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Environmental Studies Division

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ABSTRACT

This report presents current data from the HASL environmental programs, the Air Monitoring Section of the Bhabha Atomic Research Centre in India, the Health Physics Section of the Institute of Nuclear Science in Taiwan and the Radiological and Environmental Research Division at Argonne National Laboratory. The initial section consists of interpretive reports and notes on the history of long-range fallout, cesium-137 in Bombay milk, natural and fallout radioactivity in Indian diet, reporting results of radioactivity measurements at near zero levels of sample activity and background, plutonium in soil northeast of the Nevada Test Site, radon levels at the Lloyd, NY regional station, strontium-90 in New York and San Francisco diets through 1975, plutonium-239,240 in 1974 diet, up-dating stratospheric radionuclide inventories to July 1975 and a revised table of radionuclides. Subsequent sections include tabulations of radionuclide levels in stratospheric air; lead and radio-
nuclides in surface air; strontium-90 in deposition, milk, diet, tap water, and human bone; cesium-137 in Chicago foods in April 1976; and environmental radioactivity surveys for nuclear power plants in North Taiwan. A bibliography of recent publications related to environmental studies is also presented.

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Every three months, the Health and Safety Laboratory issues a report summarizing current environmental data obtained at HASL. This report, the latest in the series, contains information that became available during the period from March 1, 1976 to June 1, 1976. The next report is scheduled for publication October 1, 1976. Preceding reports in the series, starting with HASL-42, "Environmental Contamination from Weapons Tests", and continuing with HASL-306 (this report), may be purchased from the National Technical Information Service, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia 22161. A complete listing of these quarterly reports is given on the abstract page of this report.

To give a more complete picture of the current fallout situation and to provide a medium for rapid publication of radionuclide and non-nuclear pollutant data, these quarterly reports often contain information from other laboratories and programs, some of which are not part of the general ERDA program. To assist in developing, as rapidly as possible, provisional interpretations of the data, special interpretive reports and notes prepared by scientists working in the environmental field are also included from time to time. Many of these scientists are associated in some way with the general ERDA program. Information developed outside HASL is identified as such and is gratefully acknowledged by the Laboratory. In this report, data from the Air Monitoring Section of the Bhabha Atomic Research Centre in India, the Health Physics Section of the Institute of Nuclear Science in Taiwan, and the Radiological and Environmental Research Division at Argonne National Laboratory, are presented.

A portion of the analyses either have been or are being carried out by commercial laboratories under contract to the HASL Environmental Studies Division. The results of these analyses are reported as part of HASL's regular environmental program. The contractor analytical laboratories which provided data are Nuclear Science and Engineering Corp., Pittsburgh, PA.; Isotopes, Inc., Westwood, N.J.; Radiochemistry, Inc., Louisville, KY.; LFE Environmental Analysis Labs., Richmond, CA.; Controls for Radiation, Inc., Cambridge, MA.; Hazleton-Nuclear Science Corp., Palo Alto, CA. (now Teledyne Isotopes Palo Alto Labs.); Food Chemical & Research Labs., Inc., Seattle, WA.; Custom Nuclear Co., Mountainview, CA.; Ledoux and Co., Teaneck, N. J.; and U. S. Testing Co., Richland, WA.

This report is divided into four main parts:
1. Interpretive Reports and Notes
2. HASL Environmental Program Data
3. Data from Sources Other than HASL
4. Recent Publications Related to Environmental Studies.
Part I

INTERPRETIVE REPORTS

AND

NOTES
A BRIEF HISTORY OF LONG-RANGE Fallout

John H. Harley, HASL

Since 1976 is the 25th Anniversary of the first major effort on measurement of long-range radioactive fallout from nuclear weapons tests, it seems appropriate to summarize the history of the fallout program. This report will necessarily emphasize the HASL contributions since we are more familiar with this aspect of the work. Obviously, the weapons laboratories and military groups were involved in many programs, including highly sophisticated weapons diagnostics, which we had to avoid since knowing too much would have prevented us from open publication and discussion of our data. Most of their excellent work has never been published and it is unfortunate that the scientists involved have never received scientific credit for their work.

The national laboratories were largely involved in research aspects of the fallout problem such as the toxicity of strontium-90 and other radionuclides, meteorological studies, and mechanism studies. Many of them also carried out monitoring programs in their particular specialty, for example, Los Alamos, Brookhaven and Argonne had programs of whole body counter measurement for cesium-137 and Hanford was collecting and analyzing air samples. The Naval Radiological Defense Laboratory was of course very active during its existence, with particular interest in the Pacific test areas. All of these studies started up after concern had been expressed over possible hazards from long-range radioactive fallout.

Probably the most critical measurements would have been the external gamma exposure and the human intake of strontium-90, cesium-137, iodine-131 and plutonium. At the time when measurements began, however, this was not as clear, and the instrumentation then available was ill-suited to the specific measurements desired.

How it Started

The first indication of long-range fallout from nuclear weapons testing was obtained by Julian Webb of Eastman Kodak Research Laboratories in Rochester. (Webb, 1949) He found "abnormal radioactivity" in paper packing materials following the Alamagordo test in 1945. Radioactivity from natural sources had been found in film materials and packing still earlier and since these had caused some loss of sensitive x-ray film material, a general monitoring program was being carried out at Eastman.

The AEC became involved in long-range fallout on February 2nd of 1951 when the New York Operations Office (NYO) was told by AEC Headquarters that Eastman Kodak at Rochester had detected radioactivity in snow falling on January 29th. This was apparently related to the test of January 27th (one of the RANGER Series) at the Nevada Test Site. Merril Eisenbud of the Health and Safety Division dispatched laboratory teams towards the Canadian border, Watertown, Binghamton and Washington. Samples
collected by these teams were supplemented by others taken by AEC contractor groups from Luckey, Ohio, and from Rochester and Lewiston, New York. Sampling was very simple, snow was shoveled into containers and returned to the laboratory for analysis. Here, the melted snow was evaporated to dryness and beta counted in Petri dishes with geiger tubes calibrated against electroplated radium D + E standards. Activities up to 25,000 dpm/liter were found along the shore of Lake Ontario and near the Canadian border. (NYO, 1951) The University of Rochester also observed high activity in the snowfall of January 29th.

This was convincing evidence that fission products from the Nevada tests were being distributed across the country and that some monitoring system was necessary. It was considered that the health effects from these kiloton tests were not going to be significant but that sensitive industries such as film manufacturers might be affected. Thus the plans for monitoring the April-May, 1951 Pacific test series (GREENHOUSE) were developed in cooperation with the National Association of Photographic Manufacturers. They had indicated that their film problems came from particulates, since dissolved radionuclides would not produce a sufficiently high concentration to give a spot on film. It was noteworthy that, at a March 1951 meeting at NYO, the manufacturers agreed that water and air cleaning were going to be essential to reduce the effects of fallout on their industry. This was shown to be true, as later problems developed only when small paper factories without filtration produced materials containing radioactive particulates. The photographic problem essentially disappeared in a short time, although Eastman had to shut down their cellulose ester plant twice in 1952.

The spring 1951 (GREENHOUSE) test network consisted of three stations at Eastman, DuPont and Ansco film plants plus 5 stations set up by NYO and stations of the University of Rochester and Brookhaven. Deposition was collected in sheet metal trays, with the rainfall or washings being filtered and the filtered solids sent to the laboratory for counting. Airborne dust was collected by AEC contractors having facilities, and the samples forwarded to NYO. Activity was measurable at all sites, but at lower levels than those from the Nevada tests. (Blatz, 1951)

The Fall 1951 test series in Nevada (BUSTER-JANGLE) presented another problem. The long-range fallout network was well in hand, but an added requirement was then passed to NYO. The on-site radiation safety and measurements of radioactive deposition were under the control of the Test Director at the test site. Monitoring the immediate surrounding area was the responsibility of a team from UCLA under the direction of Kermit Larson. Once it became apparent that the debris was traveling long distances, the Health and Safety Division of NYO was asked to set up monitoring in the 200 to 500 mile radius. This started in the fall of 1951 and continued through a number of Nevada test series. Two laboratory men were each in charge of a group of 8 to 10 enlisted men from the Army Chemical Warfare Service. At the time of the detonation, the available trajectories were used as a basis to deploy 2 or 3 C47 aircraft with teams to various small airports within the 500 mile range. The aircraft would land, unload 2 men and their equipment and go on to additional sites. Sampling was commenced upon arrival.
and continued for 24-36 hours. Fallout trays were used for deposition and high volume samples, Aerotec centrifugal collectors and cascade impactors were used for air samples. All analyses were carried out back in the laboratory although portable GM counters were used to check for external radiation. (Health and Safety Division, 1952)

Deposition Networks

The requirements for operating many stations for a nationwide and later a worldwide fallout deposition network led to the adoption of extremely simple collection systems. The initial device, which was a metal tray about 4 ft. on a side, required scrubbing down and transfer of precipitation and dry deposition to a bottle. This was obviously impractical for remote sites with untrained personnel. In the fall of 1951, Hanson Blatz of the Health and Safety Division suggested using self-adhesive label material to collect fallout and this was tested in comparison with the large trays during the October-November series (BUSTER-JANGLE). It seemed reasonably comparable and it was adopted for the Spring 1952 tests in Nevada (TUMBLER SNAPPER). One hundred and forty stations were set up at that time and the number ranged from about 100 to over 250 up until the time that the program was discontinued in 1959.

The large number of samples required automatic counters which were developed, based on passing a roll of samples over flat GM tubes, with the ashed samples sealed between two layers of vinyl tape. At the peak period the laboratory was processing over 600 samples per day.

The original purpose of gummed film sampling was to determine the day on which debris arrived at a particular site and the relative activity compared to other stations. It was quite satisfactory for this purpose, even though the absolute collection efficiency probably varied by a factor of two depending on precipitation. When the individual small continental tests were spaced far enough apart, it was even possible to do something with the data in terms of estimating individual nuclides by measuring decay and knowing the fission yield curves. With multiple tests, the system of estimation became much too complex and depended strongly on assumptions as to which tests had produced the debris. Nevertheless, the gummed film data was all that was available and calculations were made of strontium-90 deposition and of the external gamma dose based on the total beta activity of gummed film. (Harley and Hallden, 1960) These can only be considered as reasonable approximations and the system was discontinued in 1959 as more and more direct isotopic measurements on deposition became available.

The open pot was the most useful collector for many years and HASL made its first measurements with this sampling system in 1954. The network grew to 32 in 1958. A combination plastic funnel and column packed with ion-exchange resin was developed in 1959 that could be operated by untrained personnel and it was then possible to expand the network to over 100 stations. The total of pots and columns has remained in the range of 125 to 180 up until the present time. Almost all the analytical work was devoted to strontium-90 although measurements of strontium-89 were made during periods of high
fallout. Later, multiple pots or other large area collectors were set up for estimating the deposition of plutonium-238 and 239. Most of our present effort is devoted to obtaining more detailed information from a wet/dry collector which separates dry deposition from material deposited with precipitation.

Other groups were carrying out deposition measurements simultaneously, although at single stations or with more limited networks. These ranged from the plastic sheets on roofs used to collect rainfall at the NRL and Air Force Cambridge Research Laboratory sites to large sheet metal funnels set up at the AEC analytical contractors engaged in the fallout program. The larger collectors allowed the analysis for a number of radionuclides in individual rains and furnished useful data for interpretation of deposition mechanisms and atmospheric transport.

**Troy-Albany-Schenectady**

In April 1953, Prof. Herbert Clark of RPI measured very high deposition of radioactivity near the Institute. This was noted when the background of his GM counters went up by a factor of 3. Overall contamination levels were found to be at least equivalent to values found in Utah and Nevada following earlier tests. This was due to the high-speed winds coming from the test site coinciding with a violent high-level thunderstorm over the area 36 hours after the detonation. (Clark, 1954)

The Health and Safety Division tried an aerial survey for the first time using standard geiger and scintillation counters in a rented light plane. It was very obvious that the area of deposition covered the three cities and that the levels around the Tri-City area were many times higher than the surrounding countryside. It was noted that the radiation levels dropped sharply when passing over the Hudson River. This was unfortunate because it colored our thinking in later considerations of airborne measurement of deposited fallout. We were certain that fallout particles settled rapidly in water and would not be measurable at the surface.

