-beta activities plotted on Fig. 7 because of the small number of these analyses. Observed ^{228}Ra activities generally are low, most are <3 pCi/L, compared with many of the corrected gross-beta activities. Therefore, some of the samples with gross-beta activities < 10 pCi/L would plot closer to the 2 lines if the ^{228}Ra activities were subtracted from the gross-beta values. Samples plotting between the 2 lines may have been analyzed for gross-beta activity before complete ingrowth of ^{238}U progeny. Several samples contain gross-beta activities greater than can be accounted for by a combination ^{40}K and ^{238}U progeny. The cause of these anomalously high values is not known.

The maximum possible contribution of ^{238}U progeny to gross-beta activity has been estimated from sample holding times for a few samples from the Piedmont that were analyzed by the State of New Jersey Bureau of Environmental Laboratories. This maximum is based on the assumption that ^{234}Th and ^{234}Pa are both counted with the same efficiency as the $^{90}Sr/^{90}Y$ standard. Most of the estimated values are equal to or less than the adjusted gross-beta activity (Fig. 8), where the adjusted gross-beta activity is defined as:

adjusted gross-beta activity = measured gross-beta activity − B(^{40}K) − B(^{228}Ra),

where the B-values represent the beta activity of the indicated radionuclide.

The assumption that the counting efficiencies for ^{234}Th and the $^{90}Sr/^{90}Y$ standard are equal results in an overestimate of the probable real contribution of ^{238}U progeny to the measured gross beta. The assumption that samples are held long enough for complete ingrowth of ^{234}Th and ^{234}Pa, which is implied in the values shown on Fig. 4, also overestimates the contribution in some samples. Despite these considerations, ^{238}U progeny appear to be a likely source of gross-beta activity that can not be explained by a combination of ^{40}K plus ^{228}Ra.

Decay of ^{220}Rn and ^{222}Rn produce beta-emitting radionuclides of Pb and Bi (Fig. 2). Assuming all ^{222}Rn in a sample decays before degassing, a ^{210}Pb activity of ~0.0005 times the initial ^{222}Rn activity will

