Reports

Detection of Uranium from Cosmos-1402 in the Stratosphere

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The nuclear reactor from the Soviet radar reconnaissance satellite, Cosmos-1402, reentered the earth's atmosphere on 7 February 1983 and disintegrated over the South Atlantic Ocean. The reactor was powered by approximately 50 kilograms of uranium-235 (^{235}U). In an effort to determine the fate of the reactor core, a series of aerosol samples were collected at altitudes between 27 and 36 kilometers in the Northern Hemisphere approximately 1.1 years later by high-altitude balloons. At an altitude of 36 kilometers a 53 ± 20 percent excess in the ^{235}U concentration was measured. The total excess of ^{235}U in the stratosphere was calculated to be 44 ± 15 kilograms.

Cosmos-1402, a Soviet radar reconnaissance satellite powered by a nuclear reactor that contained ~50 kg of ^{235}U, was launched into an earth orbit of 240 to 270 km on 30 August 1982 (*1, 2*). After completion of their missions, satellites of this type are normally separated into several components and the reactor is boosted into a stable long-lived orbit (~1000 km). A malfunction on 28 December 1982 prevented the boost of the reactor into its burial orbit, and on 7 February 1983 it reentered the earth's atmosphere ~2 weeks after the reentry of the main part of the satellite (*3*). It was believed that the reactor burned up in the stratosphere over the South Atlantic Ocean south of Ascension Island (*4*), but there was no direct evidence that this occurred. In an effort to determine the fate of the reactor, the ^{235}U/^{238}U ratio of the aerosol in the stratosphere was measured by mass spectrometry. This uranium isotopic ratio is a particularly sensitive tracer for this kind of event because the natural ratio, 0.0072, is small and constant, whereas the ratio for the fuel of a reactor of this type is about 10.

A previous study (*5*) provided direct verification of the burnup of a similar satellite, Cosmos-954, in the stratosphere. However, the reentry trajectories of Cosmos-954 and Cosmos-1402 were considerably different. Cosmos-954 disintegrated and burned up in the Northern Hemisphere at an approximate altitude of 40 km, whereas Cosmos-1402 disintegrated in the Southern Hemisphere at an approximate altitude of 76 km. Since balloon launches and sampling were restricted to the Northern Hemisphere (*6*), it was necessary to wait until stratospheric circulation transported the reactor aerosol to lower altitudes in the Northern Hemisphere before sampling. The timing of balloon launches for the interception of Cosmos-1402 debris was based on previously published ^{238}Pu data that was obtained after the reentry of the U.S. satellite SNAP-9A in 1964 (*7*). Based on 1983 meteorological observations, air from 76 km in the equatorial area should have been transported down to 40 km within a few months (*8*). The maximum concentration of ^{238}Pu was measured at a 35-km altitude and 33°N latitude 9 months after reentry of SNAP-9A (*9*). It was estimated that the best time for sampling Cosmos-1402 debris would be in the late winter of 1983 to 1984.

Table 1. Filter samples collected at Holloman Air Force Base, New Mexico (33°N), in the study of Cosmos-1402. Filters 1 and 2 served as blanks for filter 6, filter 3 served as a blank for filters 7 and 8, and filter 4 served as a blank for filter 5. Filter 5 was sampled on 23 January 1983 before reentry of Cosmos 1402, whereas filters 6 through 8 were sampled after reentry (filter 6, 28 February 1984; filters 7 and 8, 13 March 1984).

Filter number	Altitude (km)	Air volume* (scm)	Total weight (g)
5	38.6	40.5	111.6846
6	36.0	92.4	106.5562
7	26.6	60.5	15
8	31.9	17.9	15

*The temperature used was 288.15 K; scm is standard cubic meter.

One balloon was launched on 28 February 1984 and one on 13 March 1984 to provide samples between 27 and 36 km (Table 1). A flight launched on 23 January 1983 (16 days before the reentry of Cosmos-1402) provided an important background sample that permitted measurement of the concentration of uranium nuclides in the stratospheric aerosol prior to the burnup of Cosmos 1402. Aerosol samples were collected on IPC-1478 cellulose filter paper in samplers designed and used in the Department of Energy's (DOE) project ASHCAN (*10*). Two batches of filter papers were selected for this study because of their low uranium content. Filters 4 and 5 were cut from the same batch, whereas filters 1 to 3 and 6 to 8 came from the second batch. In the Cosmos-954 study, there was no preselection of filters, and consequently the filters had inherently large heterogeneously blank uranium concentrations. Portions (~10 g) of filters were spiked with ^{233}U, wet ashed, chemically purified by anion exchange. The uranium isotopic ratios were measured by thermal ionization mass spectrometry with pulse counting detection at the National Bureau of Standards (*11*). Chemical yields were determined for all samples and the appropriate blank corrections were applied to both the concentration and composition data.