**Surface Air Sampling**

Surface air was initially considered to be a very important possibility for human exposure. Air samples were taken following the early Nevada tests at a few fixed locations and of course were taken at the mobile locations in the 200 to 500 mile zone. The high volume samplers used at the time however did not lend themselves to network operation since they required considerable maintenance. The first well-developed surface air network was that set up by the Naval Research Laboratory in 1957. Most of the stations were distributed along the 80th meridian (West) and samples were analyzed for natural activity as well as fission products. (Lockhart, 1964) Their positive displacement pumps operated continuously for years at 1 m³/min without appreciable maintenance. This program was turned over to HASL in 1963 and the network has been considerably extended since then. Comparable stations are now being operated at the remote sites set up by the National Oceanographic and Atmospheric Administration (NOAA) for their program of geophysical monitoring for climatic change.
The overall conclusion that inhalation was relatively insignificant for long-range fallout as compared to ingestion seems correct. The only modification has been in the case of plutonium where inhalation is the main contributor to body burden. In this case, data were only available from 1965, but earlier levels could be reconstructed from known Pu/Sr-90 ratios. (Bennett, 1974)

Soil Sampling

In many cases it was necessary to assess the total deposit of strontium-90 or other radio-nuclides after the fact for inventory purposes or to cover areas where rate measurements had not been made. Our first efforts in soil sampling were part of a pasture site program designed to look at soil, plants and animal tissues as a means of studying mechanisms. This was begun in 1954 and continued at various levels of effort for about 10 years. Only rather crude estimates of the mechanisms were obtained, but useful relationships were developed for prediction models.

The measurement of soils as an indicator of total strontium-90 deposit was begun in the U. S. in 1955 and a limited worldwide survey was made in 1956. Most of the overseas samples were collected by Lyle Alexander of the U. S. Department of Agriculture and he carried out more extensive surveys in 1958 (Alexander, et al, 1961) and 1960 (Hardy, et al, 1962). A supposed final worldwide survey was made in 1966 but another sampling was required in 1970 to estimate the deposition of plutonium-238 from the SNAP 9A failure in 1964. (Hardy, et al 1972)

These soil sampling efforts were successful in showing the distribution of strontium-90 and later plutonium around the world. It was amazing how well the data from at most 150 locations fit into a recognizable pattern showing highest deposition in the mid-latitudes of the northern hemisphere with the corresponding zone in the southern hemisphere being about one third of the maximum and the equatorial and polar regions tapering off to very low values. HASL has always contended, however, that soil sampling is only indicated for after-the-fact measurements since deposition collectors yield samples that are much easier to analyze and much easier to evaluate.

Fallout over the Oceans

Over two thirds of the earth's surface is covered by the major oceans and the comparability of fallout on land and on water has frequently been questioned. Actual measurements of sea water both at the surface and at depth by Bowen of the Woods Hole Oceanographic Institution indicated that deposition per unit area was greater over the ocean. This caused some problems when working with the inventory of strontium-90 based on production, stratospheric reservoir and land deposition, since there appeared to be more Sr-90 than had been produced. This has never been completely resolved although there was some indication that high oceanic deposition does not extend to the total water area. (NAS, 1971)
Attempts to resolve this question led to installation of fallout collectors on U. S. Coast Guard weather ships on the Atlantic, to an intensive study of a large stable lake in Oregon and to constructing a small "island" to house a sampling station on the Bahama Banks. The ship's stations showed that air concentrations were the same as those at land based stations at the same altitude. (Volchok, 1970a) The Crater Lake study in Oregon showed that the total deposition was apparently the same as on adjoining land areas. (Volchok, et al, 1970b) The HASL Dolphin experiment on the Bahama Banks showed that the situation was more complicated than hoped for but suggested that fallout over the oceans was indeed greater than over land surfaces. (Volchok, 1971)

The major studies of oceanic fallout and of radioactivity applied to marine studies in the U. S. have been those of Bowen, of Broecker at Lamont and of Folsom at Scripps. These are also described in the NAS volume referenced above.

External Gamma Exposure

The estimates of external gamma dose from measurements of total beta activity on gummed film left much to be desired. Once specific radionuclides other than strontium-90 were measured some improvement was possible. The first technique was to use strontium-89 to 90 ratios as an indication of the age of the debris. The fission yield curves could then be used to estimate the deposition of gamma emitters and simple correction factors for weathering and shielding could be included. A second technique was to use the limited multi-nuclide data from our analytical contractor sites in Westwood, N. J. and Pittsburgh to estimate the gamma emitter deposition. These calculations were reported by the United Nations Scientific Committee on the Effects of Atomic Radiation. (UNSCEAR, 1966)

Very few direct measurements of gamma exposure were made at long distances from the test sites. Actually, weapons fallout was considered an annoying interference in the measurement of natural background radiation and our knowledge of the natural background field was not really sufficient to allow obtaining fallout by difference. The best efforts along this line were those of Gibson in the United Kingdom (Gibson, 1969) although data are available from Japan, Sweden and the United States. (UNSCEAR, 1972) Later work at HASL was largely based on field gamma spectrometry to distinguish between the various classes of emitters. (Beck, 1966)

The SUNSHINE Project

In the early considerations of health effects of fission products it was decided that strontium-90 was the critical radionuclide and estimates of the permissible amount of weapons testing were based on the distribution of strontium-90 in man. The first unclassified report on this subject was issued in 1956 by the RAND Corporation. (RAND, 1956) This was basically a report of a meeting of experts held in August 1953 to review earlier estimates made under an earlier project, GABRIEL. The estimate at that time was that
more than $2.5 \times 10^4$ megatons of fission would be required to give an average body burden of 1 microcurie in the world population. The actual weapons yields have been about 1% of that value and the body burdens produced have been about 0.1% of the 1 microcurie.

It was also at this meeting that the unfortunate code name of project SUNSHINE was adopted for the strontium-90 program.

At the time of the meeting only J. Lawrence Kulp of Lamont Geological Observatory and Willard F. Libby of the University of Chicago had data. The NYO Health and Safety Division program had just begun method development and routine analyses of milk, for example, did not begin until 1954 along with the first analyses of deposited strontium-90 collected in pots.

The burden of sampling and analysis continued to increase during the 50's. The Chicago group under Libby and later under Martell concentrated on measurement of stillbirths and a few deposition samples. The Lamont group also analyzed a number of bones but did carry out a wider spectrum of sampling. In 1956, the procedure of sending out the bulk of the samples to commercial analytical contractors was begun. This has continued to the present.

**Data Reporting**

The early fallout data where total beta activity was measured had been reported in the open literature for some time. (Eisenbud and Harley, 1953, 1955, 1956) On the other hand the so-called "SUNSHINE Reports" were all classified and only summaries of the data were presented. (Libby, 1956) This lack of publication was a major topic in the 1957 Congressional Hearings on Fallout and this led to the development of the HASL Quarterly Reports on Fallout with the first one appearing in 1958. The overall reporting of routine data was later transferred to the Public Health Service and they began their series of Radiological Health Data and Reports which continued up until 1974.

The 1957 Hearings also led to the declassification of many earlier reports on fallout distribution. These generally required only deletion of specific information on individual tests. While many of the reports were confined to close-in fallout, a few contained data on long-range deposition (e.g. NYO-1576 (Del), 1952; NYO-4505 (Del.), 1953; NYO-4512 (Del.), 1953; NYO-4552 (Del.), 1954; etc.). These reports were prepared by Dr. Machta's group at the U. S. Weather Bureau and by the staff at NYO.

**Human Intake**

Early in the SUNSHINE Program, the chemistry of strontium as an analog of calcium took a strong hold on everyone's thinking. Strontium-90 was sought in all high-calcium foods particularly milk, HASL began milk measurements in 1954, but did not realize the importance of total dietary intake until 1958, when 19 representative food categories were selected so that typical urban diets could be evaluated. The same categories have
been analyzed for cesium-137 by Argonne since 1961. (Karttunen, 1976) With both the Sr-90 and Cs-137, the time trends of radioactivity have been very helpful in indicating mechanisms of contamination, and in predicting body burdens of the two radionuclides. (Gustafson and Miller, 1969; Bennett, 1975)

Plutonium in diet has only been measured at HASL since 1972, but its significance as compared with inhalation seems slight. On the other hand, the intake of iodine-131 during the early continental tests may have been very significant, but our early attempts to measure it were unsuccessful. Even in 1957, a major iodine-131 experimental monitoring program was a failure. Fortunately, the advent of good gamma spectrometry made iodine-131 measurement in milk (the major source of intake) very simple.

At about the same time as the Tri-City Diet Program was beginning, PHS started an Institutional Diet Program, and FDA began monitoring individual foodstuffs sampled at the source. These were complementary to the HASL program, but the most useful data came from the PHS milk networks. Their measurements included Sr-90, Cs-137 and I-131, and were published in Radiological Health Data and Reports.

**Body Burdens**

As mentioned above, a number of the national laboratories (ANL, BNL, LASL) measured Cs-137 body burdens from fallout in humans with their whole body counters. This work was summarized by Gustafson and Miller (1969). A larger number of whole body counters were utilized for this work beginning in about 1964, but the comparability of data was plagued by calibration problems. A useful by-product of the program was the considerable amount of data on body potassium content that was generated.

Measurements of iodine-131 in human thyroids were less numerous, and the work at NYU (Eisenbud, et al, 1961) is the most widely quoted.

Strontium-90 is a bone seeker, and the analyses can only be performed on autopsy specimens. Libby, in the early days of the SUNSHINE Program relied on stillbirths, while Kulp obtained adult specimens from New York City. He made the first measurements on variability within the skeleton (a factor of 4 between femur and vertebra) which allowed normalization of data when different bones were analyzed. Their work was reported in a number of articles. (e.g. Libby, 1957; Kulp, 1960) The HASL program, beginning in 1956, was designed to measure the variation of body burden as a function of age and to relate the levels found to dietary intake. The success of this work was described first by Rivera (1969), then by Bennett (1975).

The major work on plutonium body burden from fallout has been carried out at LASL. (Campbell, et al, 1973) Their data have been shown to agree with estimates of intake and the ICRP model for plutonium metabolism. (Bennett, 1974)
Thermonuclear Testing

Thermonuclear tests at the Pacific Proving Ground were a definite challenge because of the large area that we believed might be subject to significant fallout. For the IVY tests in the fall of 1952, we attempted to do evaluation of radioactivity on the islands by measurements with an in-flight gamma scintillation counter. The equipment, which recorded radiation levels on magnetic tape along with a voice recording of location, was tested in Nevada following the continental tests in the fall of 1951. For IVY, aircraft were made available by the Navy on Kwajalein, Hawaii and Guam. A series of flights were carried out from these three bases following each of the two IVY tests. The levels of course were higher in the Marshalls and other nearby island chains but measurable activity was also found in the flights from Guam.

The measurements indicated that additional monitoring should be available for the CASTLE series in the spring of 1954. Automatic monitors for external gamma radiation and for airborne beta activity were prepared and set out on a number of islands in the Marshalls. This equipment turned out to be the means of saving many lives at the time of the BRAVO shot on March 1st.

The HASL program was designed to extend coverage outside of the immediate test area around Bikini and Enewetak atolls, which was the exclusive responsibility of the Test Director, to include the Trust Territory and beyond. Organizationally, HASL personnel were attached to Joint Task Force 7 for support and liaison but reported directly to the Commander in Chief, Pacific (CINCPAC) and to USAEC Headquarters.

