THE DECADE OF DEPLETED URANIUM

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An Update of the Quantitative Analysis of Uranium Isotopes in British, Canadian, and United States Gulf War Veterans

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The Objective of the Study

To determine the quantities and ratios of uranium isotopes in the urine and organs of the Gulf War Veterans exposed to depleted uranium (DU) by the inhalational route of internal contamination.

Patients, Materials, and Methods

Twenty-Seven British, Canadian, and United States veterans presenting with complex non-specific clinical symptomatology of the **Gulf War Illness** had their 24hrs urine samples quantitatively determined for ^{234}U, ^{235}U, ^{236}U, and ^{238}U by the method of mass spectometric analysis at Atlantic Universities Radiogenic Isotope Facility.

St. Johns, Newfoundland, Canada

Radiochemical Analysis

- The urine samples were collected and transported under controlled circumstances in sealed plastic vials, weighed into savillex-teflon screw-cap jars (15ml) and evaporated to dryness at 80-100 degrees C.
- All samples were repeatedly evaporated three times after the addition of 4ml of double distilled concentrated nitric acid.

- Each sample was separated into an isotopic concentration and isotopic dilution fraction, by adding 3.1N hydrochloric acid to each sample.

- Half of each sample was transferred to the savillex-teflon jar (7ml) & accurately weighed.

Mass Spectrometry

- Uranium was separated and collected in both fractions after ion exchange preparation with DOWEX analytical grade AGL-X8 ion exchange resins with a modified HBr technique.

- The isotopic composition was measured by a multi-collector Finnigan mass spectrometer using secondary electron multiplier (SEM) detector and ion counting system.

- The uranium blank control has been determined to by 0.45 picograms and 960U standard, measured by the same procedure.

Humans ingest or breathe in uranium. Sources are:

1) Drinking water which is filtered through the ground
2) Eating vegetables grown in our gardens
3) Breathing in dust and dirt on windy days

Hypothetical measurement of urine specimen:

$$^{238}U/^{235}U = \sim 137.88$$

Every person will have trace amounts of naturally occurring uranium in their system. We absorb it through the vegetable we eat and through the water we drink which is filtered through the soil and rocks that we live. We can also get small amounts by inhaling or ingesting dirt and dust blowing around in the breeze. So if I measured someone's urine specimen from my family, I would see this naturally occurring $^{238}U/^{235}U$ ratio of approximately 137.88.

Atlantic Universities Radiogenic Isotope Facility

(AURIF)

The Atlantic Universities Radiogenic Isotope Facility (AURIF) was created in 1989 to analyze geological samples for radiogenic isotopic tracers studies. Since its inception, AURIF has analysed samples for numerous scientists worldwide. The combined research experience of the scientific personnel in AURIF in dealing with the U/Pb isotopic system spans over 30 years with more than 5000 U/Pb analyses completed. Data from this lab has been published in peer- reviewed journals for more than 9 years.

Depleted Uranium : DU for short

"left-over" product during the enrichment process for nuclear fuel rods or nuclear weapons.