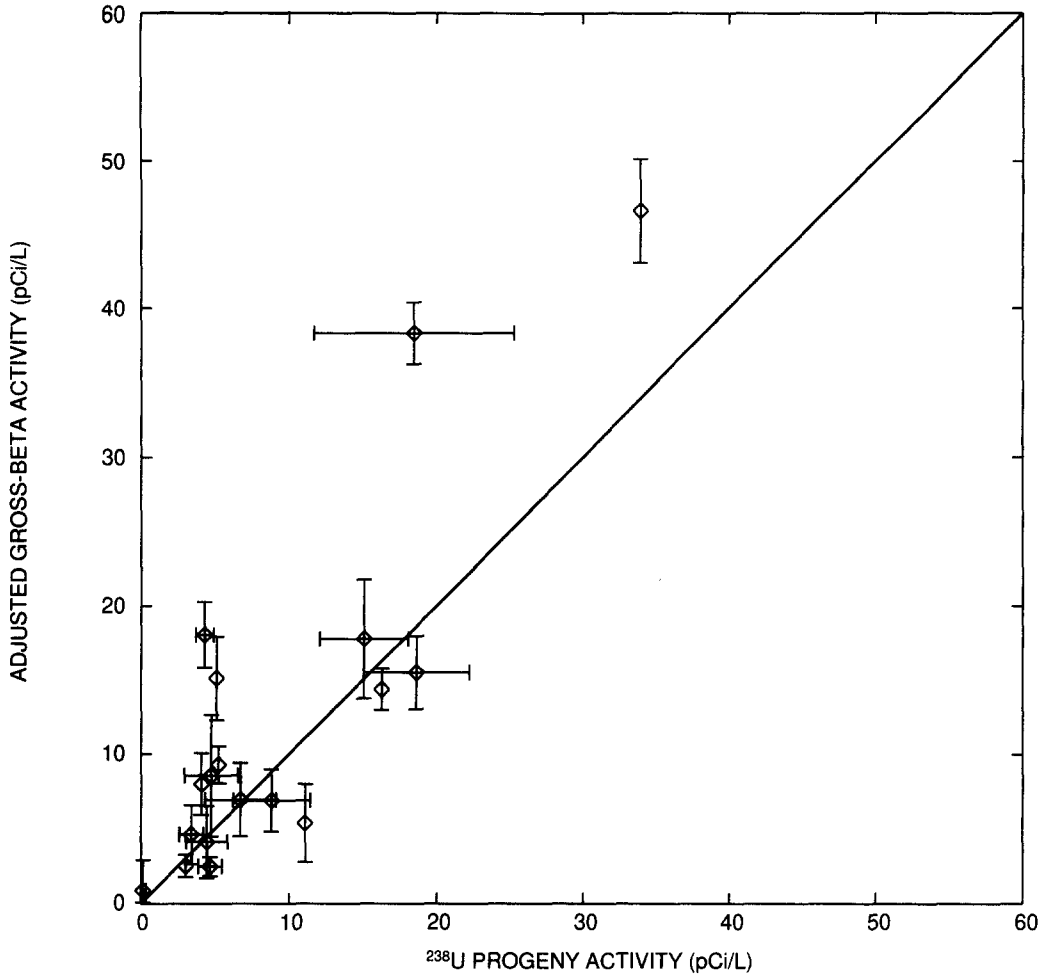

Fig. 8. Relation between gross-beta activity and the estimated contribution of beta-emitting progeny of ^{238}U. Straight lines through data points represent counting uncertainties for ^{238}U, gross-beta activity, ^{228}Ra, and analytical uncertainty for K. Sloping line represents estimated contribution from two ^{238}U progeny. Calculation of adjustment to gross-beta activity discussed in text.

be formed after about 40 days (Oural *et al.*, 1988). This conversion factor provides an estimate of the upper limit that can be produced by ^{222}Rn. Although most ^{222}Rn is undoubtedly lost from a sample due to degassing before radioactive decay, work by Krishnaswami *et al.* (1982) shows that some ^{210}Pb can grow into water samples after collection. However, activity supplied by ^{210}Pb ingrowth after sample collection is expected to be a few pCi/L or less for the systems being discussed. The median ^{222}Rn activity for the ground water under consideration here is <5000 pCi/L and corresponds to a ^{210}Pb activity of ~ 2.5 pCi/L. If about one-half the ^{222}Rn is lost to the atmosphere before decay, then ~ 1 pCi/L of beta activity would be contributed by ^{210}Pb ingrowth. Activities of ^{210}Bi would be essentially the same as ^{210}Pb due to the relation between the half-lives of these two radionuclides (Fig. 2A). The relatively nonenergetic beta particles emitted by ^{210}Pb and ^{210}Bi (Table 1) are not efficiently counted.

Lead-212 and ^{212}Bi, produced by the decay of ^{220}Rn, are unlikely sources of substantial beta activity produced after sampling. Assuming an initial ^{220}Rn activity of 1000 pCi/L, a maximum ^{210}Pb activity of about 1.4 pCi/L is reached after about 8 min. During this 8-min period, the ^{220}Rn decay would be nearly complete and therefore no longer available as a source of additional ^{210}Pb. The relatively short half-lives of ^{212}Pb and ^{212}Bi (10.64 h and 60.6 min, respectively, Fig. 2B) would cause relatively rapid further reduction of beta activity from these 2 radionuclides.

CONCLUSIONS

Gross-beta activity has been used as an indicator of beta-emitting isotopes in water since at least the early 1950s. Originally designed for detection of radioactive releases from nuclear facilities and weapons tests, gross-beta activity is widely used in studies of naturally occurring radioactivity in ground water. Analyses of about 800 samples from 5 ground-water

regions of the United States provide a basis for evaluating the utility of this measurement. The assembled data suggest that measured gross-beta activities are due to (1) long-lived radionuclides in ground water, and (2) ingrowth of beta-emitting radionuclides during holding times.

Potassium-40 and ^{228}Ra appear to be the primary sources of beta activity in ground water. The sum of ^{40}K plus ^{228}Ra appears to be less than the measured gross-beta activity in most ground-water samples. The difference between the contribution from these radionuclides and gross beta is most pronounced in ground water with gross-beta activities > 10 pCi/L. Potassium-40 and ^{228}Ra generally contribute most of beta activity in ground water with gross-beta activities < 1 pCi/L. In contrast, these two radionuclides account for less than one-half of the measured gross-beta activity in ground water having gross-beta activities > 10 pCi/L. One exception is ground water from the Coastal Plain of New Jersey, where ^{40}K plus ^{228}Ra generally contributes most of the gross-beta activity.

