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 (ν)
for the meteorite at between 3.5 and 10 (1,2,4), and for the hot springs ex-
tract at about 4.5 (1). Because of the rough linear increase of ν with mass
of the fissioning element (5), a ν > 3.8 would imply a fissioning element
heavier than Cf

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 (Xn) for each
detector efficiency (ε) is related to the number of i-neutron events occurring
(Xi) as follows:

\[ X_n = \sum_{i=n}^{\infty} X_i \binom{i}{n} ε^n (1-ε)^{i-n} (n=0,∞) \]

where C(i) is the binomial coefficient. Using the data for n=0,5 for Pu
and n=0,9 for Cf
, cubic polynomial fits to log (Xn) were obtained for each
ε. The polynomial coefficients for each ε were generalized by linear inter-
polation between the Pu and Cf
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:

\[ a_k = b_ko + mb_k1, \text{ and } \log (X_n) = \frac{3}{k=0} a_k h^k \]

The b_kj values describe a family of curves Xn 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 ν for the known fissioning isotopes. There is a very
strong correlation (r^2 = .96) between the value of m* and the ν for the iso-
topes 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 ν 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: \( \mu^* = 246.4 \pm 3.6, \nu = 3.29 \pm 0.33, A = 247.2 \pm 3.1 \)
hot springs: \( \mu^* = 246.9 \pm 1.4, \nu = 3.34 \pm 0.16, A = 247.5 \pm 2.1 \)
The Dubna data is thus consistent with the decay of an actinide of mass 244 to 250, rather than a superheavy with a high (\( > 3.8 \)) \( \nu \). 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 mcI/km² activity from Pu²³⁹ plus Pu²⁴⁰ bomb fallout (6). This corresponds to \( 3.75 \times 10^{19} \) atoms of Pu²³⁹/km². Using the yields of heavier actinides relative to Pu²³⁹, as determined in the Mike thermonuclear test (7), we calculate the fission rate/km² for each bomb produced actinide (see Table 2). The total fission activity is \( 6.4 \times 10^5 \) dis/day-km², with 93% of this activity equally divided among Cm²⁴⁶, Cm²⁵⁰, and Pu²⁴⁰. 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 \( 1.3 \times 10^7 \) dis/day-gm of water. If the Dubna group sampled \( \approx 10^8 \) grams (2.6 \( \times 10^8 \) 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 Cm²⁴⁴ 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 (\( \nu = 3.3 \)) 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 U²³⁸ in Allende. (The neutron counting data, however, cannot be explained by U²³⁸ decay.) We also note that the Dubna group ran Cf²⁵² in their neutron counter,
and contamination by $5 \times 10^4$ 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.

REFERENCES

(5) (3)
(10) Holden, N. and Walker, F. (1972) Chart of the Nuclides, USABC.

Figure 1: Fitting parameter $m^\#$ vs. $\bar{v}$.

TABLE 1

DUNNA GROUP NEUTRON COUNTING DATA (1)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Weight</th>
<th>Efficiency (e)</th>
<th>Counting Time</th>
<th>Number of Events with Multiplicity n</th>
</tr>
</thead>
<tbody>
<tr>
<td>Allende</td>
<td>3.9 kg</td>
<td>22%</td>
<td>40 days</td>
<td>n=2 n=3 n=4 n=5 n=6</td>
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<tr>
<td>Allende</td>
<td>22.5 kg</td>
<td>12%</td>
<td>55 days</td>
<td>0 0 0 0 0</td>
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<tr>
<td>Allende</td>
<td>10.5 kg</td>
<td>30%</td>
<td>45 days</td>
<td>5 2 1 0 0</td>
</tr>
<tr>
<td>Saratov</td>
<td>5.2 kg</td>
<td>22%</td>
<td>96 days</td>
<td>5 1 0 0 0</td>
</tr>
<tr>
<td>Efremovka</td>
<td>11.7 kg</td>
<td>12%</td>
<td>106 days</td>
<td>1 1 0 0 0</td>
</tr>
<tr>
<td>Hot Springs</td>
<td>6 kg</td>
<td>36%</td>
<td>250 days</td>
<td>0 0 0 0 0</td>
</tr>
<tr>
<td>Extract</td>
<td>6 kg</td>
<td>36%</td>
<td>250 days</td>
<td>0 0 0 0 0</td>
</tr>
<tr>
<td>Background</td>
<td>0 kg</td>
<td>22 and 30%</td>
<td>250 days</td>
<td>0 0 0 0 0</td>
</tr>
<tr>
<td>Cf$^{252}$</td>
<td>150 kg</td>
<td>38%</td>
<td>5 days</td>
<td>0 0 0 0 0</td>
</tr>
<tr>
<td>U$^{238}$</td>
<td>-</td>
<td>38%</td>
<td>5 days</td>
<td>0 0 0 0 0</td>
</tr>
</tbody>
</table>

TABLE 2

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

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>Concentration</th>
<th>Fission Density</th>
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</thead>
<tbody>
<tr>
<td>Cm$^{244}$</td>
<td>$1.8 \times 10^5$ atoms/km$^2$</td>
<td>$2.0 \times 10^5$ dis/day-km$^2$</td>
</tr>
<tr>
<td>Pu$^{241}$</td>
<td>$1.4 \times 10^5$ atoms/km$^2$</td>
<td>$2.0 \times 10^5$ dis/day-km$^2$</td>
</tr>
<tr>
<td>Pu$^{242}$</td>
<td>$1.9 \times 10^2$ atoms/km$^2$</td>
<td>$2.0 \times 10^5$ dis/day-km$^2$</td>
</tr>
<tr>
<td>Pu$^{243}$</td>
<td>$7.1 \times 10^5$ atoms/km$^2$</td>
<td>$1.9 \times 10^4$ dis/day-km$^2$</td>
</tr>
<tr>
<td>Pu$^{244}$</td>
<td>$4.5 \times 10^3$ atoms/km$^2$</td>
<td>$1.9 \times 10^4$ dis/day-km$^2$</td>
</tr>
</tbody>
</table>

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