The composition and concentration data are presented in Tables 2 and 3, respectively. The uncertainties reported in these tables reflect random variation due to mass spectrometry measurements, sample preparation, inhomogeneity of the blank filter paper, and, for filter 6, inhomogeneity of deposition of the uranium aerosol. Relative to these uncertainties, which have been specifically taken into account, any possible systematic errors in the measurements are negligible. The blank filters show a small

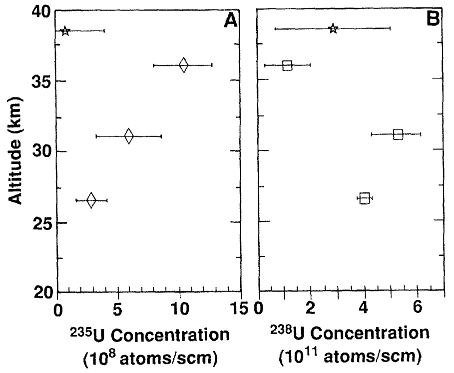

Fig. 1. Aerosol concentrations of stratospheric uranium for (**A**) ^{235}U and (**B**) ^{238}U; scm is standard cubic meter. Stars are pre-Cosmos-1402 (background) concentrations; diamonds and squares are values from post-Cosmos-1402 samples. The error bars are ±2 SEM (*19*).

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enrichment of ^{235}U compared to natural uranium. This was also observed in the previous study (*5*), but the exact reason for this is unknown. The mean and standard deviation for the ^{235}U atom percent values for the blank filters for the Cosmos-1402 and Cosmos-954 studies are 0.836 ± 0.006 and 0.789 ± 0.070, respectively. The Cosmos-1402 filters used in this study were an order of magnitude more homogeneous than the filters used previously, which decreased the perturbations in uranium concentrations.

Of the three post-Cosmos-1402 samples collected, only filter 6 showed a measured isotopic enrichment greater than the mean of the blank filters. It is not evident from the isotopic composition of filters 7 and 8 that Cosmos-1402 reactor uranium is present on the filters. We can, however, show that with the following model we can extract the values for the Cosmos-1402 components from these filters in which elevated levels of natural uranium are present.

The mass balance relation for $[^{235}\mathrm{U}]/[^{238}\mathrm{U}]$ can be written for each filter:

$$([^{235}\mathrm{U}]/[^{238}\mathrm{U}])_m = \frac{[^{235}\mathrm{U}]_b + [^{235}\mathrm{U}]_r + [^{235}\mathrm{U}]_a}{[^{238}\mathrm{U}]_b + [^{238}\mathrm{U}]_r + [^{238}\mathrm{U}]_a} \qquad (1)$$

where m, b, r, and a stand for measured, blank, reactor, and air, respectively. Since we have a good estimate of the ^{235}U and ^{238}U concentrations in the blank filter, we can calculate the concentrations of these isotopes in the natural background aerosol and the reactor-derived aerosol. We have assumed that $([^{235}\mathrm{U}]/[^{238}\mathrm{U}])_r$ is 10.6 (*12*) and have used this value to calculate the reactor ^{235}U. However, this assumption is not critical for the calculation of the $[^{235}\mathrm{U}]_r$ concentration. A ratio as low as 0.1 would not change the calculated concentrations. The results of these calculations (Table 3 and Fig. 1) give the $[^{235}\mathrm{U}]_r$ and $[^{238}\mathrm{U}]_a$ concentrations for filters 5 through 8. The error bars associated with the reactor and natural aerosol concentrations plotted in Fig. 1 reflect the errors given in Table 3. By assuming that background filter 5 is a two-component mixture of filter blank and ambient aerosol, we calculate $([^{235}\mathrm{U}]/[^{238}\mathrm{U}])_a$ to be 0.00727 ± 0.00054 (2 SD), which is indistinguishable from the natural uranium ratio of 0.0072 and indicates that the stratospheric aerosol had a natural $^{235}\mathrm{U}/^{238}\mathrm{U}$ ratio. This latter value was used for the air component in Eq. 1 for filters 6, 7, and 8.

