Equation 2 can be integrated analytically and, using as the boundary condition the present values of Hubble's constant, matter density, and radiation temperature, one obtains the curves shown in Fig. 2, which represent the physical conditions in the universe all the way back to the first seconds of its existence (*6*).

There were two important periods in the history of the expanding universe. The first 30 minutes after the singularity at the moment of the origin were responsible for the formation of light elements as a result of thermonuclear reactions in the primeval *ylem* (that is, a mixture of protons, neutrons, electrons, and, above all, light quanta). The pioneering work in this direction was carried out by Fermi and Turkevich (*7*), who integrated the equations of thermonuclear reaction in light nuclei for the variable temperatures and densities corresponding to the early periods of expansion. Their calculations have shown that the ratio of hydrogen to helium at the end of the reaction period is expected to be about 50:50, in reasonable agreement with the observed values. On the other hand, due to the absence of reasonably stable nuclei of mass 5, the amounts of lithium and all other heavier elements came out too small by a factor of about 100, as compared with the astronomical data for the sun and stars. Recently the calculations of Fermi and Turkevich were repeated more exactly by Wagoner, Fowler, and Hoyle (*8*), who used modern values for the cross sections of various nuclear reactions and the electronic computer for obtaining the curves of growth. These new calculations generally confirmed the conclusions of the earlier ones, leading to a result that heavy elements could not have been formed during the early stages of the expanding universe, but must be attributed to the vast stellar explosions (supernovae) of the early history of the universe (*9*). This double attitude is, however, quite acceptable since the recent progress of astronomical knowledge has shown that the earlier assumption of the chemical homogeneity of the universe is not quite correct. In fact, while the stars of Baade's Population I contain comparatively large amounts (up to 1 percent) of the heavier elements, the amounts of these elements in the stars of Population II is, at least, a hundred times smaller. It is generally agreed that the stars of Population II represent the original stock of stars, while the stars belonging to Population I may be formed by the recondensed material of the supernovae which exploded early in the history of the universe. Thus it is quite likely that, whereas in the former case we have agreement with the Fermi-Turkevich and later calculations, in the latter case the stars are enriched by the material synthesized in the preceding supernovae.

The second interesting period in the history of the universe is around $t = 1.0$ millieons where we have: $\rho_r = \rho_m = 5 \cdot 10^{-22}$ g/cm^3 and $T = 3000°$K, which corresponds to the transition of the expanding universe from the radiation regime to the matter regime. The early attempts (*10*) to get the correct sizes and masses of protogalaxies by using Jeans's formula for the gravitational instability were all leading to the mass values of protogalaxies that were too small by about three orders of magnitude. The change of scale, and the use of better temperature and density curves, did not help the situation, and the problem is still open. One obvious improvement would be to use in the Jeans's formula, not the mean thermal velocity of particles as is usually done, but what can be called an "effective velocity" which represents the velocity on the tail of the Maxwell's distribution, for which the evaporation rates of the condensations are sufficiently small, as compared with the rate of their separation, resulting from the expansion of space. Another important factor, which was not previously taken into account, is the role of thermal radiation, the mass density of which is comparable to that of matter. In particular, the transition of ionized hydrogen and helium into their neutral forms takes place for the temperatures characteristic for this period of the history of the universe. If everything fails, there is still the possibility that the supersonic turbulence of the primordial gas would be of some help. But we shall see.

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31 August 1967

Atmospheric Burnup of a Plutonium-238 Generator

Abstract. *The stratospheric inventory of the plutonium-238 resulting from the disintegration of a nuclear auxiliary power generator (SNAP-9A) in early 1966 accounts for essentially all the plutonium present in the original generator that reentered the atmosphere. Consequently, the pyrophoric ^{238}Pu must have completely burned up during reentry and ablated into small particles. The arithmetic mean of the distribution of the ^{238}Pu particle size was estimated to be 10 millimicrons, which confirms this conclusion.*

On 21 April 1964, a navigational satellite employing a SNAP-9A generator (Systems for Nuclear Auxiliary Power) did not reach orbital velocity because of a rocket failure after launch. The SNAP-9A generator is a nuclear-fueled power package which converts the heat developed by a radioactive source into electrical energy, contains about 17 kilocuries of ^{238}Pu and weighs 12.3 kg (*1*). Since ^{238}Pu is a highly toxic nuclide, and since bone is the critical organ for soluble plutonium and lung for insoluble plutonium, considerable interest was exhibited in the ultimate fate and disposition of the ^{238}Pu.

