AN EVALUATION OF EVIDENCE FOR SUPERHEAVY ELEMENTS IN NATURE,

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The Dubna group has recently reported the results of neutron counting experiments on several chondritic meteorites and a terrestrial hot springs sample in which they find events of high neutron multiplicity which they attribute to the decay of superheavy elements (1, 2). They further report the enrichment of this unknown element in a volatile fraction extracted from Allende (3). The discovery of a decaying superheavy element could provide a new source for heating of planetary bodies in the early solar system, and would place further constraints on the astrophysical processes to which material was subjected before the formation of the solar system.

The Dubna group's counting data for Allende, Saratov, Efremovka, and the mineral fraction extracted from a sulphur rich hot spring by an anion exchange column are reproduced in Table 1. They estimate the neutron multiplicity ($\bar{\nu}$) for the meteorite at between 3.5 and 10 (1,2,4), and for the hot springs extract at about 4.5 (1). Because of the rough linear increase of $\bar{\nu}$ with mass of the fissioning element (5), a $\bar{\nu} > 3.8$ would imply a fissioning element heavier than Cf^{252}.

We have systematically examined the counting data in Table 1, using a technique which takes full account of the statistical weight of data in each channel, as well as the available experimental data on known spontaneously fissioning elements. The number of n-neutron events detected ($\underline{X}_n$) for each detector efficiency (ε) is related to the number of i-neutron events occurring (x_i) as follows:

$$\underline{X}_n = \sum_{i=n}^{\infty} x_i \; C\binom{i}{n} \; \varepsilon^n (1-\varepsilon)^{i-n} \quad (n=o,\infty)$$

where $C\binom{i}{n}$ is the binomial coefficient. Using the data for n=0,5 for Pu^{236} and n=0,7 for Cf^{252}, cubic polynomial fits to log ($\underline{X}_n$) were obtained for each ε. The polynomial coefficients for each ε were generalized by linear interpolation between the Pu^{236} and Cf^{252} values, using a parameter "m". Thus for each efficiency, a set of eight values: b_{kj}, k=0,3, j=0,1, were obtained such that:

$$a_k = b_{ko} + mb_{k1}, \quad \text{and } \log(\underline{X}_n) \simeq \sum_{k=o}^{3} a_k h^k$$

The b_{kj} values describe a family of curves $\underline{X}_n$ as a function of the fitting parameter, m, for each efficiency examined.

We then determined the best fit value of m (denoted m*) for each set of neutron counting data for isotopes of Pu, Cm, and Cf (5) and for the Dubna counting data in Table 1. Finally we did a linear regression between m* and atomic mass (A) and $\bar{\nu}$ for the known fissioning isotopes. There is a very strong correlation ($r^2 = .96$) between the value of m* and the $\bar{\nu}$ for the isotopes of U, Pu, Cm, and Cf (Figure 1). There is also a correlation ($r^2 = .75$) between m* and the atomic mass of the fissioning isotope.

By optimizing the fitting parameter, m, for the Dubna data on Allende, Saratov, Efremovka and the hot springs extract we obtain the values denoted by the arrows in Figure 1. The values of $\bar{\nu}$ are within the range of 2.5 to 4.6, not the higher values reported by the Dubna group. If we average all the meteorite data to provide better counting statistics we obtain, to 50%

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confidence limits:
meteorites: $m^* = 246.4 \pm 3.6$, $\bar{\nu} = 3.29 \pm 0.33$, $A = 247.2 \pm 3.1$
hot springs: $m^* = 246.9 \pm 1.4$, $\bar{\nu} = 3.34 \pm 0.16$, $A = 247.5 \pm 2.1$

The Dubna data is thus consistent with the decay of an actinide of mass 244 to 250, rather than a superheavy with a high ($\gtrsim 3.8$) $\bar{\nu}$. But how would an actinide find its way into the hot springs extract and several carbonaceous chondrites?

In the case of the hot springs extract there are three possible sources of actinides: 1) thermonuclear bomb production, 2) nuclear reactor waste, and 3) laboratory contamination. We have attempted to estimate the quantity of bomb produced actinides which might be present. The hot spring is in the 40° to 50° N. latitude band, where there is 2.2 miCi/km^2 activity from Pu^{239} plus Pu^{240} bomb fallout (6). This corresponds to 3.75×10^{19} atoms of Pu^{239}/km^2. Using the yields of heavier actinides relative to Pu^{239}, as determined in the Mike thermonuclear test (7), we calculate the fission rate/km^2 for each bomb produced actinide (see Table 2). The total fission activity is 6.4×10^5 dis/day-km^2, with 93% of this activity equally divided among Cm^{246}, Cm^{250}, and Pu^{240}. The bomb produced actinides deposited in a region 4.5 meters on a side would be sufficient to account for the hot springs counting results.

Presumably these actinides would be dissolved in rainfall, carried into the groundwater, and extracted by the anion exchange column. Dahlman et al report that 12% of the Pu and 50% of the Cm in an actinide release at Oak Ridge is water soluble (8), and that almost all of the water soluble Pu and Cm is extracted by an anion exchange column (8).