The gross-beta technique does not measure all beta activity in ground water. Although ^{3}H contributes beta activity to some ground water, it is driven from the sample before counting and therefore is not detected by gross-beta measurements. Beta-emitting radionuclides with half-lives shorter than a few days can decay to low values between sampling and counting. Therefore, only longer-lived radionuclides measured by the gross-beta technique represent the activities in ground water. Although little is known about concentrations of most short-lived beta-emitting radionuclides in environmental ground water, their activities are expected generally to be low.

Ingrowth of beta-emitting radionuclides during sample holding times can contribute to gross-beta activity, particularly in ground water with gross-beta activities > 10 pCi/L. Ingrowth of beta-emitting progeny of ^{238}U, specifically ^{234}Pa and ^{234}Th, contributes much of the measured gross-beta activity in ground water from 4 of 5 areas studied. Consequently, gross-beta activity measurements commonly overestimate the abundance of beta-emitting radionuclides actually present in ground water. Differing sample holding times before analysis leads to differing amounts of ingrowth of the two progeny. Therefore, holding times can affect observed gross-beta measurements, particularly in ground water with ^{238}U activities that are moderate to high compared to the activity of ^{40}K plus ^{228}Ra. Uncertainties associated with counting efficiencies of beta particles with different energies further complicate the interpretation of gross-beta measurements.

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MEMORANDUM

TO: [redacted] ~ USACE, LRB
FROM: [redacted] ~ SAIC
SUBJECT: **POTENTIAL CESIUM-137 CONCENTRATIONS IN THE IWCS**
DATE: September 22, 2003
CC: [redacted]

During our September 12, 2003 conference call on potential COCs in the IWCS, we discussed cesium contamination and how we might arrive at a reasonable concentration in the soils within the IWCS. We conducted a quick review of the historical documents that we have and offer the following as a potential "cesium source term" in the IWCS for modeling purposes.

- 29,000 cubic yards at ~8 pCi/g – placed in the northern section of the IWCS

The above is based on the following:

- The 1980 "Comprehensive Radiological Characterization of the DOE-Niagara Falls Storage Site" identifies the primary radiological contaminant in the northwest portion of the site as being cesium-137. They cite data for that area of Cesium activities up to 59,000 pCi/g although they do indicate that the cesium was swamping the detectors and not allowing detection of other radionuclides. The earlier 1979 progress report (Battelle 1979) indicates that two locations had Cs-137 contamination ranging from1,200 pCi/g to 208,000 pCi/g depending on the depth and replicate number. According to the Battelle Hazard Assessment, the volume of the contaminated soil is only 2 yd^3. However documents from the subsequent years consistently show a large area of contamination in the 'Baker-Smith' area. It would appear as if there may have been as much as several thousand cubic yards affected by Cs-137.

- The 1984 "Comprehensive Radiological Survey Off-Site Property G Niagara Falls Storage Site Lewiston, New York" identifies cesium as being one of the contaminants of property G. The levels at property G are much lower than those in the Baker Smith area (Northwest) with maximum activities around 3-5 pCi/g and most being below 1 pCi/g. However they did discover one 'spark-gap' with a high cesium content (27.4 μCi of Cs-137). The Property G soils would then appear to be essentially at background from a total volume perspective.

- In addition to the northwest quadrant of the site, Cs-137 was found in the northeast quadrant in the "New Naval Dump" area (between O and N streets east of Campbell St). Only 3 samples in this area exceeded background and a maximum detection of 220 pCi/g was found. The volume of soil material removed from this area is about 4,000 yd^3 (ANL June 1983 action memorandum). Because only three detections of

cesium were identified, SAIC assumed the Cs-137 concentration in much of it would be at or below 1 pCi/g.

- In the southwest quadrant (South of O St and West of Campbell) Cs-137 activities up to an order of magnitude above background were found. This would put the levels at about 3 pCi/g. These levels were encountered near the central ditch and along R St. east of the central ditch. Roughly 35,000 yd^3 of material was removed from both the central ditch and west ditch in 1983. Assuming that 60% (the proportion of the length of ditch remediated) of that was from the Central Ditch which did contain cesium, 21,000 yd^3 of soil with cesium was moved to the IWCF.