Korsmayer (*2*) estimated that the satellite entered the atmosphere at about 150,000 feet (46 km or 46,000 m) over the Indian Ocean in the Southern Hemisphere. There are three alternatives as to what could have happened when the SNAP-9A reentered the atmosphere. One is that it plunged intact into the Indian Ocean leaving little or no remnants in the atmosphere. A second is that the heat of reentry into the atmosphere completely consumed the device and the pyrophoric ^{238}Pu ablated into small particles. The third alternative is some combination of the

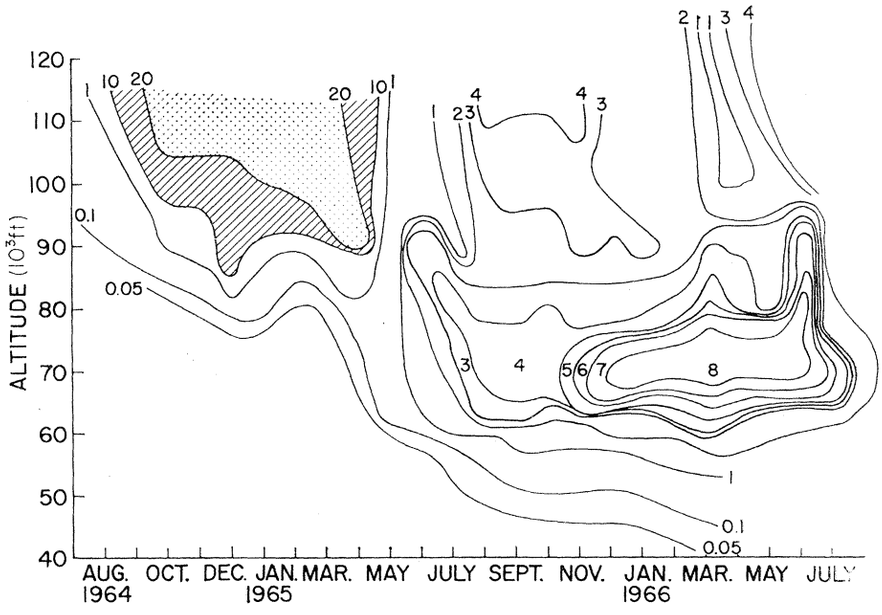

Fig. 1. Stratospheric SNAP-9A ^{238}Pu concentration dpm/10^3 SCF (standard cubic feet), at 34°S. Concentrations at 90,000 feet (27 km) and above show a cyclic pattern and reach a maximum during the winter and spring, which is followed by a reduction in the fall. Cell of high concentration in the layer from 60,000 to 90,000 feet formed in June 1965 and enhanced during first half of 1966.

first two, that is, a partial burnup with some bulk loss to the Indian Ocean. Various atmospheric sampling programs were alerted to the generator's loss and the search for its whereabouts and disposition was underway.

Our first objective in this search was to identify positively, as quickly as possible, even the slightest remnants of the generator. This identification would demonstrate that some burnup of the device did occur in the atmosphere. Second, the subsequent behavior of the SNAP-9A debris with latitude, altitude, and season was to be studied. This behavior would reflect the stratospheric transport processes going on and the interrelation between latitude and concentration of SNAP-9A debris. Third, a synoptic structure of the debris in the stratosphere was to be developed which would permit an estimate of the stratospheric burden of ^{238}Pu. Finally, the particle size of the ^{238}Pu from the generator was to be measured, and the result indicate the intensity of the burn-up.