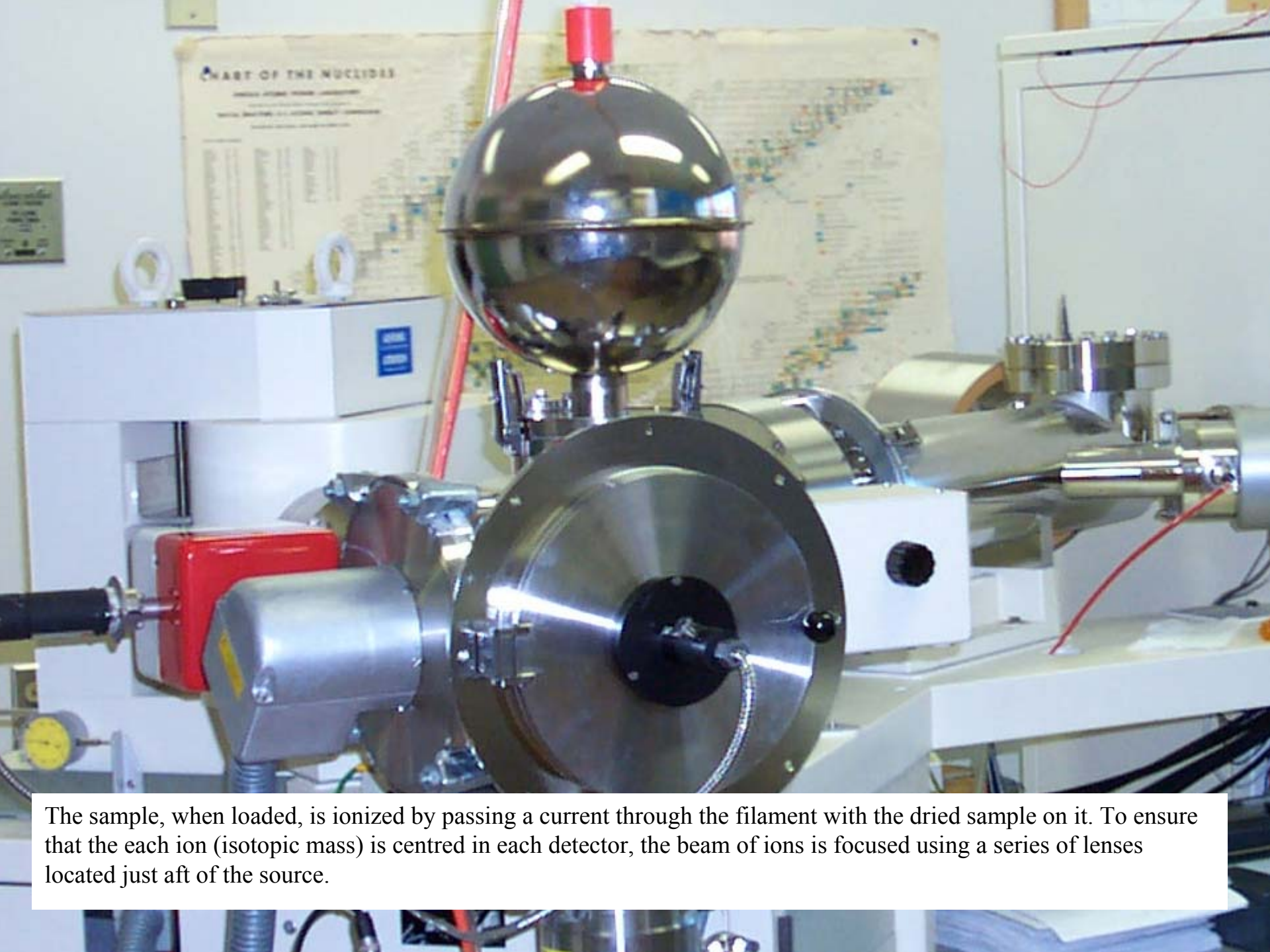
DU shrapnel measured in AURIF lab

$^{238}U = 99.7945\%$, $^{235}U = 0.2026\%$, $^{234}U = 0.0012\%$.

Key ratio of interest: $^{238}U/^{235}U = 491.87 \pm 0.16$ (2σ absolute)

The nuclear industry refines mined U_3O_8 and removes the majority of ^{235}U and ^{234}U for use in nuclear fuel rods. This enrichment process creates a "left over" form of uranium with substantially less ^{235}U and increased ^{238}U abundance as seen in this slide. This "left over" uranium is referred to as depleted uranium. The $^{238}U/^{235}U$ ratio measured from one particular piece of shrapnel is 491.87 ± 0.16 ($2\ \sigma$). I already mentioned one use of DU as ballast in airplanes manufactured before the mid –1980's. Another use of DU has been in shell casing for military armaments. The shells are very hard and will easily pierce armour. Upon impact, the DU in the shell casing ignites and burns causing massive damage.

AURIF uses a Finnigan MAT 262V solid source thermal ionization mass spectrometer with 8 faraday detectors and one secondary electron multiplier – ion-counting system.



The sample, when loaded, is ionized by passing a current through the filament with the dried sample on it. To ensure that the each ion (isotopic mass) is centred in each detector, the beam of ions is focused using a series of lenses located just aft of the source.

The mass spectrometer has a large electromagnet that facilitates the separation of charged particles or ions.

Illustration of mass separation with the Finnigan electromagnet

Courtesy of Finnigan MAT

As the beam of ions pass out through the “line of sight” or beam valve, it encounters a strong magnetic field created by the large electromagnet. Since the beam contains ions of different masses, it splits into individual beams, one per isotopic mass present in the sample. So for uranium, one beam enters the magnetic field and produces individual ^{238}U, ^{235}U and ^{234}U (and even sometimes ^{236}U if present in the sample).