Monitoring was performed by two complementary methods, a network of manned ground stations that reported gamma radiation data periodically by radio, and aerial surveys by Navy patrol aircraft over specified islands following each detonation. There were 18 ground stations in all, concentrated chiefly in the Marshall, Caroline, and Marianna Islands but extending to the Phillipines, Japan, Hawaii, and the Aleutians. Gamma readings were transmitted several times a day from monitoring instruments provided by HASL. In addition, the stations within a radius of 1500 nautical miles from the test area were equipped with automatic recording monitors that documented the time of occurrence and magnitude of abnormal gamma intensity. On receiving a report of significant increase of gamma level at any station following a test detonation, the HASL project officer dispatched a survey aircraft to the appropriate island group. The patrol aircraft, operating from Kwajalein, Guam and Oahu, were equipped with sensitive gamma detectors of HASL design which enabled them to estimate fallout at ground level from an altitude of two hundred feet during brief passes over each atoll. The measurements were relayed in flight by radio to the Task Force, giving the HASL project officer prompt information about the extent and intensity of fallout.
The most dramatic example of the effectiveness of this dual arrangement occurred on the day of the very first detonation, BRAVO. During the late afternoon, the automatic monitor on Rongerik, 120 nautical miles east of Bikini, went off scale at 100 mR/hr. This information, received at the Task Force Headquarters in less than half an hour, was the first indication of high fallout levels outside of the ships of the task force and Bikini atoll itself where the detonation occurred. The Navy patrol squadron on Kwajelein was notified to execute a survey pattern over the northern Marshall Islands including Rongerik. Due to communications difficulties, the flight was delayed until the following day when in-flight reports indicated hazardous levels of fallout on Rongerik and neighboring atolls. By then, personnel on Rongerik were already being evacuated and based on the air survey reports, plans were implemented to evacuate Rongelap and Ailinginae. Actual doses received were in the range of 100-200 rad. Within a matter of days, aerial surveys had been completed in the Marshalls, Carolines, Marianas, Gilbert, and Hawaiian Islands from which the extent and severity of fallout was clearly defined.

The March 1st incident made it obvious that additional rapid monitoring over a broad area was required. The aerial survey system was believed to be adequate for checking out islands but it was felt necessary to have additional locations in between the islands. The first system tested was to lay an oil slick on the water as a trap for deposited fallout. This was tested off the New Jersey Coast and it appeared that a relatively small area would be adequate. This changed our thinking to consider distributing rafts by dropping them from aircraft. This also was tried just outside New York Harbor and was partly successful. Due to the pressure of time it was decided to risk the large scale deployment of rafts before the next test. This project was appropriately named DUMBO, since in operation almost all of the rafts disintegrated before hitting the water. Fortunately when the aircraft were sent out after the test to check the islands and any remaining rafts it became apparent that no fallout collector was needed. The distribution of radioactivity in the water could be measured for several days. (Breslin and Cassidy, 1955)

The fallout from the March 1st test had contaminated a number of the Marshalese on Rongelap and the crew of the Japanese fishing boat Fortunate Dragon. NRDL and HASL were involved in the measurement of radioactivity in the urine samples from the Marshalese over a period of several weeks. These were mostly total fission product measurements and only a few strontium-90 analyses and one iodine-131 determination were made. It was a source of continuing wonder to the laboratory staff that these hundreds and hundreds of urine samples could all be sent in in liquor bottles but it was apparent that the Joint Task Force contingent on the islands was very helpful in providing empty bottles.

The situation with the Japanese fishermen was of course more serious since their exposure had not been reduced other than by washing down the vessel.
The aftermath of CASTLE series in the spring of 1954 included a great deal of public reaction in Japan. Their Meteorological Service in particular sampled the area around Japan and reported elevated beta activity in the water. Based on their data, Allyn Vine at the Woods Hole Oceanographic Institution worked out that the radioactivity deposited on the ocean surface had probably followed the currents towards Japan and the following year it would be in the more Northern Pacific in the Japanese Current. To test this hypothesis, Operation TROLL was set up for the spring of 1955. A group of scientists with sampling and counting equipment operated from the U. S. Coast Guard Cutter Taney and followed the track of radioactivity for seven weeks. Most of the measurements were surface water but depth samples were taken every few hundred miles and biological specimens were measured whenever they became available. (Harley, Ed., 1956)

A gamma scintillation probe was also tested during the cruise. This was a large volume plastic scintillator with photomultiplier and associated electronic equipment housed in a stainless steel shell. This was designed to be towed behind the vessel at all times and to transmit the radiation reading by cable to the recorder on shipboard. This was not completely successful but a later improved version was used by the University of Washington Applied Fisheries Laboratory group following the REDWING test series in 1956. (Seymour, et al, 1957) A parallel development by Riel of the Naval Ordnance Laboratory has been used extensively in looking at cesium-137 in estuaries (Riel, 1975), as well as for ocean fallout. (Riel, 1965)

**Stratospheric Inventories**

It was apparent that the thermonuclear tests, starting with IVY MIKE in 1952, carried the radioactive debris well up into the stratosphere. Dr. Libby in 1953 proposed that the stratospheric storage would have a half life of perhaps 10 years which would reduce exposure from strontium-90 fallout. This was disputed later by Machta and Martell and it appears now that the half life is of the order of 1 year, with equatorial tests giving a longer residence time and arctic tests giving a shorter residence time.

The first actual measurements of activity in the stratosphere were with the ASHCAN balloon program in 1956. Sampling equipment was designed by General Mills and the actual flight operations were carried out by the Air Force. The balloons reached altitudes of over 100,000 ft. and this altitude was later extended to 140,000 ft. HASL was made responsible for the analytical phase of the high altitude balloon sampling in 1961.

The aircraft sampling up to 70,000 ft. with U2 aircraft was initially set up by the Defense Atomic Support Agency. Responsibility for the analytical and interpretive phases of the high altitude sampling program (HASP) was assigned to Isotopes, Inc. The HASP program ran from 1957 through 1960. (Friend, Ed., 1961) With the resumption of testing in 1961, DASA set up the STARDUST program utilizing the RB-57F aircraft. (Feely, et al, 1967) This program continued until 1967 when the work was assigned to HASL and the program was renamed AIRSTREAM. (Krey, et al, 1975)
The two stratospheric programs allowed us to maintain a balanced inventory for strontium-90 and to predict deposition at least a year in advance. The distribution of the debris and the isotopic ratios in various samples allowed us to develop models of stratospheric transport and the transfer of material to the troposphere. This was particularly useful in the cases where tracers such as tungsten-185 and cadmium-109 were produced in individual tests. The most striking application of stratospheric data was to the burn up of a SNAP 9A satellite containing 17 kilocuries of plutonium-238 at 150,000 ft. This occurred in April 1964 and the Pu-238 was first detected in the balloon samples taken in the Southern Hemisphere in the late summer. (Salter, 1964) As the material came down into aircraft sampling altitude it became possible to develop an inventory and to show that the Pu-238 source had burned up completely and was present as particles with the mean diameter of the order of $10^{-7}$ centimeters. (Krey, 1967) Soil measurements later were found to account for the Pu-238 as it deposited with a half residence time in the order of a year. (Hardy, et al, 1972)

Other atmospheric data became available through tropospheric sampling with B-57 aircraft from time to time. These data were of most interest in tracking debris from individual tests rather than inventories since concentrations in the troposphere were normally considerably lower than those in the stratosphere. The carbon-14 and krypton-85 data were particularly useful. (Telegadas, 1971; Telegadas, 1975)

Summary

The overall program of work on long-range radioactive fallout from nuclear tests has been an interesting example of scientific response to a problem. Naturally, the bulk of the studies in the United States were sponsored by either the Atomic Energy Commission or the Defense Department. In other countries, the work was also usually government-funded. Under these circumstances, it is surprising that the data came into public view as rapidly as they did. Once the general situation became known, the scientific community was presented with an enormous "tracer experiment" that led to a number of significant advances.

In the physical sciences, meteorological concepts could be tested, oceanographic current movements could be monitored and instrumentation developments in low-level counting, spectrometry and external gamma exposure measurements were numerous. On the biological side, information on the metabolism of strontium, cesium and even potassium and calcium became available and the whole field of radioecology blossomed. These were responses to a need, for the most part, but were made possible through the radioactivity introduced into the environment. Thus, although fallout was not a "good thing", it was possible to take advantage of its existence.

The nuclear fission tests in the Pacific and Nevada produced only about 0.1 MCi of Sr-90 and even the thermonuclear tests carried out before the test moratorium brought the yield to less than 10 MCi in 15 years of testing. Then in 1961-62, another 10 MCi was produced. Thus it is not surprising that fallout peaked in 1963, with the highest levels in
diet and man occurring a year or two later. Since the test ban treaty, the contributions to world-wide contamination have been less than 10% of the total. Thus the stratospheric inventories and fallout rates have fallen off to relatively low values. The total deposit on the earth’s surface is decaying away slowly with the half-life of Sr-90 and levels in diet and man are declining at intermediate rates. Many of these quantities should be followed a few years longer to help fill out our knowledge of transfer mechanisms but there is no public or government pressure for further studies.

After 25 years, it should be possible to sum up the biological damage caused by the weapons testing program, however, we still lack the key element - the effect of low-level radiation on man. Some damage may have occurred but it has not been detected. A great deal of money and a tremendous scientific effort has produced many valuable results but not a final answer.

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Cesium-137 and Potassium in Whole Milk Supply of Bombay City

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A B S T R A C T

Naturally occurring potassium-40 and man made cesium-137 constitute the two important sources of gamma activity in milk and milk products. Since milk is an important source of uptake of cesium-137 by human beings and is the major diet of growing children, its measurements on a country wide scale was started at the Bhabha Atomic Research Centre, Bombay, in 1958. With the cessation of large scale testing of nuclear weapons in the atmosphere, the levels started decreasing after 1964 and hence this extensive monitoring was substituted with limited monitoring program. The results of these measurements for the period 1969-1973 are presented for some of the milk sampling stations supplying milk to the city of Bombay. The parameters responsible for some of the variations observed in $^{137}\text{Cs}$ levels are discussed. The results of these measurements are used to assess daily intake of $^{137}\text{Cs}$ for people of Bombay in relation to health hazard.
Nuclear weapon testing programs have introduced radioactive materials into the environment on a global scale. Some of the short-lived isotopes from fallout are disappearing rapidly from the man's environment due to radioactive decay. However, some long-lived isotopes like strontium-90 and cesium-137 are still present in our environment. These isotopes find their way to plants, animals and to man due to their metabolic similarity to essential elements calcium and potassium. Continuing measurements of cesium-137 levels in milk and diet present one of the simpler methods of assessing the long-term radiation hazard to man from radioactive fallout. In this short communication, results on cesium-137 in milk are presented for 1969-1973 period. Milk constitutes major diet of growing children and is an important constituent of food for adults. Hence it is given special attention in our fallout monitoring program.

**SAMPLING AND MEASUREMENT PROCEDURES**

Ten litres of liquid milk samples are collected from Bombay(Aarey), Anand and Dhulia dairies on monthly basis. These dairies are responsible
for major supply of milk to Bombay city. Sporadic samples are also collected from Kolhapur, Miraj, Nasik, Ratnagiri and Solapur, as these places also contribute to Bombay's milk supply. These samples are ashed below 400°C (to minimize losses of $^{137}$Cs) and the ash, put in standard containers (diameter 6.5 cm. x height 7.5 cm), is directly counted for $^{137}$Cs and $^{40}$K using a 12.5 cm x 10 cm NaI(Tl) crystal coupled to 256-channel pulse height analyser. The measurement procedures are described in detail in our earlier publications$^{(1,2)}$.

RESULTS

Fig. 1 gives the levels of $^{137}$Cs in Bombay and Anand milk on monthly basis for 1969-73 period. The insert in Fig. 1 gives average annual values of $^{137}$Cs in milk from 1958 onwards for Bombay and 1960 onwards for Anand. This insert shows the nature of $^{137}$Cs variations in milk since the inception of the program as given in our earlier publications$^{(1,2)}$. Table 1 lists the annual average values of $^{137}$Cs in milk from all the places mentioned above. The levels of cesium-137 are reported in pCi of $^{137}$Cs per gram of potassium in milk. The yearly average of potassium content in milk in table 1 is given in grams of potassium per litre of milk. Monthly variations of $^{137}$Cs content in Dhulia milk follow the pattern of variations in $^{137}$Cs content of Bombay and Anand milk (Fig.1).