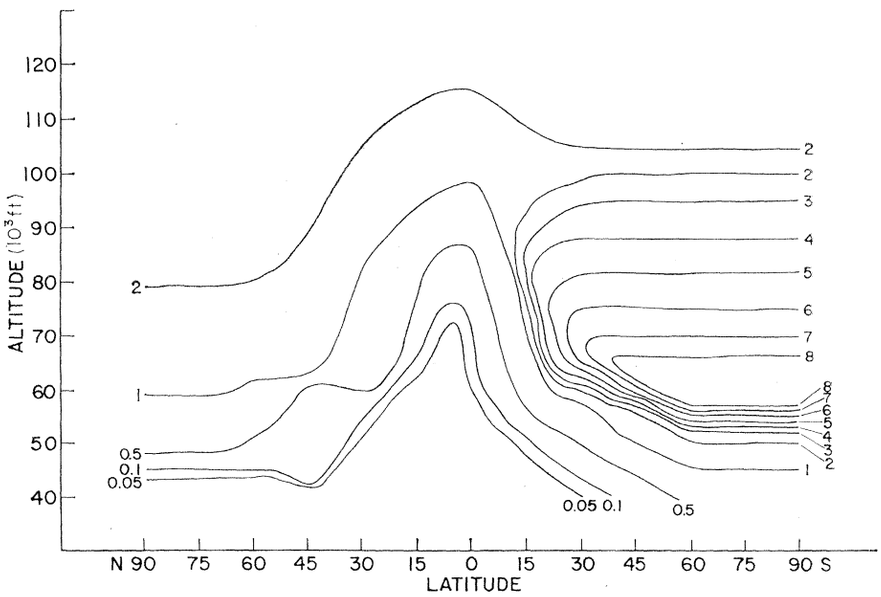

Fig. 2. Synoptic distribution of SNAP-9A ^{238}Pu concentrations dpm/10^3 SCF, for the period January through March 1966. The bulk of the debris resides in the middle to polar latitudes of the Southern Hemisphere. Contours of equal concentration increase with altitude near the equator. Little debris descended into the troposphere by this time.

The initial identification of SNAP-9A debris was made by the AEC's High Altitude Balloon Sampling Program at 108,000 feet over 34°S on 26 August 1964, 4 months after the injection. The subsequent pattern of SNAP-9A debris measured in the stratosphere at this latitude is shown in Fig. 1. The concentrations below 80,000 feet were derived from Project Stardust, an aircraft sampling program sponsored by the Defense Atomic Support Agency (DASA). The Pu data from these two programs have already been reported (*3*).

There was a small quantity of ^{238}Pu in the stratosphere produced by previous nuclear weapon testing. This background ^{238}Pu is equivalent to about 3 percent of the $^{239,240}Pu$ present (*4*). In Fig. 1 and subsequent figures, the background ^{238}Pu has been removed by subtracting 3 percent of the $^{239,240}Pu$ concentration in each sample from the total ^{238}Pu concentration measured to yield only the SNAP-9A ^{238}Pu concentrations.

An interesting feature of Fig. 1 is the relatively large incursion of SNAP-9A debris at the upper altitude, beginning in the winter season of each year and followed by a sharp reduction in the fall. This behavior agrees with transport models of the stratosphere, describing a subsidence of air from upper altitudes in the winter and spring (*4*) with subsequent upwelling of air from lower altitudes in the summer and fall (*5*). The downward transport of SNAP-9A debris from 115,000 to 80,000 feet in the spring of 1964 and 1965 averaged 0.09 cm/sec (7500 feet/month), a value in good agreement with the downward velocity of ^{90}Sr at similar times and altitudes (*5*). The overall downward transport of the SNAP-9A material through the lower altitudes from 80,000 to 45,000 feet was slower, averaging about 0.03 cm/sec.

Beginning in June 1965, concentrations in the layer at 60,000 to 90,000 feet of Fig. 1 were higher than the concentrations either above or below this region. This condition became progressively more pronounced during late 1965 and through the first half of 1966.

Unfortunately, the range between 65,000 and 80,000 feet where the core of this high-concentration cell apparently resided is not well documented. Since there is no reasonable way for this cell of high concentration to be supported from upper altitudes, it must have been advected from other latitudes, presumably the Antarctic stratosphere. It is interesting that the contours of this high activity cell also reflect an upwelling of air in the fall, since lobes of this cell extend upward during the month of June in both 1965 and 1966. A similar pattern developed in the polar and mid-latitudes of the Northern Hemisphere during the first winter and spring after the reentry, although at reduced concentrations. The behavior in the equatorial stratosphere is markedly different. By early 1966, 2 years after the burnup, SNAP-9A debris had still not descended into the equatorial stratosphere at the relatively high concentrations or to the low altitudes, as it had at other latitudes.