Table 1: Quantitative Data for Individual Samples

No.	Patient	U 238	U 235	U238/U235	Sigma
1	G.B.	99.2782	0.7145	139.0	1.3
2	B.B	99.2742	0.7076	140.3	0.2
3	R.B.	99.2782	0.7145	139.0	1.3
4	L.B.	99.2738	0.7180	138.3	0.5
5	D.B.	99.2701	0.7233	137.5	0.5
6	P.C.	99.2570	0.7210	137.7	0.5
7	C.C.	99.2738	0.7113	139.6	0.4
8	R.G.D.	99.3154	0.6758	147.0	0.7
9	J.G.	99.7565	0.2339	426.6	3.7
10	W.H.	- - -	- - -	- - -	- - -
11	J.H.	- - -	- - -	153.0	0.1
12	M.K.	99.2762	0.7152	138.8	0.8
13	C.P.L.	99.2702	0.7200	137.9	0.5
14	G.L.	- - -	- - -	- - -	- - -
15	K.I.M.	99.4280	0.5663	175.6	1.7
16	T.N.	99.2963	0.6925	143.4	3.4
17	C.O.	99.2811	0.7135	139.1	0.9
18	A.P.	99.3456	0.6495	153.0	0.3
19	T.R.	99.5564	0.4346	229.1	1.3
20	P.R.	99.2742	0.7189	138.1	0.8
21	S.R.	99.5603	0.4304	231.3	1.6
22	F.S. (A)	99.4876	0.4945	201.2	5.9
23	F.S. (B)	99.2693	0.7189	138.1	1.7
24	V.S.	99.7113	0.2830	352.4	1.5
25	M.D.T	- - -	- - -	- - -	- - -
26	R.W.	99.3025	0.6825	145.5	1.4
27	A.W.	99.3862	0.4966	200.1	1.2

Table 2: Summary of Quantitative Data for Individual Samples

	U 238	U 235	U238/U235	Sigma
Negative	99.2726	0.7166	138.6	0.8
Std. Dev.	0.00625	0.0046	0.862	
Std. Error	0.00188	0.0014	0.260	
Positive	99.4561	0.5248	208.4	1.42
Std. Dev.	0.1598	0.1575	87.51	
Std. Error	0.0461	0.0455	24.27	
Totals	99.3683	0.6165	176.4	1.18
Std. Dev.	0.1469	0.1483	72.51	
Std. Error	0.0306	0.0309	14.80	
P-Value	0.00108	0.00072	0.00696	

Table 3: Quantitative Data for Positive Samples

No.	Patient	U 238	U 235	U238/U235	Sigma
3	R.B.	99.2782	0.7145	139.0	1.3
8	R.G.D.	99.3154	0.6758	147.0	0.7
9	J.G.	99.7565	0.2339	426.6	3.7
11	J.H.	- - -	- - -	153.0	0.1
15	K.I.M.	99.4280	0.5663	175.6	1.7
16	T.N.	99.2963	0.6925	143.4	3.4
18	A.P.	99.3456	0.6495	153.0	0.3
19	T.R.	99.5564	0.4346	229.1	1.3
21	S.R.	99.5603	0.4304	231.3	1.6
22	F.S. (A)	99.4876	0.4945	201.2	5.9
24	V.S.	99.7113	0.2830	352.4	1.5
26	R.W.	99.3025	0.6825	145.5	1.4
27	A.W.	99.3862	0.4966	200.1	1.2
Totals		99.4561	0.5248	208.4	1.42
Std. Dev.		0.1598	0.1575	87.51	
Std. Error		0.0461	0.0455	24.27	

Table 4: Quantitative Data for Negative Samples

No.	Patient	U 238	U 235	U238/U235	Sigma
1	G.B.	99.2782	0.7145	139.0	1.3
2	B.B	99.2742	0.7076	140.3	0.2
4	L.B.	99.2738	0.7180	138.3	0.5
5	D.B.	99.2701	0.7233	137.5	0.5
6	P.C.	99.2570	0.7210	137.7	0.5
7	C.C.	99.2738	0.7113	139.6	0.4
12	M.K.	99.2762	0.7152	138.8	0.8
13	C.P.L.	99.2702	0.7200	137.9	0.5
17	C.O.	99.2811	0.7135	139.1	0.9
20	P.R.	99.2742	0.7189	138.1	0.8
Totals		99.2726	0.7166	138.6	0.91
Std. Dev		0.00625	0.0046	0.862	
Std. Error		0.00188	0.0014	0.260	