- No reported analyses for the southeast quadrant are above background for Cs-137.

Assuming that the Baker-Smith volume (~4000 yd^3 based on comparisons of the area excavated with the New Naval Dump) averaged 40 pCi/g, that the New Naval Dump soils were at 1 pCi/g average and that the central ditch soils were at 3 pCi/g would give a roughly 29,000 yd^3 of soil averaging 8 pCi/g Cs-137. A further assumption could be made that because of the timeframes of most of these relocations the material was placed in the northern portion of the IWCF.

The table below summarizes the contributions to the cesium source term.

Area Remediated	**Cs-137 Concentration (pCi/g)**	**Volume (yd^3)**
Baker Smith	40	4,000
New Naval Dump	1	4,000
Central Ditch	3	21,000
Resulting average in IWCS		
Northern Section of the IWCS	~8	29,000

References:

ANL, 1983, *Action Description Memorandum, Niagara Falls Storage Site Proposed Interim Remedial Actions for FY 1983-85 Accelerated Program*, Argonne National Lab, June.

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Table 9-1. NFSS Fall 2008 Environmental Surveillance Program Findings for Radiological Constituents in Groundwater

NFSS Well ID*	CAS Number	Analyte	Result	Units	Qualifier*
201A	13981-16-3	Plutonium-238	-0.014	pCi/L	U
201A	OER-100-70	Plutonium-239/240	-0.014	pCi/L	U
201A	14133-76-7	Technetium-99	8.570	pCi/L	U
201A	10045-97-3	Cesium-137	0.106	pCi/L	U
201A	10098-97-2	Strontium-90	0.053	pCi/L	U
201A	10028-17-8	Tritium	-43.900	pCi/L	U
BH49A	13981-16-3	Plutonium-238	-0.067	pCi/L	U
BH49A	OER-100-70	Plutonium-239/240	-0.040	pCi/L	U
BH49A	14133-76-7	Technetium-99	3.040	pCi/L	U
BH49A	10045-97-3	Cesium-137	1.430	pCi/L	U
BH49A	10098-97-2	Strontium-90	0.025	pCi/L	U
BH49A	10028-17-8	Tritium	59.800	pCi/L	U
BH49A		Uranium-233/234	10.900	pCi/L	
BH49A	13982-70-2	Uranium-235/236	0.562	pCi/L	
BH49A	7440-61-1	Uranium-238	9.860	pCi/L	
OW11B	13981-16-3	Plutonium-238	-0.013	pCi/L	U
OW11B	OER-100-70	Plutonium-239/240	0.000	pCi/L	U
OW11B	14133-76-7	Technetium-99	-6.720	pCi/L	U
OW11B	10045-97-3	Cesium-137	0.075	pCi/L	U
OW11B	10098-97-2	Strontium-90	-0.008	pCi/L	U
OW11B	10028-17-8	Tritium	164.000	pCi/L	U
OW11B		Uranium-233/234	87.600	pCi/L	
OW11B	13982-70-2	Uranium-235/236	4.270	pCi/L	
OW11B	7440-61-1	Uranium-238	84.100	pCi/L	

U - Compound was analyzed for but not detected.

Table 9-2. NFSS Fall 2009 Environmental Surveillance Program Findings for Radiological Constituents in Groundwater

NFSS Well ID*	CAS Number	Analyte	Result	Units	Qualifier
201A	10045-97-3	Cesium-137	1.406	pCi/L	U
201A	13981-16-3	Plutonium-238	0.01513	pCi/L	U
201A	15117-48-3	Plutonium-239	0.02561	pCi/L	U
201A	14158-27-1	Strontium-89	-0.88473	pCi/L	U
201A	10098-97-2	Strontium-90	1.839368	pCi/L	U
201A		Total Strontium	1.242982	pCi/L	U
BH49A	10045-97-3	Cesium-137	-0.5934	pCi/L	U
BH49A	13981-16-3	Plutonium-238	-0.01777	pCi/L	U
BH49A	15117-48-3	Plutonium-239	-0.00863	pCi/L	U
BH49A	14158-27-1	Strontium-89	0.552565	pCi/L	U
BH49A	10098-97-2	Strontium-90	1.036426	pCi/L	U
BH49A		Total Strontium	1.40846	pCi/L	U
BH49A	13966-29-5	Uranium-234	7.665	pCi/L	
BH49A	13982-70-2	Uranium-235	0.6051	pCi/L	
BH49A	7440-61-1	Uranium-238	6.586	pCi/L	
BH49A		Total Uranium	14.8561	pCi/L	
OW11B	10045-97-3	Cesium-137	-0.1617	pCi/L	U
OW11B	13981-16-3	Plutonium-238	0.01221	pCi/L	U
OW11B	15117-48-3	Plutonium-239	0.02722	pCi/L	U
OW11B	14158-27-1	Strontium-89	-0.67835	pCi/L	U
OW11B	10098-97-2	Strontium-90	1.465329	pCi/L	U
OW11B		Total Strontium	1.008735	pCi/L	U
OW11B	13966-29-5	Uranium-234	137.9	pCi/L	
OW11B	13982-70-2	Uranium-235	12.4	pCi/L	
OW11B	7440-61-1	Uranium-238	123.7	pCi/L	
OW11B		Total Uranium	274	pCi/L	