DISCUSSIONS

Since the commencement of the sampling program in 1958, the levels of $^{137}$Cs content in milk were the highest during the period 1963-1964
following the high yield nuclear weapon tests carried out in 1961-1962\(^{(2)}\). The highest annual average levels were 28.6 pCi \(^{137}\text{Cs}\)/g.K and 11.7 pCi \(^{137}\text{Cs}\)/g.K in Bombay and Anand milk samples respectively. By 1968 end, the \(^{137}\text{Cs}\) levels dropped down to 3.7 and 2.2 pCi/g.K in milk for these two places (Fig. 1 insert). The content of \(^{137}\text{Cs}\) in milk was lowest in the year 1969 during the 1969-1973 period (table 1). The levels of \(^{137}\text{Cs}\) content in milk started rising from 1970 onwards. This has been explained on the basis of fallout from recent nuclear weapon tests by China and France as follows.

Fallout measurements at Bombay show that nearly 80-90\% of the annual deposition of fallout \(^{90}\text{Sr}\) activity takes place during rainy season (June-October)\(^{3}\). This explains the higher levels of fallout \(^{137}\text{Cs}\) found in milk during rainy season as \(^{137}\text{Cs}\) deposition also follows similar trends. French nuclear explosions have taken place during the months of June to September of every year during 1970-1973 period. Study of short lived radionuclides in surface air at Bombay has indicated the arrival of fallout from these tests over the west coast of India within two to three weeks\(^{4}\). It has also been shown that the levels of activity on the west coast of India are an order of magnitude higher than at other areas of the northern hemisphere. Comparison with the activity from the Chinese tests of northern hemisphere (40°N) shows that the levels on the west coast of India are comparable to other areas of the northern hemisphere\(^{(4,5)}\). Thus rapid transport of air masses across the equator by the southerly jet during south west monsoon\(^{(4)}\) and the time coincidence of French weapon testing
program with monsoon resulted in higher deposition of radioactive fallout from these tests. When the data given in Fig. 1 is compared with data on $^{90}$Sr levels in New York milk (6) or data on $^{90}$Sr and $^{137}$Cs levels in milk from country wide survey in U.K (7), it is found that $^{137}$Cs levels in milk are slowly rising during 1970-1973 period while those in New York or U.K. milk are steadily decreasing. This can however, be explained on the basis of higher fallout received by west coast of India from French nuclear explosions when compared to other places in the northern hemisphere (5). All the milk sampling stations supplying milk to Bombay city are in the west zone of India. This is also the reason for somewhat higher levels of $^{137}$Cs in milk from Kolhapur, Nasik and Poona in 1970 as these stray samples happened to be collected during rainy season. Similarly somewhat higher levels observed during January-March period of 1969, 1970, 1971 and 1972 have been attributed to fallout from Chinese nuclear explosions on December 27, 1968, September 29, 1969, October 14, 1970, November 18, 1971 and January 7, 1972. The values of $^{137}$Cs concentration in milk from Anand and Dhulia are somewhat lower than those for Bombay due to lower rainfall at these places as compared to Bombay.

Average daily consumption of milk and milk products per adult is about 100 ml for Bombay (8). On this basis, the daily intake of $^{137}$Cs through milk and milk products range between 0.1 - 1.8 pCi. This is negligible in comparison with the limit of 44000 pCi of $^{137}$Cs per day recommended for daily intake for individual members of the population at large (9).
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<td>1972</td>
<td>2.60</td>
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<td></td>
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<td>1973</td>
<td>5.40</td>
<td>1.15</td>
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<td>DHULIA</td>
<td>$20.9^\circ N$, $74.8^\circ E$</td>
<td>58.4</td>
<td>1970</td>
<td>1.97</td>
<td>1.32</td>
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<td></td>
<td></td>
<td></td>
<td>1971</td>
<td>3.96</td>
<td>1.40</td>
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<td></td>
<td></td>
<td></td>
<td>1972</td>
<td>3.96</td>
<td>1.35</td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>1973</td>
<td>3.63</td>
<td>1.03</td>
</tr>
<tr>
<td>KOLHAPUR</td>
<td>$16.7^\circ N$, $74.2^\circ E$</td>
<td>104.0</td>
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<td>7.65</td>
<td>1.25</td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>1971</td>
<td>4.47</td>
<td>1.18</td>
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<td></td>
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<td>1972</td>
<td>2.32</td>
<td>1.54</td>
</tr>
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<td>NASIK</td>
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<td>76.2</td>
<td>1970</td>
<td>5.31</td>
<td>1.54</td>
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<td>$18.3^\circ N$, $74.0^\circ E$</td>
<td>71.5</td>
<td>1970</td>
<td>4.09</td>
<td>1.28</td>
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<tr>
<td>RATNAGIRI</td>
<td>$17.0^\circ N$, $73.3^\circ E$</td>
<td>254.0</td>
<td>1971</td>
<td>3.35</td>
<td>1.08</td>
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<td>SOLAPUR</td>
<td>$17.5^\circ N$, $75.9^\circ E$</td>
<td>72.3</td>
<td>1971</td>
<td>4.30</td>
<td>1.42</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>1972</td>
<td>5.82</td>
<td>1.02</td>
</tr>
</tbody>
</table>
FIGURE 1. CESIUM-137 LEVELS IN BOMBAY AND ANAND MILK SAMPLES.
ABSTRACT

The paper presents results of gamma spectrometric measurements of natural and fallout radioisotopes in composite Indian diet. The details of sampling and measurement procedures are also given. The natural radioactivity due to potassium-40, radium-226 and thorium-228 and cesium-137 of fallout origin are reported in this paper and the assessment of daily intake of these isotopes has been made.
INTRODUCTION

The radioactivity in the diet of a man consists of natural radiations due to the two radioactive series with uranium-238 and thorium-232 as parents, potassium-40 and long lived radioisotopes introduced in the biosphere by fallout from nuclear weapon tests. The authors' laboratory had started the program of fallout measurements in environment in 1958. The data obtained since then has been published in various reports from this centre. The decline in weapon testing program since test moratorium in 1962 has resulted in significant decrease in fallout radioactivity after 1968. This enabled us to devote more time to the measurements of natural radioactivity in food-stuffs by using simple technique of gamma-ray spectrometry. The study of the natural levels of radioactivity in diet and the manner in which it varies, helps in understanding the environmental influences of radioactivity produced by nuclear era vis-a-vis natural radioactivity. This paper gives the results of measurements on radioactivity of natural and fallout origin on composite diet samples (called as "thali" in local language or "lunch pack") collected during 1970-1974 period.

SAMPLING AND MEASUREMENT PROCEDURES

The composition of the sample and the average weight are given in the beginning of tables 2 and 3. Two such composite
meals in a day is the major diet for an average Indian of poor and lower middle class people (As regards economic standards). "Thali" samples were collected from two canteens - one in the heart of city and another at this Research Centre. Ten "Thali" samples were collected each canteen on quarterly basis. The samples were ashed at about 350°C to minimize losses of $^{137}$Cs during ashing. After ashing, the samples were transferred to plastic containers (7.5 cm. in diameter and 6.5 cm. in height) and stored air-tight for minimum period of one month to allow for the equilibration of radon and thoron daughter products with their parents radium-226 and thorium-228.

The sources have been prepared using calibrated solutions of various isotopes and mixing them in a known weight of AnalaR sodium oxalate to approximate ash matrix of the sample. Thorium source was prepared by mixing thorium nitrate powder with sodium oxalate. The source for $^{40}$K was 129.9 gms of AnalaR potassium chloride. The sources for $^{226}$Ra and $^{228}$Th were also stored "air-tight" in plastic containers for minimum period of one month.

The samples and standards were counted in identical containers placed concentrically on a 12.5 cm.x 10.0 cm. NaI(Tl)integral assembly housed in a 15 cm. thick lead shield lined with cadmium and copper. The detector was coupled to a 256-channel pulse height analyser. All the "thali" samples were counted for 1000 minutes. The energy regions used for the estimation of various isotopes, the conversion factors
(concentration of the isotope/cpm due to the isotope in its region as derived from standard sources) and the minimum detectable levels are included in the table 1. The energies used for estimation of $^{228}\text{Th}(2.62 \text{ Mev})$ and $^{226}\text{Ra}(1.76 \text{ Mev})$ and the analyser calibration for $0.2-3.0 \text{ Mev}$ were similar to those used by others (1). As the ratio of $^{40}\text{K}$ content to $^{226}\text{Ra}$ or $^{228}\text{Th}$ content in food-stuffs was very high compared to that in soils, it was not possible to use the $1.6 \text{ Mev}$ calibration used advantageously for soil samples (2). The $0.2-3.0 \text{ Mev}$ calibration used in present work avoided the interference of Compton contribution of predominant $^{40}\text{K}$ in $^{226}\text{Ra}$ and $^{228}\text{Th}$ regions. The concentrations of $^{228}\text{Th}$, $^{226}\text{Ra}$, $^{40}\text{K}$ and $^{137}\text{Cs}$ were determined in the energy regions by means of the following equations:

\[
\begin{align*}
C_4 &= T_{2.62} \\
C_3 &= T_{1.76} - 0.81C_4 \\
C_2 &= T_{1.46} - 0.89C_4 - 1.01C_3 \\
C_1 &= T_{0.66} - 5.11C_4 - 3.39C_3 - 0.30C_2
\end{align*}
\]

Where $C_4$, $C_3$, $C_2$ and $C_1$ represented actual counts in their respective energy regions due to $^{228}\text{Th}$, $^{226}\text{Ra}$, $^{40}\text{K}$ and $^{137}\text{Cs}$. $T_{2.62}$, $T_{1.76}$, $T_{1.46}$ and $T_{0.66}$ were total counts (corrected for background) observed in the spectrum in the above respective energy regions of $^{228}\text{Th}$, $^{226}\text{Ra}$, $^{40}\text{K}$ & $^{137}\text{Cs}$. The total counts were used for estimation of the concentrations of the above isotopes if and only if they were above the level of one standard deviation in the background in their respective energy region.
RESULTS AND DISCUSSIONS

The results of the above study are given in Tables 2 and 3. All the concentrations of isotopes are reported in picocuries per kilogram of "Thali" weight. The overall errors in the results are about ± 20% except for $^{40}$K where it is less than ± 10%. Table 4 gives average daily intake of natural and fallout radioactivity in picocuries as calculated from the data given in tables 2 and 3.

It is known that $^{40}$K is the predominant radioactive component in normal foods and human tissue. Tables 2, 3 also show that the major intake of radioactivity is from $^{40}$K. It enters human body mainly through food-stuffs and is under close homeostatic control. Variations in the composition of diet have some effect on value of $^{40}$K in the diet. The $^{40}$K content in "thali" has varied between 1100 pCi to 2200 pCi per Kg. of "thali" weight. This approximates to an intake of 1250 to 3600 pCi per day on the basis of two such meals per day. This is equivalent to 1.5 to 4.2 g of potassium/day through diet and 0.2 g K through milk and milk products based on its average consumption of 100 ml/day for Bombay. The daily intake of potassium reported in literature for some other places in the world is 1.4 to 6.5 g. Large variations were observed in $^{226}$Ra and $^{228}$Th content of "thali". The levels of $^{226}$Ra varied between 0.7 pCi to 11.2 pCi per Kg. of "thali" weight and those for $^{228}$Th between 0.7 pCi to 11.2 pCi. The mean daily intake of
226Ra for 1970-1974 period was 4.5 pCi/day and that for 228Th for the same period was 3.9 pCi/day (Table 4). It is also evident from tables 2 and 3 that the variations in 226Ra and 228Th content in "thali" samples collected from canteen located in B.A.R.C. campus are more when compared to those in "thali" samples collected from city area. Average content of these two isotopes in B.A.R.C. thali is also more than that in the samples collected from city area for the period 1970-1974. However, all these daily intakes are much lower than the recommended limits for individual members of the population at large (Table 4)5.

Cesium-137 was the only isotope of fallout origin that could be detected in some of the "thali" samples. The levels of 137Cs varied from the detection limit to about 12 pCi per Kg. weight of "thali" samples. Analysis of "thali" samples for Strontium-90 content gave variations of 2.0 pCi to 14.0 pCi of 90Sr per Kg. of "thali" for 1970-1973 period (6). Wheat and rice constitute 45% of the "thali" composition. The supply of these food-grains to Bombay city is at times made from stocks received from countries like America, Canada, Egypt and Australia which have widely different fallout distribution patterns. Some of the imported wheat samples were analysed during January 1971, April 1971 and July 1972 and had 137Cs levels of 12-14 pCi/Kg. of wheat and this could be the reason for higher values of 137Cs levels in April 1971, July 1971 and July 1972 "thali" samples (7). However, the
levels of $^{137}$Cs in composite diet samples during the measurement period were much below the average level of 44000 pCi/day prescribed for daily intake of this isotope for any period of 12 months (5).