Table 5: Ratio of Uranium Isotopes

	U 238	U 235	U238/U235	U235/U238
Natural Uranium	99.2739	0.7200	137.88	0.0073
Shrapnel (DU)	99.7945	0.2026	492.60	0.0020
Urine	99.3728	0.6119	178.1	0.0062

Table 6: Isotopic Ratio in Individual Samples

No.	Patient	235/238	Sigma	234/238	Sigma	236/238	Sigma
1	G.B.	0.007195	0.000034	0.000069	0.000005	0.000004	0.000003
2	B.B	0.007130	0.000090	0.000100	0.000002	0.000090	0.000020
3	R.B.	0.006628	0.000143	0.000080	0.000012	0.000072	0.000006
4	L.B.	0.007230	0.000037	0.000057	0.000003	0.000006	0.000002
5	D.B.	0.007280	0.000034	0.000065	0.000003	0.000011	0.000004
6	P.C.	0.007264	0.000018	0.000128	0.000006	0.000094	0.000013
7	C.C.	0.007170	0.000020	0.000080	0.000010	0.000070	0.000010
8	R.G.D.	0.006805	0.000032	0.000070	0.000007	0.000019	0.000006
9	J.G.	0.002345	0.000020	0.000035	0.000003	0.000059	0.000007
10	W.H.	- - -	- - -	- - -	- - -	- - -	- - -
11	J.H.	0.006535	0.000004	0.000066	0.000004	0.000092	0.000005
12	M.K.	0.007205	0.000041	0.000080	0.000004	0.000007	0.000003
13	C.P.L.	0.007255	0.000026	0.000075	0.000004	0.000023	0.000003
14	G.L.	- - -	- - -	- - -	- - -	- - -	- - -
15	K.I.M.	0.005696	0.000055	0.000041	0.000006	0.000026	0.000009
16	T.N.	0.006970	0.000175	0.000100	0.000044	0.000013	0.000009
17	C.O.	0.007188	0.000044	0.000052	0.000010	0.000003	0.000001
18	A.P.	0.006540	0.000010	0.000050	0.000002	0.000000	0.000000
19	T.R.	0.004366	0.000025	0.000032	0.000002	0.000058	0.000003
20	P.R.	0.007241	0.000039	0.000059	0.000005	0.000011	0.000003
21	S.R.	0.004323	0.000030	0.000031	0.000002	0.000063	0.000006
22	F.S. (A)	0.004981	0.000146	0.000046	0.000012	0.000123	0.000033
23	F.S. (B)	0.007242	0.000090	0.000080	0.000077	0.000047	0.000030
24	V.S.	0.002838	0.000012	0.000016	0.000001	0.000043	0.000002
25	M.D.T	- - -	- - -	- - -	- - -	- - -	- - -
26	R.W.	0.006870	0.000065	0.000116	0.000003	0.000037	0.000011
27	A.W.	0.004992	0.000016	0.000081	0.000007	0.000042	0.000010

Table 7: Summary of Isotopic Ratio for Individual Samples

	235/238	Sigma	234/238	Sigma	236/238	Sigma
Negative	0.007218	0.000049	0.000077	0.000007	0.000040	0.000013
Std. Dev.	0.000045		0.000022		0.000045	
Std. Error	0.000014		0.000007		0.000014	
Positive	0.005376	0.000072	0.000059	0.000013	0.000050	0.000008
Std. Dev.	0.001561		0.000030		0.000034	
Std. Error	0.000433		0.000008		0.000009	
Totals	0.006176	0.000061	0.000066	0.000011	0.000042	0.000008
Std. Dev.	0.001467		0.000027		0.000039	
Std. Error	0.000299		0.000006		0.000008	
P-Value	0.000564		0.060110		0.125050	

Table 8: Quantitative Data for ^{234}U and ^{236}U

No.	Patient	U 234	U 236
1	G.B.	0.0069	0.0008
2	B.B	0.0096	0.0085
3	R.B.	0.0079	0.0071
4	L.B.	0.0057	0.0006
5	D.B.	0.0065	0.0011
6	P.C.	0.0127	0.0094
7	C.C.	0.0077	0.0072
8	R.G.D.	0.0070	0.0019
9	J.G.	0.0037	0.0060
10	W.H.	- - -	- - -
11	J.H.	- - -	- - -
12	M.K.	0.0080	0.0007
13	C.P.L.	0.0075	0.0023
14	G.L.	- - -	- - -
15	K.I.M.	0.0041	0.0016
16	T.N.	0.0099	0.0013
17	C.O.	0.0051	0.0003
18	A.P.	0.0049	0.0000
19	T.R.	0.0032	0.0057
20	P.R.	0.0058	0.0011
21	S.R.	0.0031	0.0062
22	F.S. (A)	0.0046	0.0123
23	F.S. (B)	0.0076	0.0028
24	V.S.	0.0016	0.0043
25	M.D.T	- - -	- - -
26	R.W.	0.0115	0.0036
27	A.W.	0.0058	0.0045