Table 2. Uranium isotopic analysis of balloon filter samples. Only a part of the total filter was analyzed for filters 5 through 8 (filter 5, 33.3%; filter 6, 41.8%; filter 7, 23%; and filter 8, 37%). The uncertainties of the mean values for the measurements at the filter blanks are given by $\pm 2s_{\mathrm{bl}}\, n^{-1/2}$, where $n = 4$ is the number of filter pieces measured and s_{bl} is the standard deviation reflecting variations due to filter inhomogenity and measurement error (*16*). For filters 5 and 6 the uncertainties are twice the standard error for the weighted mean (*17*), and for filters 7 and 8 the standard errors for the isotopic abundances are approximated as twice the raw standard deviations (s_{bl}) for the blanks (*18*).

Filter number	Filter weight* (g)	Atoms ^{238}U/g ($\times 10^{10}$)	Atom percent*			
			^{234}U ($\times 10^{-4}$)	^{235}U ($\times 10^{-3}$)	^{236}U ($\times 10^{-4}$)	^{238}U
			Natural uranium			
			55	720	0	99.28
			Cosmos-1402 reactor†			
			10,000	90,000	5,000	8.5
			Filter blanks			
1	4.2430	48.1	85.3	848.7	3.0	99.147
2	4.3307	47.6	95.2	831.4	32.7	99.157
3	6.1505	57.4	79.3	818.7	2.0	99.177
4	8.1178	47.3	80.3	833.9	0.0	99.158
Mean		50.1 ± 4.9	85.0 ± 7.3	833.2 ± 12.3	9.4 ± 15.6	99.160 ± 0.013
			Pre-Cosmos-1402 sample			
5	37.1756	60.9 ± 5.7	88.1 ± 13.6	817.3 ± 5.8	8.8 ± 4.0	99.173 ± 0.006
			Post-Cosmos-1402 samples			
6	44.5526	61.0 ± 5.4	101.9 ± 10.2	958.3 ± 21.4	17.7 ± 3.7	99.031 ± 0.021
7	3.4602	210.5 ± 9.8	71.9 ± 14.5	800.5 ± 24.6	1.5 ± 31.2	99.192 ± 0.025
8	5.4806	113.9 ± 9.8	98.0 ± 14.5	830.0 ± 24.6	11.4 ± 31.2	99.159 ± 0.025

*Mass spectrometric repeatabilities for atom percent values (*15*). †Assumed to be similar to a National Bureau of Standards enriched uranium sample (*12*).

Table 3. Uranium isotopic concentrations in atoms per standard cubic meter.

Filter	$[^{235}\mathrm{U}]_r$ ($\times 10^{-9}$)	$[^{235}\mathrm{U}]_a$ ($\times 10^{-9}$)	$[^{238}\mathrm{U}]_r$ ($\times 10^{-7}$)	$[^{238}\mathrm{U}]_a$ ($\times 10^{-11}$)
5	0.072 ± 0.305	2.2 ± 1.6	0.68 ± 2.88	3.0 ± 2.2
6	1.042 ± 0.239	0.9 ± 0.6	9.84 ± 2.46	1.3 ± 0.9
7	0.284 ± 0.131	2.88 ± 0.23	2.68 ± 1.26	4.0 ± 0.3
8	0.584 ± 0.273	3.88 ± 0.76	5.52 ± 2.63	5.35 ± 1.05

The concentrations are calculated from Eq. 1. The stated uncertainties are ±2 SEM (*19*).

The average mass of Cosmos-1402 ^{235}U per gram of filter 6 corresponds to $0.9 \times 10^9 \pm 0.2 \times 10^9$ atoms (*13*) or a total amount for the filter of 38 pg of ^{235}U. This corresponds to an excess in ^{235}U of 53 ± 20% relative to natural uranium. The highest measured ^{235}U atom percent found in the Cosmos-1402 study was 0.958 ± 0.021, which is significantly smaller than the value obtained in the Cosmos-954 study (1.564 ± 0.003). This is ascribed to the higher altitude of injection of Cosmos-1402 than Cosmos-954, the later sampling time of the Cosmos-1402 study, and the subsequently greater dispersion of the material.