A synoptic distribution of the debris on a global scale for the period of January to March 1966 is shown in Fig. 2. One can see from this distribution that little SNAP-9A has passed from the stratosphere into the troposphere by this time. The low concentrations in the equatorial stratosphere, increase in altitude of the concentration contours at the equator, uniform concentrations in the Northern polar stratosphere, and the bulk of the SNAP-9A debris in the middle to upper latitudes of the Southern Hemisphere are clearly discernible. This distribution is in accord with Machta's model of stratospheric winter circulation with each hemisphere (*4*); Machta describes a rising air column in the equatorial regions, a poleward flow at about 110,000 feet, and a downward flux at middle and upper latitudes.

Since the SNAP-9A debris entered the upper atmosphere of the Southern Hemisphere in the fall of 1964, the debris, according to Machta's model, could then be largely available in the very next season for transport poleward by the winter flow at the upper altitudes. Residual or subsequently available debris near the site of injection could later make its way into the Northern Hemisphere by a similar winter circulation of that hemisphere. The rising column of air in the equatorial regions could explain the minimum concentrations of SNAP-9A debris in that region of the stratosphere and in the increase in altitude of the concentration contours at the equator.

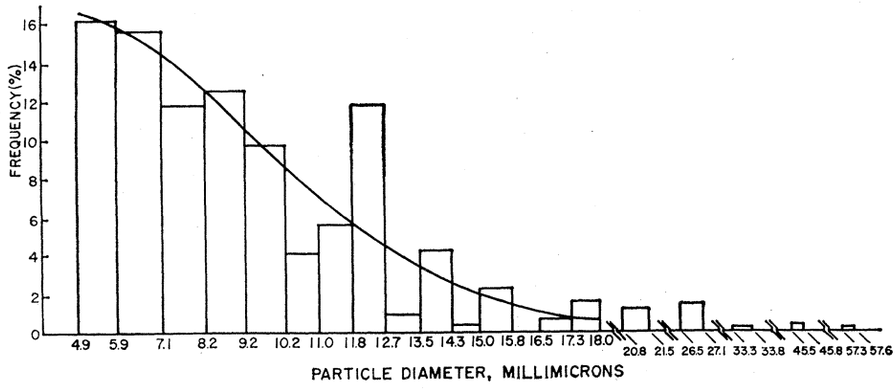

Fig. 3. Size distribution of SNAP-9A $^{238}PuO_2$ spherical particles. This distribution was estimated by first measuring the mass of ^{238}Pu in each SNAP-9A particle by autoradiography. Then the size of a spherical particle of $^{238}PuO_2$ containing an equivalent mass of ^{238}Pu was calculated. The histogram represents data for 1008 ^{238}Pu SNAP-9A particles exposed to a nuclear emulsion for 1 year.

The downward flux at middle latitudes could account for the early arrival of the debris at 34°S only 4 months after burnup and for the analogous arrival at 31°N in the winter of 1965. The bulk of the debris could continue poleward at the upper altitudes until it finally reaches the polar latitudes where it descends to lower altitudes. Although not included in Machta's model, the cells of high SNAP-9A concentrations in the 60,000 to 90,000 feet levels over each hemisphere suggest a subsequent equatorial flow from the polar regions.

By integrating the contours in Fig. 2 (*6*), a total stratospheric inventory of 15 kc of SNAP-9A ^{238}Pu or 88 percent of the 17 kc in the original generator can be accounted for. Of this, 80 percent resides in the Southern Hemisphere stratosphere, while only 20 percent was transported into the Northern Hemisphere. Surface air concentrations and deposition values of SNAP-9A ^{238}Pu in the Northern and Southern Hemispheres will ultimately reflect this 4 to 1 proportion. Based upon this inventory of SNAP-9A ^{238}Pu, we conclude that the generator completely burned up during reentry and ablated into small particles.

Figure 3 is an estimate of the particle-size distribution of the ^{238}Pu from the SNAP-9A debris. This distribution is based upon an autoradiographic technique developed by Tracerlab (*7*); the result represents the size of the SNAP-9A ^{238}Pu particles as equivalent PuO_2 spheres. The diameter of the spheres ranged from 5 to 58 mμ with an arithmetic mean of about 10 mμ. These measurements of particle size illustrate that the burnup of the generator was intense and verify the conclusion reached from the behavior of the ^{238}Pu concentrations, namely, that the generator ablated into very small particles.

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