Table 9: Summary of Quantitative Data for ^{234}U and ^{236}U

	U 234	U 236
Negative	0.0076	0.0032
Std. Dev.	0.0021	0.0035
Std. Error	0.0006	0.0010
Positive	0.0058	0.0045
Std. Dev.	0.0030	0.0033
Std. Error	0.0009	0.0010
Totals	0.0066	0.0039
Std. Dev.	0.0027	0.0034
Std. Error	0.0006	0.0007
P-Value	0.0682	0.1967

Table 10: Gravimetric Data for Individual Samples

No.	Patient	U pg/g	U pg/24hr
1	G.B.	5.01	10196.99
2	B.B	- - -	- - -
3	R.B.	- - -	- - -
4	L.B.	- - -	- - -
5	D.B.	- - -	- - -
6	P.C.	7.33	12149.63
7	C.C.	- - -	- - -
8	R.G.D.	13.07	1290.24
9	J.G.	- - -	- - -
10	W.H.	8.55	960.00
11	J.H.	- - -	- - -
12	M.K.	4.01	35.94
13	C.P.L.	0.20	545.44
14	G.L.	1.49	141.90
15	K.I.M.	2.77	14111.26
16	T.N.	- - -	- - -
17	C.O.	- - -	- - -
18	A.P.	- - -	- - -
19	T.R.	- - -	- - -
20	P.R.	15.21	7604.85
21	S.R.	77.96	268225.11
22	F.S. (A)	163.02	10780.19
23	F.S. (B)	162.49	10745.42
24	V.S.	- - -	- - -
25	M.D.T	0.0150	1.60
26	R.W.	- - -	- - -
27	A.W.	2217.04	11426.01

Table 11: Summary of Gravimetric Data for Individual Samples

	U pg/g	U pg/24hr
Negative	32.38	6879.71
Std. Dev.	63.94	5314.25
Std. Error	26.10	2169.53
Positive	494.77	75409.84
Std. Dev.	964.90	112434.73
Std. Error	431.51	50282.34
Totals	209.64	34057.00
Std. Dev.	585.90	71715.11
Std. Error	156.59	19166.67
P-Value	0.16031	0.11574

Table 12: Autopsy Specimens

	U 238	U 235	U238/U235
Lung	99.2348	0.6932	143.20
Liver	99.2792	0.7082	140.20
Bone	99.3220	0.6718	147.80

Table 13: Uranium Concentration in Man

	Daily Urinary Excretion (in mg)	**Estimated Body Content (in mg)**
UK	0.380	100
USA	0.154	80

