COSMOS 954 - DETECTION OF BURNUP IN THE STRATOSPHERE

INTRODUCTION

During the month of December 1977, U. S. observers noticed the orbit of a recently launched Russian Sattelite misbehaving( ). On January 24, 1978, the sattelite re-entered the earth's atmosphere at a height above 40 km and passed over Great Slave Lake (62°N, 115°W) and extended 800 km to Bakers Lake (65°N, 95°W) in Canada. Large sections of pipe, associated with a 40 kw Romashta type thermal reactor used on the satellite, were found over an extensive impact area(). The core of the reactor containing 50 kgr of U-235 was never found, and presumably lost under ice or burned up in the stratosphere.

Assuming a major portion of the core ablated in the stratosphere, the Environmental Measurements Laboratory (EML) of the U. S. Department of Energy initiated a program of balloon flights to investigate the potential atmospheric enrichment of isotopic uranium by Cosmos 954 debris.

This paper describes the detection of debris from Cosmos 954 in filter samples collected in the stratosphere during the spring and summer of 1978.

BALLOON LAUNCH FACILITIES

The U. S. Department of Energy (DOE) has an active program investigating the distribution of nuclear and non-nuclear materials injected into the stratosphere by natural and anthropogenic processes. One program, "Project Airstream" ( ) involves flights in the Northern Hemisphere, using a WB-57 aircraft to provide gas and filter samples to 19 km. A second program "Project Ashcan" ( ) involves equipment mounted on balloons to obtain gas and filter samples at three altitudes (21, 24.5 and 27 km). With the Air Force ( ) Geophysics Laboratory and New Mexico State University ( ) launches are carried out at Howard AFB Panama (6°N), Holloman AFB, New Mexico (33°N) and Eielson AFB, Alaska (65°N). During the spring sampling period all three bases are used but in the summer and fall, flights are scheduled only at Holloman AFB. To intercept Cosmos 954 debris, special balloons, using a high volume filter sampler were used to provide samples to a height of 39 km.

As shown in Table 1 filters for this study were collected at the three launch sites from April to July. Included in the Table is information related to each flight. An additional high altitude flight was arranged by Air Force Geophysics Laboratory in Alaska for early September. A complete description of the balloon facilities and filter sampling equipment can be found in an EML Quarterly Report ( ).

CHEMICAL ANALYSIS OF FILTERS

Each filter is divided into three parts, one section for analysis of γ emitting radionuclides at EML and two sections for bulk isotopic analysis of uranium and individual particle analysis using Lexan techniques ( ) at Knolls Atomic Power Laboratory (KAPL).)

Irradiation of a filter sample (approximately 10 cm^2) in contact with a Lexan plastic strip, allows the identification of single uranium bearing particles. Any particles on the filter containing sufficient uranium-235 will produce fission tracks on the plastic, which when properly etched produce visible stars . A single particle containing app approximately 10^8 atoms of U-235 would produce 50 tracks. These stars can be optically seen and single particles transferred onto a micro grid for mass spectrographic analysis. [illegible] ut. The only requirement for the above analysis is that the uranium particles need to be greater than a few tenths of a micrometer in diameter. The above techniques allow the identification of ng quantities of uranium.

In addition, bulk analysis by isotopic dilution was carried out on subdivided sections of filters for isotopic uranium.

A detailed discussion of the analytical procedures used in this study can be found in KAPL? EML report No. ________.

RESULTS OF SINGLE PARTICLE ANALYSIS

Approximately 200 cm^2 of the June 4, 1978 high altitude filter sample (39 km) was analyzed using the Lexan technique and a total of 345 stars were observed. Only six stars containing double tracks could be seen, the rest being single track stars. Multi track stars could not be found eliminating the possibility of isotopic analysis of individual particles.

Using the fact that production of stars by thermal neutron irradiation follows a binomial probability distribution one is able to place an upper limit on the size of particles associated with the Cosmos 954 burnup.

S_i = = 5.3×10^{-7}

where (thermal neutron cross section) = 5.9×10^{-22} cm^2

(thermal neutron flux) = 2.03×10^{11} cm^{-2} Sec^{-1}

t (irradiation time) = 4500 seconds

Because of this low probability and the fact of discrete track formation, i.e., either a track is or is not formed, the Poisson probability distribution may be used as an approximation to the binomial probability distribution to calculate the average number of atoms in a given particle that forms tracks. This distribution is given by

P_r =

where r = 0, 1, 2.... represents the number of fission tracks per particle formed.

and P_r is the frequency of occurrence of stars with "r" tracks

and $\lambda = NS_i$ is the expected number of tracks

S_i is given by equation (1)

N is number of atoms per particle

the ratio of double to single track stars is given by

and n = 6 atoms

This represents the total number of atoms per particle of uranium-235 necessary to produce the distribution of stars observed. Using the fact that each particle contains 89%, U-235 the total atoms of uranium in each particle is approximately 78,000 atoms. The equivalent mean radius of a uranium oxide spherical particle (density of 11g cm^{-3}) containing these uranium atoms is 9 mμm. The error associated with this count is   or 41%. Physical factors which would tend to increase the number of double tracks, such as coincidences of tracks in the Lexan and small particles of natural uranium background in the filter paper will cause an increase in the equivalent diameter.