The concentration of reactor ^{235}U/scm (standard cubic meter) of air as determined on filters 6, 7, and 8 can be used to estimate the total mass of ^{235}U injected into the atmosphere by Cosmos-1402. As a first approximation, we assume the ablation aerosol is distributed uniformly around the globe with the concentrations shown in Fig. 1. The reactor aerosol concentration is assumed to be constant between 36 km and 76 km, the altitude at which the reactor was estimated to begin burnup. The concentration of reactor aerosol below 26.6 km was assumed to be zero. We calculated that the total mass of ^{235}U injected into the stratosphere was 18 ± 6 kg (*14*). If the Southern to Northern Hemisphere inventories are assumed to be in the ratio of 4 to 1, as was observed for Snap-9A (*9*), then the calculated total mass of ^{235}U would increase to 44 ± 15 kg. If the ablation material is kept below 40 km, the overall inventory would decrease by 27%. These values are near the reported reactor mass of 50 kg of ^{235}U and suggest that all of the excess ^{235}U in the upper atmosphere could have come from this one reactor. The natural uranium isotopic composition calculated for filter 5 (*12*), which is the pre-Cosmos-1402 sample, indicates that the residence time of the reactor aerosol at 39 km from the 24 January 1978 burnup of Cosmos-954 was less than 5 years.

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13. Based on the method of calculation from (*5*), in which paired isotope data was used, the average mass of Cosmos-1402 ^{235}U per gram is $0.7 \times 10^9 \pm 0.2 \times 10^9$ atoms per gram. This value is in good agreement with the value from the more rigorous calculation in Eq1.
14. If the calculated ^{235}U concentrations are uniformly distributed worldwide above 26.6 km, the total stratospheric inventory of ^{235}U would be 18 ± 6 kg. If the ablation material is kept below 40 km, the overall inventory would decrease by 27%.
15. Mass spectrometry repeatabilities (±2 SD) for atom percent values are: ^{234}U: $\pm 31 \times 10^{-4}$; ^{235}U: $\pm 17 \times 10^{-3}$; ^{236}U: $\pm 12 \times 10^{-4}$; and ^{238}U: ±0.017. These values were calculated by pooling the standard deviations from all samples for which two or more mass spectrometry measurements were run.
16. The uncertainties for the measurements of the filter blanks are given as $\pm 2\ s_{\text{bl}} n^{-1/2}$ where $n = 4$ filter pieces measured. The standard deviation, s_{bl}, reflects variation due to filter inhomogeneity plus measurement error. Blanks 1, 2, and 3 were from the same batch of filter paper, which was the same paper used for filters 6, 7, and 8. Blank 4 was from the same paper used for blank 5.
17. The ^{235}U/^{238}U ratio data for filter 6 showed clear evidence of inhomogeneity among the seven filter sections analyzed. This was taken into account statistically by treating the filter sections analyzed as a random sample drawn without replacement from the filter. The atom percent data for the analyzed filter sections were summarized by the weighted mean, X_{w}, of the values for the individual sections with the use of weights equal to the measured masses of the filter sections. The standard error, $s(X_{\text{w}})$, used to summarize the uncertainty was calculated with the formula:

$$S\,(\bar{X}_{\text{w}}) = \left[\frac{S_{\text{B}}^2}{W_{\text{s}}}\left(1 - \frac{W_{\text{s}}}{W}\right) + \frac{S_\epsilon^2}{W_{\text{s}}^2}\left(\sum_1^n \frac{w_i^2}{m_i}\right)\right]^{1/2} \quad (1)$$

where W_{s} is the total weight of the analyzed filter sections, W is the total weight of the unanalyzed plus analyzed filter sections, w_i is the weight of the *i*th section of filter (in grams), m_i is the number of mass spectrometry runs for section "*i*," n is the number of filter sections analyzed ($n = 7$ for filter 6 and $n = 5$ for filter 5), s_ϵ is the standard deviation due to mass spectrometry, and S_{B} is the "between" filter section component of variation, computed as:

$$S_{\text{B}} = \left[\frac{\sum_1^n w_i(\bar{X}_i - \bar{X}_{\text{w}})^2}{n-1} - \frac{S_\epsilon^2}{n-1} \left(\sum_1^n \frac{w_i}{m_i} - \frac{\sum_1^n \frac{w_i^2}{m_i}}{W_{\text{s}}}\right)\right]^{1/2} \quad (2)$$

(S_{B} is taken to be zero if the quantity inside the brackets is negative, as it was for filter 5.)