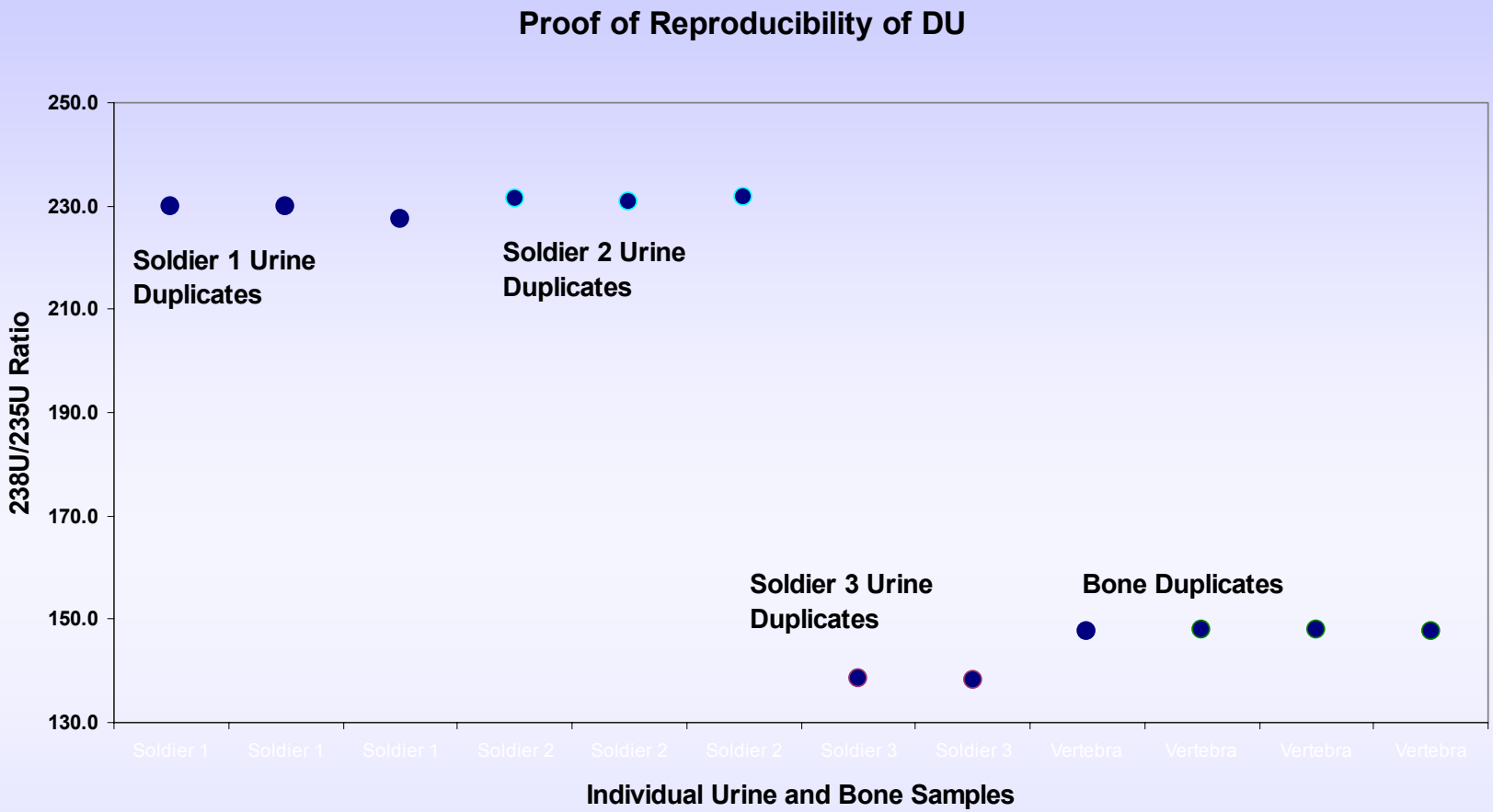
UK – Hamilton E.I.: Nature 227, 501-502. 1970

USA – Welford G.A., Baird R., Fisenne I.M.: Annual Bioassy and Analytical Chemistry Conf. Bethesda MD, 1970.

Results of autopsied bone fragments from deceased Canadian veteran

Sample#	238U/235U	2 sigma	234/238	2 sigma	236/238	2 sigma
Vertebra	147.6721	0.190	0.000057	0.0000360	0.000013	0.000002
Vertebra	147.8660	0.413	0.000052	0.0000005	0.000009	0.000002
Vertebra	148.0673	0.562	0.000052	0.0000007	0.000009	0.000001
Vertebra	147.7731	0.352	0.000051	0.0000014	0.000009	0.000002
Sample#	U238%	U235%	U234%	U236%		
Vertebra	99.3205%	0.6726%	0.0056%	0.0013%		
Vertebra	99.3222%	0.6717%	0.0051%	0.0009%		
Vertebra	99.3232%	0.6708%	0.0051%	0.0009%		
Vertebra	99.3219%	0.6721%	0.0051%	0.0009%		

This slide shows the results for the bone analyses from a deceased Canadian veteran. As you can see the bone sample shows a shifted 238U/235U ratio indicating the presence of DU.



This slide show the proof of reproducibility of DU analyses. Each one of these analyses are complete duplicates starting with a fresh aliquot of urine or a new bone fragment. The 2 sigma absolute errors have been plotted along with the sample but in most cases you can’t see them as they are so small.

Original Results of Urine Analysis

- DU present in 13/27 samples

$^{238}U > 99.45\%$

$^{235}U < 0.52\%$

- The average ratio

$^{238}U / {}^{235}U > 208.4$

- The results confirm the definitive presence of

$^{234}U > 0.0066\%$

and

$^{236}U > 0.0039\%$

Conclusion

The results demonstrate a significant presence of DU in the urine of Gulf War Veterans nine years after inhalational exposure and warrants further investigation.

When a book hits the head and a hollow sound is heard, it is not always the fault of the book.

Schopenhauer