SNAP-9A - a nuclear reactor containing plutonium burnt up in the equatorial stratosphere in 1964 producing particles whose sizes were estimated to be between 10 and 15 mμm in radius ( ), similar to the Cosmos 954 Burnup.

RESULTS OF MASS SPECTROGRAPHIC ANALYSIS OF FILTERS

Results of mass spectrographic analysis of filters using bulk concentration techniques are shown in Table 3. Reported atom percentages are based on the addition of known spikes of U-233 to each sample. The blank filter paper shows a small enrichment of U-236 indicating an unknown source of contamination occurring during or after manufacture of the filter paper. The collected samples indicate significant enrichment of the minor isotopes above the blank. Errors listed represented one standard deviation and are based on information associated with mass spectrographic analysis. The absolute accuracy of the data is traceable to NBS standard and has shown to be better than 1% ( ). Included in Table 3 is a mass spectrographic analysis (Canadian) of a particle removed from a pipe section, associated with the Cosmos 954 reactor found on the ground in Canada ( ). This represents the atom percentage of isotopic uranium in Cosmos 954 debris.

If one makes the assumption that the only contribution to uranium measured in our filter samples are from existing stratospheric aerosols, background filter composition and Cosmos-954 debris, then a set of four simultaneous equations can be obtained, that are based on the atom percentage of isotopic uranium found in the sample. Using any two isotopes of uranium allows the calculation of the Cosmos-954 fraction of uranium.

let

x = the total atoms of Cosmos-954 uranium on the filter

y = the total atoms of background uranium on the filter

Ut = the total atoms of uranium measured on the filter.

then

$$S_i = \frac{C_i X + B_i Y}{U_t}$$

Where "i" can be anyone of four uranium isotopes (U-234, U-235, U-246 and U-238) and S_1, B_1 and C_1 represents the atom percentages of isotopic uranium for the sample, background and Cosmos-954 respectively. Calculations based on pairs of isotopes are shown in Table 4 and confirm the existence of Cosmos-954 debris in the stratosphere. Because of the variability of U-234 and U-236 atom percents found in the blank filters, the internal agreement between pairs of isotopes is not exact. Using the data for the U-235 and U-238 isotopes does show significant enrichment over the blank filter and the July 20, Holloman flight. These calculations support the concept that Cosmos-954 debris was present in the filter sample. The September Alaska flight also shows a Cosmos-954 uranium enrichment of U-235.

The ability to detect Cosmos-954 debris in such a large uranium background is due to the unique uranium isotopic composition of Cosmos-954. The filter collected on July 20, 1978 at 30°N and 37 km represents a true background sample as the Cosmos-954 debris had not penetrated into this latitude band. The isotopic composition of this filter is very similar to the blank unexposed filter.

In addition, the filter collected on June 3, 1978 at 65°N and 31 km altitude has an isotopic composition similar to blank filters indicating that prior to Cosmos-954, stratospheric aerosols, appears to contain negligible quantities of uranium. It is evident from the data presented that the debris from the Cosmos-954 burnup had not extended to 33°N latitude in July 1978 nor down to 31 km at 65°N in June 1978.

CONCLUSIONS

Using the best available analytical techniques it has been possible to detect subnanogram quantities of Cosmos-954 uranium in filter samples collected at 39 km over Alaska. This data supports the hypothesis that a fraction of Cosmos-954 burned up in the stratosphere.

Using Lexan techniques an estimate of the average size of the Cosmos-954 particles in the stratosphere was found to be 9mμm (equivalent radius of spherical UO_2 particles), similar to the findings of the SNAP-9A sattelite burnup.

As the debris penetrates into lower altitudes, our aircraft sampling could provide filters for analysis of Cosmos-954 debris. Spatial distributions of the debris could be obtained for the Northern and possibly Southern hemisphere but would depend on the development of a new low blank uranium filter paper. Data from these flights could provide insight into what fraction of the Cosmos-954 reactor burned in the stratosphere and be a unique tracer of stratospheric motion.