ACKNOWLEDGEMENTS

The authors are grateful to Dr. K.G. Vohra, Head, Division of Radiological Protection and Dr. U.C. Mishra, Head, Air Monitoring Section, for their valuable suggestions and keen interest in the present work. Authors are also grateful to Shri S. Sadasivan for cooperation given in Standardization procedures.
REFERENCES


### Table 1

**Prominent Gamma Energies of the Isotopes Analyzed and the Details of the Analyser System Used**

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy used for estimation (MeV)</th>
<th>'Energy regions' for the present work (MeV)</th>
<th>Concentration per Cpm in the region (pCi)</th>
<th>Minimum detectable limit (pCi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs+ $^{137m}$Ba</td>
<td>0.662</td>
<td>0.54 - 0.76</td>
<td>4.00</td>
<td>0.80</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>1.460</td>
<td>1.28 - 1.58</td>
<td>97.80</td>
<td>15.45</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>1.760</td>
<td>1.62 - 2.02</td>
<td>36.92</td>
<td>4.00</td>
</tr>
<tr>
<td>($^{214}$Bi)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>2.620</td>
<td>2.42 - 2.77</td>
<td>67.23</td>
<td>5.00</td>
</tr>
<tr>
<td>($^{208}$Tl)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

I - 39
**TABLE-2**

**NATURAL AND FALLOUT RADIOACTIVITY IN COMPOSITE DIET**

**SAMPLES: SAMPLES COLLECTED AT BARC CANTEEN**

Composition of diet sample: Rice-100 g., Pulses-100 g., Wheat-150 g., Vegetables-100 g., Curd-75 g., Miscellaneous-25 g., Average weight of a Thali- 570g.

<table>
<thead>
<tr>
<th>Month</th>
<th>$^{137}$Cs (pCi/Kg)</th>
<th>$^{226}$Ra (pCi/Kg)</th>
<th>$^{228}$Th (pCi/Kg)</th>
<th>$^{40}$K (pCi/Kg)</th>
<th>Potassium content (g/Kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>JUNE, 1970</td>
<td>-</td>
<td>6.6</td>
<td>3.5</td>
<td>1957</td>
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<td>JULY,</td>
<td>3.0</td>
<td>0.7</td>
<td>2.4</td>
<td>1544</td>
<td>1.84</td>
</tr>
<tr>
<td>AUGUST,</td>
<td>-</td>
<td>-</td>
<td>3.6</td>
<td>1910</td>
<td>2.28</td>
</tr>
<tr>
<td>OCTOBER,</td>
<td>-</td>
<td>3.0</td>
<td>1.6</td>
<td>1500</td>
<td>1.79</td>
</tr>
<tr>
<td>JANUARY, 1971</td>
<td>-</td>
<td>4.5</td>
<td>4.0</td>
<td>1880</td>
<td>2.24</td>
</tr>
<tr>
<td>APRIL,</td>
<td>4.4</td>
<td>2.4</td>
<td>0.8</td>
<td>1960</td>
<td>2.34</td>
</tr>
<tr>
<td>AUGUST,</td>
<td>6.5</td>
<td>0.6</td>
<td>3.8</td>
<td>2210</td>
<td>2.64</td>
</tr>
<tr>
<td>DECEMBER,</td>
<td>3.3</td>
<td>-</td>
<td>-</td>
<td>1950</td>
<td>2.33</td>
</tr>
<tr>
<td>FEBRUARY, 1972</td>
<td>-</td>
<td>11.2</td>
<td>10.8</td>
<td>2020</td>
<td>2.41</td>
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<tr>
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<td>9.7</td>
<td>1.9</td>
<td>1.5</td>
<td>1734</td>
<td>2.07</td>
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<td>1.99</td>
</tr>
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<td>JANUARY, 1973</td>
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<td>1132</td>
<td>1.35</td>
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<td>APRIL,</td>
<td>2.1</td>
<td>2.6</td>
<td>1.1</td>
<td>1350</td>
<td>1.61</td>
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<td>1.2</td>
<td>1474</td>
<td>1.76</td>
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<tr>
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<td>3.8</td>
<td>9.2</td>
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<td>1.75</td>
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<td>1.9</td>
<td>1.1</td>
<td>1457</td>
<td>1.74</td>
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<td>4.1</td>
<td>1297</td>
<td>1.55</td>
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<td>4.8</td>
<td>5.8</td>
<td>1759</td>
<td>2.10</td>
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</table>
TABLE-3

NATURAL AND FALLOUT RADIOACTIVITY IN COMPOSITE DIET SAMPLES: SAMPLES COLLECTED IN CITY AREA

Composition of diet sample: Rice-150 g., Pulses-125 g., Wheat-150 g., Vegetables-200 g., Curd-100 gm (includes Sweet dish—many times made from curd), Miscellaneous-25 g., Average Weight of a thali - 750 to 800 gm.

<table>
<thead>
<tr>
<th>Month</th>
<th>$^{137}$Cs (pCi/Kg)</th>
<th>$^{226}$Ra (pCi/Kg)</th>
<th>$^{228}$Th (pCi/Kg)</th>
<th>$^{40}$K (pCi/Kg)</th>
<th>Potassium content (g/Kg)</th>
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</thead>
<tbody>
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<td>1540</td>
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<td>1.71</td>
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<tr>
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<td>-</td>
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</tr>
<tr>
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<td>2.0</td>
<td>-</td>
<td>1603</td>
<td>1.91</td>
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<td>1.4</td>
<td>0.5</td>
<td>1995</td>
<td>2.38</td>
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<td>2.2</td>
<td>1680</td>
<td>2.00</td>
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<tr>
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<td>1.95</td>
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<td>1.5</td>
<td>1171</td>
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<tr>
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<td>1.1</td>
<td>1526</td>
<td>1.82</td>
</tr>
<tr>
<td>OCTOBER,</td>
<td>-</td>
<td>3.8</td>
<td>4.5</td>
<td>1291</td>
<td>1.54</td>
</tr>
</tbody>
</table>
### TABLE 4

**AVERAGE DAILY INTAKE OF NATURAL AND FALLOUT ISOTOPES THROUGH COMPOSITE MEAL FOR PEOPLE OF BOMBAY**

(pCi/day)

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
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</thead>
<tbody>
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<td>ISOTOPE</td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>2.0</td>
<td>8.8</td>
<td>6.2</td>
<td>1.7</td>
<td>5.3</td>
<td>44,000</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>4.3</td>
<td>3.6</td>
<td>7.0</td>
<td>3.6</td>
<td>4.2</td>
<td>22</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>2.8</td>
<td>2.5</td>
<td>6.4</td>
<td>3.7</td>
<td>4.0</td>
<td>-</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>2210</td>
<td>2490</td>
<td>2380</td>
<td>1900</td>
<td>1820</td>
<td>-</td>
</tr>
<tr>
<td>Potassium</td>
<td>2.64</td>
<td>2.96</td>
<td>2.84</td>
<td>2.27</td>
<td>2.17</td>
<td>-</td>
</tr>
</tbody>
</table>

**These are recommended limits of Daily Intake for individual members of the population at large.**

(Average Values for any period of 12 months). \(^{(5)}\)
REPORTING RESULTS OF RADIOACTIVITY MEASUREMENTS AT NEAR ZERO LEVELS OF SAMPLE ACTIVITY AND BACKGROUND

Naomi H. Harley, NYU Institute of Environmental Medicine
Isabel M. Fisenne, HASL

ABSTRACT

The measurement of background samples and baseline samples in environmental studies of alpha emitting radionuclides can give rise to situations where neither a sample nor a background count will appear in reasonable counting times. The present paper describes the calculation technique for estimating the lower limit of detection in such cases.

The reporting of results at near zero levels of radioactivity presents a special problem. When the measurement of background yields a positive value, its variance and standard deviation may be calculated based on Poisson counting statistics. In any subsequent measurement of a sample, the net activity and its standard deviation may be evaluated to determine whether significant radioactivity is present. If this net activity is less than or equal to zero, it has been the convention at HASL to report that the activity is less than or equal to the standard deviation of this net activity.
A special case arises when neither counter background nor sample activity is detectable over a reasonably long count interval. It is of practical interest to be able to quantitate the upper limit of radioactivity which could be present in this sample and yet yield this result. This is especially true at this time, since increasingly more environmental nuclide measurements may fall into this category.

The measurement and identification of low levels (<0.01 dpm/sample) of alpha emitting nuclides by spectrometry represents a specific example. This measurement is performed with a silicon surface barrier detector. Any background counts measured in these detectors usually fall within the energy intervals corresponding to naturally occurring nuclides, primarily radium and its daughters. The background count rate for these detectors usually range from 1 to 5 counts per 1000 minutes in any of the energy intervals containing peaks of the uranium series, plutonium isotopes and most other alpha emitters of interest.

Thorium-232 is a special case, since the energy interval where it appears (3.9 MeV) normally exhibits very low background. Recently, a measurement for thorium-232 yielded zero counts in 5000 minutes and zero background counts over a similar count interval. It is of value to calculate rigorously an upper limit of thorium-232 activity which could be present in such a sample.
Two cases are derived here. One, where the counting interval is fixed and the other where the count time can be calculated based upon a desired fixed value for the upper limit of activity in the sample.

**PREDETERMINED COUNT INTERVAL**

We may apply the bionomial distribution to a group of $N_0$ atoms to calculate the probability that there will be no alpha decays in counting interval $t$.

The binomial distribution for the probability of $x$ number of disintegrations out of a total of $N_0$ atoms is

$$Pr(x) = \frac{N_0!}{(N_0-x)!} \frac{x}{x!} p^x q^{N_0-x}$$  \hspace{1cm} (1)

where

$Pr(x) = \text{probability of } x \text{ disintegrations during time } t,$

$N_0 = \text{initial number of radioactive atoms},$

$p = \text{probability that an atom will decay in time } t,$ and

$q = \text{probability that an atom will not decay in time } t.$

Since the probability, $q$, that an atom will not decay is

$$q = e^{-\lambda t}$$
and

\[ p = (1 - e^{-\lambda t}) \]

then

\[ Pr(x) = \frac{N_0!}{(N_0-x)!} \frac{(1-e^{-\lambda t})^x}{x!} (e^{-\lambda t})^{N_0-x} \]

The probability of no disintegrations in time t is then

\[ Pr(0) = \frac{N_0!}{N_0!} (1-e^{-\lambda t})^0 (e^{-\lambda t})^{N_0} = e^{-\lambda N_0 t} \quad (2) \]

Because the detector efficiency is less than 100%, there is also the possibility that one or more disintegrations occur but that they are not detected. If the detector efficiency is G and one disintegration occurs, the probability of no count being detected is \((1-G)\). If two disintegrations occur, the probability of zero counts is \((1-G)^2\). In general, whenever \(x\) disintegrations occur, the probability that zero counts will be seen is \((1-G)^x\). Then the probability that one or more of \(N_0\) atoms present disintegrates during time t but that none is detected is

\[ Pr(1) \cdot (1-G) + Pr(2) \cdot (1-G)^2 + Pr(3) \cdot (1-G)^3 + \ldots \quad (3) \]

From (1), the probability of one disintegration occurring is

\[ Pr(1) = \frac{N_0!}{(N_0-1)!} \frac{(1-e^{-\lambda t})^1}{1!} (e^{-\lambda t})^{N_0-1} \quad (4a) \]
The probability of two disintegrations occurring is:

\[ \Pr(2) = \frac{N_0!}{(N_0-2)! \cdot 2!} (1-e^{-\lambda t})^2 \left( e^{-\lambda t} \right) N_0 - 2 \]  

etc.