U - Compound was analyzed for but not detected.

Table 4.2
Summary of Contaminants of Potential Concern for Simulation

			Medium	Fraction	Parameter	Units	Screening Level	UTL[1]	MCL[2]
U-238	1	^{238}U	GW	RAD	Uranium-238, Dissolved	pCi/L	6.32	6.32	30 µg/L [(3)]
	2	^{234}U	GW	RAD	Uranium-234, Dissolved	pCi/L	8.94	8.94	
	3	^{230}Th	GW	RAD	Thorium-230, Dissolved	pCi/L	0.229	0.229	
	4	^{226}Ra	GW	RAD	Radium-226, Dissolved	pCi/L	1.31	1.31	5 [(4)]
	5	^{210}Pb	GW	RAD	Lead-210, Dissolved	pCi/L	NA		
Actinium	1	^{235}U	GW	RAD	Uranium-235, Dissolved	pCi/L	0.51	0.51	
	2	^{231}Pa	GW	RAD	Protactinium-231, Dissolved	pCi/L	NA		
	3	^{227}Ac	GW	RAD	Actinium-227, Dissolved	pCi/L	NA		
Thorium	1	^{232}Th	GW	RAD	Thorium-232, Dissolved	pCi/L	0.39	0.39	
Metals	1	Arsenic	GW	METAL	Arsenic, Dissolved	ug/L	10	10	10
	2	Barium	GW	METAL	Barium, Dissolved	ug/L	42.8	42.8	2,000
	3	Boron	GW	METAL	Boron, Dissolved	ug/L	4750	4750	
	4	Cadmium	GW	METAL	Cadmium, Dissolved	ug/L	2.32	2.32	5
	5	Iron	GW	METAL	Iron, Dissolved	ug/L	9280	9280	
	6	Lead	GW	METAL	Lead, Dissolved	ug/L	0.935	0.935	15
	7	Molybdenum	GW	METAL	Molybdenum, Dissolved	ug/L	40(5)		40[(5)]
	8	Manganese	GW	METAL	Manganese, Dissolved	ug/L	966	966	
PCE-TCE-DCE-VC	1	PCE	GW	VOA	Tetrachloroethene	ug/L	5		5
	2	TCE	GW	VOA	Trichloroethene	ug/L	5		5
	3	cis-DCE	GW	VOA	cis-1,2-Dichloroethene	ug/L	70		70
	4	VC	GW	VOA	Vinyl chloride	ug/L	1.48	1.48	2

Table 4.2
Summary of Contaminants of Potential Concern for Simulation

			Medium	Fraction	Parameter	Units	Screening Level	UTL[1]	MCL[2]
Other	1	Antimony	GW	METAL	Antimony, Dissolved	ug/L	2.4	2.4	
	2	bis(2-eh)phthalate	GW	SVOA	bis(2-Ethylhexyl)phthalate	ug/L	6		6
	3	methylene chloride	GW	VOA	Methylene chloride	ug/L	5		5

total=24

[1] UTL - Upper Tolerance Limit for NFSS (SAIC and Tetra Tech, 2006)

[2] MCL - Maximum Contaminant Level (USEPA)

[3] The MCL of 30 µg/L is for Total Uranium

[4] The MCL is for combined ^{226}Ra and ^{228}Ra

[5] The USEPA drinking water standard lifetime health advisory level for a 10 kg child.