If we assume that 50% of the surface actinides have been dissolved in the rainfall over the last 20 years near the hot springs site, rainfall ≈ 0" to 10" per year (9), the groundwater fission rate would be 1.3×10^{-7} dis/day-gm of water. If the Dubna group sampled ≈ 10^8 grams (2.6×10^4 gallons) of this "model water" they would concentrate sufficient actinides to explain their counting results. Their paper (1) does not indicate the amount of water sampled. However, we note the two Cm isotopes would be the most water soluble, and their masses, 246 and 250, bracket our mass determination ($A = 247.5 \pm 2.1$) for the hot spring extract.

Nuclear reactors also produce large quantities of actinides (227 gm of Cm^{244} per metric ton of Pu fuel per 100 mw-years is typical)(9). However, estimation of the local effects is impossible without detailed knowledge of Russian waste disposal techniques and sites.

The meteorite data is more puzzling. Our calculations indicate that several sources of transuranics including: 1) the slowing down of primary cosmic ray actinides, 2) production by heavy ion collisions, 3) production by multiple neutron capture on U and Th, and 4) production in giant flare events can all be ruled out. In situ fission of heavy elements due to cosmic rays can also be excluded. We do note, however, that Zavara et al claim to have extracted most of the "superheavy" in their volatilization experiment. However, when the correct neutron multiplicity ($\bar{\nu} \simeq 3.3$) is taken into account, they extracted no more than 20% of the fissioning material in Allende. Furthermore, the fission rate of the volatile extract given by Zavara et al, 0.02 decays/kg-day (3), is the fission rate that would be expected from the spontaneous fission of U^{238} in Allende. (The neutron counting data, however, cannot be explained by U^{238} decay.) We also note that the Dubna group ran Cf^{252} in their neutron counter,

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and contamination by 5 x 10^4 atoms of Cf^{252} in their apparatus would account for their meteorite count rate. Their blank and Pb counting data, however, give null results; and the source of the multiple neutron events in the meteorites remains a mystery.

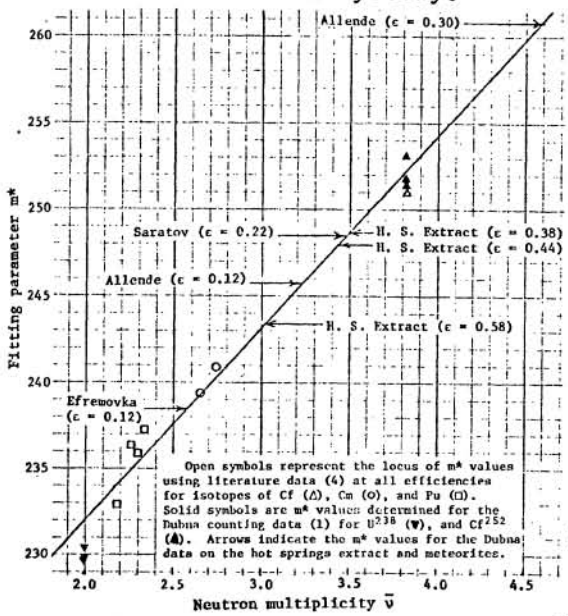

Figure 1: Fitting parameter m* vs. $\bar{\nu}$.

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TABLE 1

DUBNA GROUP NEUTRON COUNTING DATA (1)

Sample	Weight	Efficiency (ε)	Counting Time	Number of Events with Multiplicity n				
				n=2	n=3	n=4	n=5	n=6
Allende	3.9 kg	22%	40 days	3	0	0	0	0
Allende	22.5 kg	12%	55 days	10	1	0	0	0
Allende	10.5 kg	30%	45 days	5	2	1	0	0
Saratov	5.2 kg	22%	94 days	4	1	0	0	0
Efremovka	11.7 kg	12%	106 days	14	1	0	0	0
Hot	6 kg	38%	10 days	36	15	1	0	1
Springs	6 kg	44%	7 days	24	11	3	0	0
Extract	6 kg	58%	6 days	23	10	3	1	0
Background	0	22 and 30%	250 days	0	0	0	0	0
Lead	150 kg	30%	5 days	0	0	0	0	0
Cf^{252}	-	38%		1000	420	105	17	2
U^{238}	-	38%		1000	159	11	0	0

TABLE 2

FISSION DENSITY FROM BOMB ACTINIDES, 40° to 50° N. LATITUDE, 20 YEARS AFTER BOMB TEST

Isotope	Concentration	Fission Density	% of Total Fission
Cm^{246}	1.8 x 10^5 atoms/km^2	2.0 x 10^5 dis/day-km^2	31%
Pu^{240}	1.4 x 10^{19} atoms/km^2	2.0 x 10^5 dis/day-km^2	31%
Cm^{250}	1.9 x 10^{12} atoms/km^2	2.0 x 10^5 dis/day-km^2	31%
Pu^{242}	7.1 x 10^{17} atoms/km^2	1.9 x 10^4 dis/day-km^2	3%
Cm^{248}	4.5 x 10^{13} atoms/km^2	1.9 x 10^4 dis/day-km^2	3%
Others		<8 x 10^3 dis/day-km^2	< 2%
	Total	6.4 x 10^5 dis/day-km^2	