If the assumption can be made that \( \lambda t \) is small and that \( N_0 \) is much greater than one then, \( \Pr(1), \Pr(2) \) etc. simplify to

\[ \Pr(1) \approx N_0 (\lambda t)^1 \left( e^{-\lambda N_0 t} \right) \]  

\[ \Pr(2) \approx \frac{N_0 N_0}{2!} (\lambda t)^2 \left( e^{-\lambda N_0 t} \right) \]  

\[ \Pr(3) \approx \frac{N_0 N_0 N_0}{3!} (\lambda t)^3 \left( e^{-\lambda N_0 t} \right) \]

The probability for observing zero counts is the sum of the probabilities for no disintegration to occur plus those for the occurrence of one or more disintegrations which are not detected.

\[ P = \Pr(0) + \Pr(1) \cdot (1-G) + \Pr(2) \cdot (1-G)^2 + \Pr(3) \cdot (1-G)^3 + \ldots \]

\[ \approx e^{-\lambda N_0 t} + \lambda N_0 t e^{-\lambda N_0 t} (1-G) + \frac{[\lambda N_0 t (1-G)]^2}{2!} e^{-\lambda N_0 t} + \ldots \]

\[ \approx e^{-\lambda N_0 t} \left[ 1 + \frac{\lambda N_0 t (1-G)}{1!} + \frac{[\lambda N_0 t (1-G)]^2}{2!} + \frac{[\lambda N_0 t (1-G)]^3}{3!} + \ldots \right] \]

\[ \approx e^{-\lambda N_0 t} \cdot e^{+\lambda N_0 t (1-G)} \]

\[ \approx e^{-\lambda N_0 t G} \]

It should be noted that \( \lambda N_0 t G \) is the number of counts to be expected during the interval \( t \).
We would like the occurrence of zero counts when activity is actually present to have a small probability. If this probability is arbitrarily set to 0.05 so that zero counts will be detected only five percent of the time when \( N_0 \) atoms are actually present and the counting interval is \( t \) minutes, then (6b) becomes

\[
e^{-\lambda N_0 tG} = 0.05
\]

\[
\lambda N_0 tG = - \ln (0.05)
\]

\[
(\lambda N_0) = A_0 = - \frac{1}{tG} \ln (0.05)
\]

\[
A_0 = \frac{3}{tG}
\]

(7)

where \( \lambda N_0 = A_0 \) is the upper limit of sample activity in dpm at the 95\% confidence level, when \( t \) is in minutes and \( G \) is expressed as fractional efficiency.

PREDETERMINED UPPER LIMIT OF ACTIVITY

It may be of interest to establish the counting time required to determine that a chosen upper limit of activity is present. Solving (7) for the counting time yields

\[
t = \frac{3}{A_0 G}
\]

(8)
As an example, the measured data for the thorium-232 sample will be used. The efficiency of the surface barrier detector is 0.30 and zero counts were recorded in 5000 minutes. From (7)

\[ K_0 = \frac{3}{(5000)(0.3)} = 0.002 \text{ dpm} \]  

(9)

The upper limit of activity is this sample is thus 0.002 dpm at the 95% confidence level.

BACKGROUND NOT ZERO

All of the above expressions apply equally well to background activity. It is possible that either a background count did not occur during the count interval or that one or more occurred but were not detected. In this case the upper limit to background activity is the same as that calculated in (9) except that there is no efficiency for background and the background activity is expressed as counts per minute. Then expression (9) would become,

\[ B_0 G = \frac{3}{5000} = 0.0006 \text{ cpm} \]

Then according to the convention at HASL, any value of net activity equal to zero would be evaluated in comparison with

\[ S_{\text{net}} = \sqrt{\frac{0.0006}{5000} + \frac{0.0006}{5000}} = 0.0005 \text{ cpm} \]
and the activity would be reported as $\leq \frac{0.0005}{0.3}$ or $\leq 0.002$ dpm at the 95% confidence level.*

Acknowledgement—Dr. Bernard S. Pasternack of NYU made several helpful suggestions when reviewing the manuscript.

*This convention would be used for reporting single values. For averaging purposes, the actual value of the net activity, positive or negative, would be used.
ABSTRACT

Analyses of soil collected at 31 locations northeast of the Nevada Test Site (NTS) as far as south central Idaho, southwestern Wyoming and eastern Utah show higher plutonium levels than expected from global fallout alone. The presence of a second source of plutonium was demonstrated by mass spectrometry, and its origin identified as the NTS. Resolution of the plutonium fallout leads to an uneven dispersion for NTS derived debris. This reinforces the conclusion that safety tests and other detonations which resulted in incomplete fission, conducted from 1956 through 1958, created individual deposition patterns dictated by the wind trajectories at shot time.

INTRODUCTION

Over a period of 14 years (1957-1971, the annual fallout rates of Sr–90 in New York City and Salt Lake City were very similar despite the two and one-half fold difference in mean annual precipitation. Because of the drier climate in Salt Lake City, a proportionately lower fallout of Sr–90 compared to New York City would have been expected, since precipitation is a known scavenger of air-borne debris.

The accumulated deposit was measured at 13 sites in north central and southeast Utah in 1971 by analyzing soil samples (1). Although the deposited amount was not uniform because of differences in climate and topography, there was a clear pattern of higher deposition per unit
amount of precipitation than in New York City and most others areas of the United States. We showed that the fallout from 1957-1971 in New York City and Salt Lake City must have originated from the same global source - the stratosphere. It was reasonable to hypothesize that the Salt Lake City area is a preferential region for direct intrusion of stratospheric air into the troposphere. Prior to 1957, the accumulated Sr-90 deposit in Salt Lake City was 10 - 15 mCi per km² higher than in New York City which implied that Salt Lake City must have received fallout from another source, presumably the Nevada Test Site.

Subsequently, the 13 soil samples collected in 1971 were analyzed for Pu-239-240 and Pu-238 and it was surprising to find higher plutonium levels than predicted from the Sr-90 fallout. By comparing the Pu-239-240 to Sr-90 activity ratios of the Utah soils with the average ratio of thirty-two northern hemisphere soils collected in 1970-71 and containing only global fallout, we estimated that up to 60 percent of the total Pu-239-240 activity deposited at some Utah sites represented a source other than the stratospheric reservoir (2). Soil samples taken at the University of Utah from 1959 through 1971 revealed that the excess plutonium must have been delivered prior to 1959. Mass isotopic analyses indicated that the Nevada Test Site was the probable second source and that twice the level expected from
global fallout, deposited in the Salt Lake City area from NTS. We surmised that this plutonium from NTS was predominantly from some of the tests in which plutonium was physically dispersed by high explosives or tests in which little fission occurred.

Since the two reports summarized above were published, two further steps have been taken to determine the extent and distribution of the off-site plutonium contamination from the NTS: (1) during June 1974 soil was sampled at 18 additional sites in Utah, Nevada, Wyoming and Idaho and analyzed for plutonium, and (2) the plutonium fractions representing all sites sampled in 1971 and 1974 were subjected to mass isotope spectrometry as a means of distinguishing the two sources of debris. This report brings together all the data associated with the two sampling efforts.

METHODS

The 1971 and 1974 soil sampling sites are identified in Table 1. Included are values for the mean annual precipitation (3) and altitude. The sites are mapped in Figure 1 where the numbers correspond to those in the first column of Table 1.

The 1971 samples were taken by the core method (4a) to a depth of 30 cm, while both the template (4b) and the core techniques were used to sample to a depth of 15 cm in 1974. Data on depth distributions, acquired following the 1971 sampling, indicated that over
TABLE 1

SOIL SAMPLING SITES FOR PLUTONIUM DEPOSITION

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Location</th>
<th>Site</th>
<th>Mean ann. precip. (cm)</th>
<th>Altitude (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Provo, UT</td>
<td>Utah State Hospital</td>
<td>34</td>
<td>1400</td>
</tr>
<tr>
<td>2</td>
<td>Salt Lake City, UT</td>
<td>Liberty Park</td>
<td>39</td>
<td>1310</td>
</tr>
<tr>
<td>3</td>
<td>Salt Lake City, UT</td>
<td>Univ. of Utah</td>
<td>41</td>
<td>1460</td>
</tr>
<tr>
<td>4</td>
<td>Ogden, UT</td>
<td>Cache NF</td>
<td>53</td>
<td>1590</td>
</tr>
<tr>
<td>5</td>
<td>Brigham, UT</td>
<td>Tabernacle</td>
<td>46</td>
<td>1370</td>
</tr>
<tr>
<td>6</td>
<td>Heber, UT</td>
<td>Tabernacle</td>
<td>39</td>
<td>1710</td>
</tr>
<tr>
<td>7</td>
<td>Marion, UT</td>
<td>Cemetery</td>
<td>51</td>
<td>2010</td>
</tr>
<tr>
<td>8</td>
<td>Wanship, UT</td>
<td>Rockport LakePk.</td>
<td>42</td>
<td>1860</td>
</tr>
<tr>
<td>9</td>
<td>Heber, UT</td>
<td>private meadow</td>
<td>38</td>
<td>1680</td>
</tr>
<tr>
<td>10</td>
<td>Uinta Mtns, UT</td>
<td>Trial Lake’area</td>
<td>76</td>
<td>2900</td>
</tr>
<tr>
<td>11</td>
<td>Moab, UT</td>
<td>BLM area</td>
<td>20</td>
<td>1620</td>
</tr>
<tr>
<td>12</td>
<td>Manti Lasal NF, UT</td>
<td>Geyser Pass</td>
<td>81</td>
<td>2900</td>
</tr>
<tr>
<td>13</td>
<td>Manti Lasal NF, UT</td>
<td>Lasal Guard Station</td>
<td>41</td>
<td>2260</td>
</tr>
</tbody>
</table>

Sites Sampled in June 1971
(map location designated by open circle)

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Location</th>
<th>Site</th>
<th>Mean ann. precip. (cm)</th>
<th>Altitude (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Tybo, NV</td>
<td>grazing land</td>
<td>&lt;20</td>
<td>1620</td>
</tr>
<tr>
<td>2</td>
<td>Cherry Creek, NV</td>
<td>Humboldt NF, Quinn Canyon Range</td>
<td>33</td>
<td>2440</td>
</tr>
<tr>
<td>3</td>
<td>Timber Mtn.Pass, NV</td>
<td>Black Cliff area</td>
<td>&lt;20</td>
<td>1710</td>
</tr>
<tr>
<td>4</td>
<td>Panaca, NV</td>
<td>grazing land</td>
<td>&lt;20</td>
<td>1460</td>
</tr>
<tr>
<td>5</td>
<td>St.George, UT</td>
<td>Dixie NF, Cottonwood Canyon</td>
<td>25</td>
<td>1650</td>
</tr>
<tr>
<td>6</td>
<td>Panguitch, UT</td>
<td>Dixie NF, Panguitch Lake</td>
<td>41</td>
<td>2200</td>
</tr>
<tr>
<td>7</td>
<td>W.Milford, UT</td>
<td>W.of Frisco Pass &amp; E. of Wah Wah Valley</td>
<td>&lt;20</td>
<td>1460</td>
</tr>
<tr>
<td>8</td>
<td>Baker, NV</td>
<td>F. Baker ranch</td>
<td>20</td>
<td>1590</td>
</tr>
<tr>
<td>9</td>
<td>Ely, NV</td>
<td>city pasture</td>
<td>27</td>
<td>1950</td>
</tr>
<tr>
<td>10</td>
<td>Eureka, NV</td>
<td>grazing land</td>
<td>30</td>
<td>1770</td>
</tr>
<tr>
<td>11</td>
<td>Elko, NV</td>
<td>grazing land</td>
<td>23</td>
<td>1650</td>
</tr>
<tr>
<td>12</td>
<td>Wendover, NV</td>
<td>west of salt flat</td>
<td>13</td>
<td>1370</td>
</tr>
<tr>
<td>13</td>
<td>Veron-Eureka, UT</td>
<td>mountain meadow</td>
<td>38</td>
<td>1830</td>
</tr>
<tr>
<td>14</td>
<td>Wales, UT</td>
<td>mountain meadow</td>
<td>41</td>
<td>2380</td>
</tr>
<tr>
<td>15</td>
<td>Vernal, UT</td>
<td>meadow</td>
<td>20</td>
<td>1830</td>
</tr>
<tr>
<td>16</td>
<td>Robertson, WY</td>
<td>meadow</td>
<td>36</td>
<td>2380</td>
</tr>
<tr>
<td>17</td>
<td>Cache NF, UT</td>
<td>Bear River Range, Tony Grove, R.S.</td>
<td>71</td>
<td>2230</td>
</tr>
<tr>
<td>18</td>
<td>Twin Falls, ID</td>
<td>Old County Hospital</td>
<td>23</td>
<td>1070</td>
</tr>
</tbody>
</table>
FIG. 1 SOIL SAMPLING SITES: open circles, 1971 samples
dots, 1974
95% of the plutonium should be in the top 15 cm of soil (9).