For the concentration measurements (that is, atoms ^{238}U/g), the same weighted mean was used to summarize the data for the filter sections analyzed. Since only one concentration measurement was made per filter section, formula 1 was modified to allow for the unavailability of the repeatability standard deviation, S_ϵ. Thus the standard error for the weighted mean was calculated with the formula:

$$S\,(\bar{X}_{\text{w}}) = \left[\left(\frac{1}{W_{\text{s}}}\right)\frac{\sum_1^n w_i\,(\bar{X}_i - \bar{X}_{\text{w}})^2}{n = 1}\right]^{1/2} \quad (3)$$

Formula 3 is conservative in that it gives a larger result than would be obtained if S_ϵ were available and formula 1 were used.
18. A single filter section ($n = 1$) was analyzed for both filter 7 and 8 so that the formulas described in (*17*) were not applicable. Instead, the data for these filters were treated as if the between-section component of variance was the same as for the blanks 1 through 4. Thus the standard errors for the isotopic abundance and concentration data were approximated by the raw standard deviations, s_{bl}, obtained for the blanks.
19. The stated uncertainties are ±2 SEM. These were calculated by propagation of errors from the standard errors given in Table 2. The standard errors for the ^{235}U/^{238}U ratios, which are needed for Table 3, may be calculated from the standard errors for the ^{235}U and ^{238}U abundances as:

$$\sigma(A_5/A_8) = \frac{A_5}{A_8}\left\{\left[\left(\frac{\sigma\,(A_5)}{A_5}\right)^2 - \left(\frac{\sigma\,(A_8)}{A_8}\right)^2\right] \Big/ (1 - 2A_5)\right\}^{1/2}$$

where A_5 is the isotopic abundance of ^{235}U, A_8 is the isotopic abundance of ^{238}U, and the σ function gives the standard error of the argument. This formula takes into account the correlation between the ^{235}U and ^{238}U abundances, A_5 and A_8. In the above formula, A_8 and A_5 must be expressed as proportions (that is, numbers between 0.0 and 1.0) and not as percentages.
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X-ray Holograms at Improved Resolution: A Study of Zymogen Granules

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X-ray holography offers the possibility of three-dimensional microscopy with resolution higher than that of the light microscope and with contrast based on x-ray edges. In principle, the method is especially advantageous for biological samples if x-rays in the wavelength region between the carbon and oxygen K edges are used. However, until now the achieved resolution has not exceeded that of the light microscope because of the poor coherence properties of the x-ray sources and the low resolution of the detectors that were available. With a recently developed x-ray source based on an undulator on an electron storage ring, and high resolution x-ray resist, a hologram has been recorded at about 400-angstrom resolution. The experiment utilized x-rays with wavelengths of 24.7 angstroms and required a 1-hour exposure of the pancreatic zymogen granules under study.

A NUMBER OF THEORETICAL STUDIES (*1, 2*) have suggested the possibility of using soft x-rays (wavelength 10 to 50 Å) to make three-dimensional images of microscopic biological objects. One of the methods proposed is x-ray holography, a technique that is capable, in principle, of three-dimensional imaging of biological samples with resolution of features in the 100- to 1000-Å range. Moreover, this capability should be applicable to cells and subcellular structures in something closely related to their natural state: that is, without dehydration, sectioning, or staining (*3*).

We have now verified some of these predictions experimentally. We report results that demonstrate that x-ray holograms, which contain information with 400-Å resolution, of biological objects can be recorded on resist with 25- to 32-Å soft x-rays (*4*) with reasonable exposure times. The significance of such a recording is that this has hitherto been the missing step in the sequence of operations needed to make a holographic image at that resolution level. We shall comment on the other steps in the sequence. They present some difficulties, but have all been demonstrated in other contexts.

The achievement of recordings of this quality has been prevented in the past by the poor coherence properties of available x-ray sources. We have addressed this in the present experiments by using one of the recently developed undulator sources (*5*). These devices provide a copious flux of spatially and temporally coherent soft x-rays and have many laser-like properties. Earlier sources,

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