The sampling method depends upon the soil type and moisture content. The template was used in those situations where a soil core would not hold together.

The 1974 sampling sites were selected from an area defined by the composite trajectory of the winds which existed at the detonation times of 12 safety experiments at NTS. A total of 26 safety tests were carried out at NTS from 1956 through 1958 (5).

As far as could be ascertained, the sites represented undisturbed soil for a period of 20 years. Contacts were made with Forest Service and Soil Conservation personnel who were familiar with the general areas and suggested sites where undisturbed soil could be found.

The soils were prepared for analysis at HASL by drying, crushing, and blending. A five kilogram portion was pulverized (4c). Kilogram aliquots of all the soils were acid extracted. In addition, one hundred gram aliquots of the 1974 soils were analyzed. Ten and one hundred gram aliquots of some of the samples were completely solubilized to compare with the acid extractions. Plutonium was chemically separated and purified by the HASL procedure (6). After electrodeposition on platinum and alpha counting with a silicon barrier detector, the plutonium fractions were isotopically
analyzed by mass spectrometry.

Radiochemical analyses were carried out at HASL, Teledyne-
Isotopes (Palo Alto) and LFE Environmental Analysis Laboratories.'
Mass spectrometry was performed at Knolls Atomic Power Laboratory,
McClellan Central Laboratory and Lawrence Livermore Laboratory.

Aliquots of all the soils were non-destructively analyzed for
Cs-137 at HASL by lithium-drifted germanium diode spectrometry (4d).

RESULTS

Single values or averages of replicate analyses for Cs-137,
Pu-239,240, and Pu-238 are given in Table 2. Error terms represent
one standard deviation due to counting in the case of a single
analysis and one standard deviation about the mean in the case of
replicate analyses. The activity ratios, Pu-239,240 to Cs-137 and
Pu-238 to Pu-239,240 are listed in the table for future reference
and the mass ratio, Pu-240 to Pu-239 is given in the last column.
Table A in the Appendix provides the individual analytical results
for these samples including mass ratios involving Pu-241 and Pu-242.

Some of the Pu-239,240 values in Table 2 show a rather large
spread about the mean of replicate analyses, and in one case the
error term is close to 50 percent. Our previous experience with
<table>
<thead>
<tr>
<th>No.</th>
<th>Site</th>
<th>Location</th>
<th>Site</th>
<th>Activity Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>S1718</td>
<td>Provo, UT</td>
<td>institution lawn</td>
<td>4.91±0.4</td>
</tr>
<tr>
<td>2</td>
<td>S1712</td>
<td>Cherry Creek, NV</td>
<td>mountain meadow</td>
<td>9.71±1.9</td>
</tr>
<tr>
<td>3</td>
<td>S1911</td>
<td>Timber Mtn., NV</td>
<td>sparses veg.</td>
<td>3.80±0.5</td>
</tr>
<tr>
<td>4</td>
<td>S1909</td>
<td>Panaca, NV</td>
<td>grazing land</td>
<td>4.54±0.0</td>
</tr>
<tr>
<td>5</td>
<td>S1908</td>
<td>St. George, UT</td>
<td>mountain meadow</td>
<td>3.23±0.1</td>
</tr>
<tr>
<td>6</td>
<td>S1907</td>
<td>Panguitch, UT</td>
<td>grassed field</td>
<td>2.40±0.1</td>
</tr>
<tr>
<td>7</td>
<td>S1906</td>
<td>W. Milford, UT</td>
<td>west of Frisco Pass</td>
<td>2.25±0.2</td>
</tr>
<tr>
<td>8</td>
<td>S1901</td>
<td>Baker, NV</td>
<td>Ranch pasture</td>
<td>9.04±0.0</td>
</tr>
<tr>
<td>9</td>
<td>S1900</td>
<td>Ely, NV</td>
<td>City pasturand</td>
<td>4.52±0.5</td>
</tr>
<tr>
<td>10</td>
<td>S1899</td>
<td>Eureka, NV</td>
<td>grazing land</td>
<td>3.59±0.5</td>
</tr>
<tr>
<td>11</td>
<td>S1898</td>
<td>Elko, NV</td>
<td>grazing land</td>
<td>2.45±0.4</td>
</tr>
<tr>
<td>12</td>
<td>S1897</td>
<td>Wendover, NV</td>
<td>west of salt flat</td>
<td>2.18±0.1</td>
</tr>
<tr>
<td>13</td>
<td>S1892</td>
<td>Vernon-Eureka, UT</td>
<td>mountain meadow</td>
<td>2.42±0.4</td>
</tr>
<tr>
<td>14</td>
<td>S1891</td>
<td>Wades, UT</td>
<td>mountain meadow</td>
<td>3.78±1.1</td>
</tr>
<tr>
<td>15</td>
<td>S1890</td>
<td>Vernal, UT</td>
<td>meadow</td>
<td>1.76±0.3</td>
</tr>
<tr>
<td>16</td>
<td>S1888</td>
<td>Robertson, NV</td>
<td>meadow</td>
<td>3.58±0.7</td>
</tr>
<tr>
<td>17</td>
<td>S1887</td>
<td>Cache N.F., UT</td>
<td>Tony Grove meadow</td>
<td>4.34±1.0</td>
</tr>
<tr>
<td>18</td>
<td>S1886</td>
<td>Twin Falls, ID</td>
<td>Co. Hospital lawn</td>
<td>2.44±0.3</td>
</tr>
</tbody>
</table>

**Table 2. Plutonium Fallout Soils**

*June 1974 Sampling (Sampling depth 0-15 cm)*

**Table Notes:**
- No. of analyses:
- Activity Ratio:
- Mass Isotope Ratio:
- Sampling Depth: 0-30 cm

---

*Not included: single HASL value of 16.1.*

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soils from other areas has shown that our analytical reproducibility was less than twenty percent (7). There are five sites where duplicate samples were taken. Table 3 shows the sampling reproducibility (the difference between pairs expressed as a percent of the mean) for the three isotopes and the Pu-240 to Pu-239 mass ratio. For Pu-239-240, the sampling reproducibility ranged from 7 to 57 percent whereas in previous studies a sampling reproducibility of 20 percent or less was usually encountered (7). The corresponding values for Cs-137 and the mass ratio Pu-240 to Pu-239 are less than 15 percent. The most plausible explanation for large analytical and sampling errors associated with the plutonium analyses is that the plutonium containing particles are not uniformly distributed and that in some samples relatively few particles contain the bulk of the plutonium activity. It would appear that we are dealing with the so-called "hot particle problem" which has plagued the EPA in their off-site plutonium in soil program (8). Obviously the much larger samples collected and analyzed in this work were still not adequate to entirely overcome this problem.

In analyzing soil containing plutonium from NTS activities, the possibility exists that some fraction of the plutonium can
### TABLE 3. DUPLICATE SAMPLING

<table>
<thead>
<tr>
<th>Map Site</th>
<th>Sampling Year</th>
<th>Site</th>
<th>Cs-137</th>
<th>Pu-239,240</th>
<th>Pu-238</th>
<th>Pu-240 to Pu-239</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>1971</td>
<td>Salt Lake C., UT</td>
<td>12</td>
<td>12</td>
<td>6</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>1974</td>
<td>Timber Mtn., NV</td>
<td>14</td>
<td>35</td>
<td>28</td>
<td>13</td>
</tr>
<tr>
<td>8</td>
<td>&quot;</td>
<td>Baker, NV</td>
<td>1</td>
<td>57</td>
<td>64</td>
<td>10</td>
</tr>
<tr>
<td>13</td>
<td>&quot;</td>
<td>Vernon-Eureka, UT</td>
<td>7</td>
<td>7</td>
<td>22</td>
<td>10</td>
</tr>
<tr>
<td>16</td>
<td>&quot;</td>
<td>Robertson, WY</td>
<td>7</td>
<td>29</td>
<td>8</td>
<td>6</td>
</tr>
</tbody>
</table>
not be extracted with acid. In this study there were ten pairs of data which could be used to compare acid extraction with complete dissolution (see Table B-1 in the Appendix). Using a two-sided T-test, \( \alpha = 0.05 \), which would indicate that at the 95 percent confidence level, acid extraction was as effective as complete dissolution in recovering plutonium. Individual pairs of data, however, show large variations which are only plausible if the "hot particle" explanation is invoked. Using the same statistical test we compared the acid extraction of 100 and 1000 gram aliquots and the complete dissolution of 10 and 100 gram aliquots (see Tables B-2 and B-3 in the Appendix). No difference between the sets of data in either case, at the 95 percent confidence level, was observed.

In addition to blind replicates, aliquots of pre-bomb (or blank) soil and reference soils were analyzed as indicators of analytical quality. No evidence of contamination during analyses was found. Tables C-1 through C-3 in the Appendix summarize the supporting data for this observation.

DISCUSSION

Although the emphasis in this work is on plutonium deposited northeast of the Nevada Test Site, the soils sampled were analyzed for the fission product Cs-137 to compare distributions. The
Cs-137 results in mCi per km$^2$ are mapped in Figure 2 and show a random but fairly uniform deposition pattern. The highest values reflect high amounts of precipitation and the lowest values are from the drier areas. Although there is no obvious gradation from high to low deposits with increasing distance from the NTS, presumably some variable fraction of the total Cs-137 measured originated from nuclear tests in Nevada. The Pu-239,240 to Cs-137 ratio for soils containing only global fallout is 0.016±0.003 (10). Table 2 shows that this ratio is higher in almost all cases indicating the presence of plutonium from a source other than global fallout.

The total (measured) deposit of Pu-239,240 mapped in Figure 3 does not show a clear pattern of decreasing levels with distance from NTS. From a deposit of 1.7 mCi per km$^2$ at Vernal, UT to 19.5 mCi per km$^2$ at Eureka, NV, these values range from about equal to five times the levels measured elsewhere in the conterminous United States (11).

By using mass spectrometry, we were able to determine the plutonium isotopic composition of these integrated fallout samples. The ratio of Pu-240 to Pu-239 is given for each site in Table 2. The mass isotopic composition of global fallout plutonium in soil has been determined from analysis of 65 soil samples collected throughout the world during 1970-1971 (12). The global mean atom
FIG. 2 CUMULATIVE TOTAL DEPOSIT OF Cs-137
(mCi per km²)
FIG. 3 CUMULATIVE TOTAL DEPOSIT OF Pu-239,240 (mCi per km²)
ratio for Pu-240 to Pu-239 is 0.176±0.014. It has been demonstrated that by mass spectrometric analysis, it is possible to differentiate between global fallout plutonium and plutonium from another source (13,14). It is obvious from the last column in Table 2 that most of the soil samples contain plutonium from a second source. If the mass isotopic composition of plutonium from two sources is sufficiently different, a mixture of these sources can be resolved by applying the following equation (12).

\[
\frac{(Pu \text{ activity})_1}{(Pu \text{ activity})_2} = \frac{(R_2 - R)}{(R - R_1)} \frac{(1 + 3.6R_1)}{(1 + 3.6R_2)}
\]

where \( R \) is the Pu-240 to Pu-239 atom ratio of the mixture and \( R_1 \) and \( R_2 \) the ratios for source 1 and 2, respectively. We used a Pu-240 to Pu-239 value for NTS of 0.05±0.01 (12) and for global fallout, 0.18±0.01 (12). Based on this relationship it is straightforward to calculate the global and the excess plutonium deposit for each site. These values are given in Table 4. The last column of this table shows the percent of the total (measured) deposit of plutonium that is attributed to the NTS. All but two of the sites show the presence of NTS plutonium.

The deposition values for global fallout Pu-239,240 are mapped in Fig. 4 and show a random, rather homogeneous pattern with higher values
### Table 4. Components of Plutonium Fallout in Soil

(Determined from the Mass Ratio: Pu-240 to Pu-239)

#### 1971 Samples

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Location</th>
<th>mCi Pu-239,240 per km²</th>
<th>% of Total Attributed to NTS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Provo, UT</td>
<td>1.2±0.7 4.8±0.8</td>
<td>80±14</td>
</tr>
<tr>
<td>2</td>
<td>Salt Lake C., UT</td>
<td>1.5±0.4 2.6±0.4</td>
<td>63±10</td>
</tr>
<tr>
<td>3</td>
<td>Salt Lake C., UT</td>
<td>1.7±0.5 3.5±0.6</td>
<td>67±12</td>
</tr>
<tr>
<td>4</td>
<td>Ogden, UT</td>
<td>3.3±0.3 2.2±0.3</td>
<td>40±5</td>
</tr>
<tr>
<td>5</td>
<td>Brigham, UT</td>
<td>2.1±0.2 0.8±0.4</td>
<td>27±14</td>
</tr>
<tr>
<td>6</td>
<td>Heber, UT</td>
<td>1.4±0.3 1.5±0.3</td>
<td>52±10</td>
</tr>
<tr>
<td>7</td>
<td>Marion, UT</td>
<td>2.1±0.4 2.5±0.4</td>
<td>52±9</td>
</tr>
<tr>
<td>8</td>
<td>Wanship, UT</td>
<td>2.0±0.2 0.9±0.2</td>
<td>32±6</td>
</tr>
<tr>
<td>9</td>
<td>Heber, UT</td>
<td>2.0±0.2 0.9±0.2</td>
<td>30±6</td>
</tr>
<tr>
<td>10</td>
<td>Uinta Mtns., UT</td>
<td>1.0±0.1 0.9±0.1</td>
<td>32±6</td>
</tr>
<tr>
<td>11</td>
<td>Moab, UT</td>
<td>2.0±0.1 1.1±0.1</td>
<td>34±5</td>
</tr>
<tr>
<td>12</td>
<td>Geyser Pass, UT</td>
<td>2.2±0.1 1.1±0.2</td>
<td>33±5</td>
</tr>
<tr>
<td>13</td>
<td>Lasal, G.S., UT</td>
<td>2.3±0.1 0.0</td>
<td>0</td>
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</tbody>
</table>

#### 1974 Samples

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Location</th>
<th>mCi Pu-239,240 per km²</th>
<th>% of Total Attributed to NTS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Tybo, NV</td>
<td>2.0±0.7 2.9±1.6</td>
<td>60±37</td>
</tr>
<tr>
<td>2</td>
<td>Cherry Creek, NV</td>
<td>1.1±0.5 8.6±2.0</td>
<td>89±27</td>
</tr>
<tr>
<td>3</td>
<td>Timber Mtn., NV</td>
<td>1.0±0.4 2.8±0.6</td>
<td>75±19</td>
</tr>
<tr>
<td></td>
<td>duplicate sampl.</td>
<td>1.9±0.5 3.6±1.3</td>
<td>67±28</td>
</tr>
<tr>
<td>4</td>
<td>Panaca, NV</td>
<td>1.5±0.7 3.0±2.1</td>
<td>67±55</td>
</tr>
<tr>
<td>5</td>
<td>St. George, UT</td>
<td>1.8±0.2 1.4±0.2</td>
<td>44±6</td>
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<tr>
<td>6</td>
<td>Panguitch, UT</td>
<td>1.7±0.1 0.7±0.2</td>
<td>31±7</td>
</tr>
<tr>
<td>7</td>
<td>W. Milford, UT</td>
<td>1.1±0.1 1.1±0.2</td>
<td>49±12</td>
</tr>
<tr>
<td>8</td>
<td>Baker, NV</td>
<td>2.7±0.8 6.3±0.8</td>
<td>70±10</td>
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<td></td>
<td>duplicate sampl.</td>
<td>1.2±0.3 3.8±0.3</td>
<td>76±6</td>
</tr>
<tr>
<td>9</td>
<td>Ely, NV</td>
<td>1.7±0.3 3.7±0.6</td>
<td>68±13</td>
</tr>
<tr>
<td>10</td>
<td>Eureka, NV</td>
<td>2.8±1.0 16.7±1.1</td>
<td>86±6</td>
</tr>
<tr>
<td>11</td>
<td>Elko, NV</td>
<td>1.8±0.3 0.6±0.5</td>
<td>24±22</td>
</tr>
<tr>
<td>12</td>
<td>Wendover, NV</td>
<td>1.9±0.1 0.2±0.2</td>
<td>84±8</td>
</tr>
<tr>
<td>13</td>
<td>Vernon-Eureka, UT</td>
<td>2.3±0.3 1.9±0.5</td>
<td>45±12</td>
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<td></td>
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<td>2.8±1.0 1.7±1.9</td>
<td>37±44</td>
</tr>
<tr>
<td>14</td>
<td>Wals, UT</td>
<td>1.2±0.4 2.5±1.2</td>
<td>69±38</td>
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<tr>
<td>15</td>
<td>Vernal, UT</td>
<td>1.0±0.2 0.7±0.4</td>
<td>43±22</td>
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<td>16</td>
<td>Robertson, NV</td>
<td>1.7±0.5 1.8±0.6</td>
<td>52±20</td>
</tr>
<tr>
<td></td>
<td>duplicate sampl.</td>
<td>2.1±1.0 2.6±2.1</td>
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<tr>
<td>17</td>
<td>Cache N.F., UT</td>
<td>3.3±0.2 1.0±0.2</td>
<td>24±5</td>
</tr>
<tr>
<td>18</td>
<td>Twin Falls, ID</td>
<td>1.1±0.3 1.3±0.4</td>
<td>52±18</td>
</tr>
</tbody>
</table>
FIG. 4 CUMULATIVE GLOBAL FALLOUT DEPOSIT OF Pu-239,240
(mCi per km²)
again reflecting higher amounts of rainfall. The ratios of the Cs-137 values to these calculated global fallout Pu-239,240 values average 0.014±0.005 which is in good agreement with the ratio observed in soils containing only global fallout (10). This means that the measured Cs-137 deposits and the calculated global fallout Pu-239,240 deposits for these sites reflect debris originating from the stratosphere, within the limits of error imposed upon these values.

Perhaps the most convincing evidence we have that the excess or second source of plutonium is from NTS safety tests and other detonations which resulted in incomplete fission, is the radiochemical and mass isotope spectrometric data we have on a series of soil samples collected at the University of Utah from 1959 through 1971. Table 5 presents these data as well as data for three soils from Cedar City, UT from 1963 through 1965. The mass ratio of Pu-240 to Pu-239 at the University site is 0.050 in 1959 and increases to 0.084 in 1971. This indicates that the second source of plutonium was deposited prior to 1959 and the increase in the ratio reflects dilution of this debris with global plutonium fallout. Since all of the safety tests at NTS were conducted before 1959, it seems reasonable to attribute a substantial portion of the excess plutonium to this source. The Cedar City data show that the NTS contribution to the total measured plutonium deposit is much less in this area compared to the Salt Lake City area.
<table>
<thead>
<tr>
<th>Year</th>
<th>Total Pu-239,240 (mCi/km²)</th>
<th>Mass Isotope Ratio Pu-239,240</th>
<th>Global Fallout Pu-239,240 (mCi/km²)</th>
<th>Excess Pu-239,240 from NTS (mCi/km²)</th>
<th>% of Total Fallout Pu from NTS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1971</td>
<td>5.2</td>
<td>0.084</td>
<td>KAPL</td>
<td>1.7</td>
<td>3.6</td>
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<tr>
<td>1965</td>
<td>4.7</td>
<td>0.082</td>
<td>LLL</td>
<td>1.4</td>
<td>3.3</td>
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<tr>
<td>1964</td>
<td>4.9</td>
<td>0.079</td>
<td>LLL</td>
<td>1.4</td>
<td>3.5</td>
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<tr>
<td>1963</td>
<td>4.5</td>
<td>0.060</td>
<td>KAPL</td>
<td>0.5</td>
<td>4.0</td>
</tr>
<tr>
<td>1962</td>
<td>4.2</td>
<td>0.058</td>
<td>KAPL</td>
<td>&lt;0.4</td>
<td>3.8</td>
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<td>1960</td>
<td>3.8</td>
<td>0.051</td>
<td>KAPL</td>
<td>&lt;0.4</td>
<td>3.4</td>
</tr>
<tr>
<td>1959</td>
<td>4.2</td>
<td>0.050</td>
<td>LLL</td>
<td>&lt;0.4</td>
<td>3.8</td>
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</tbody>
</table>

**University of Utah**

**Cedar City, Utah**

<table>
<thead>
<tr>
<th>Year</th>
<th>Total Pu-239,240 (mCi/km²)</th>
<th>Mass Isotope Ratio Pu-239,240</th>
<th>Global Fallout Pu-239,240 (mCi/km²)</th>
<th>Excess Pu-239,240 from NTS (mCi/km²)</th>
<th>% of Total Fallout Pu from NTS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1965</td>
<td>1.4</td>
<td>0.141</td>
<td>LLL</td>
<td>1.1</td>
<td>0.3</td>
</tr>
<tr>
<td>1964</td>
<td>1.5</td>
<td>0.132</td>
<td>KAPL</td>
<td>1.1</td>
<td>0.4</td>
</tr>
<tr>
<td>1963</td>
<td>1.4</td>
<td>0.121</td>
<td>KAPL</td>
<td>0.9</td>
<td>0.5</td>
</tr>
</tbody>
</table>
The final figure (Figure 5) shows the Pu-239,240 deposition pattern from NTS. Considering the errors associated with these data, we attempted to draw isopleths to show the dispersion pattern. The 2 mCi per km² isopleth was the lower limit but it is obvious that a much wider area has received plutonium fallout from NTS at levels less than 2 mCi per km². The soil sampling that has been carried out to date does not encompass a large enough area and is not sufficiently dense to provide a reliable value for the total amount of plutonium released from the NTS. Furthermore, it is clear that the dispersion pattern is more complex than we are able to show. Apparently each individual detonation produced a separate downwind deposition pattern and the composite picture contains small area highs and lows that cannot be defined by the data available here. Within the isopleths that we have drawn, however, we estimate by contouring that 700 curies of Pu-239-240 from NTS have deposited northeast of the test site as far as the Salt Lake City area. By comparison the global fallout Pu-239,240 within the same area amounts to about 250 curies.

Twelve of the twenty-six safety tests had wind trajectories that followed a northeast pattern from NTS. The wind trajectories for the remaining fourteen detonations overlap the other three segments of the compass. Our coverage, therefore, from an inventory
FIG. 5 CUMULATIVE NTS DEPOSIT OF Pu-239,240 (mCi per km²)
standpoint is very limited. We know, for example, that the soil we sampled at Burbank, CA in 1970 includes some plutonium from NTS because the Pu-240 to Pu-239 mass ratio was 0.144 compared with the average of 0.176±0.014 for global fallout (12). The total Pu-239,240 deposit was only 0.7 mCi per km² so the NTS contribution is considerably less at this site than in the Salt Lake City area, by comparison.

CONCLUSIONS

Integrated plutonium fallout levels as measured in soil samples taken northeast of the Nevada Test Site in 1971 and 1974 are in most cases several times higher than expected from global fallout. Mass spectrometric analysis of the radiochemically separated plutonium fractions demonstrates the presence of a second source of plutonium. A sequence of soil samples collected from 1959 to 1971 at the University of Utah indicate that this second source of plutonium fallout was the safety tests and other detonations where little or no fissioning occurred, which were carried out from 1956 through 1958 at the NTS. Resolution of the plutonium fallout levels into global and NTS source components leads to a reasonably uniform global fallout pattern as far north as Twin Falls, ID and as far east as Vernal, UT. Twenty-nine of the thirty-one sites sampled
show the presence of NTS plutonium but the dispersion does not show a smoothly decreasing pattern with distance from NTS. The twelve safety tests which had wind trajectories northeast of the NTS apparently created distinctly different deposition patterns. Within the 2 mCi per km² isopleth which extends northeast as far as the Salt Lake City area, about 700 curies of NTS Pu-239,240 has deposited as compared to about 250 curies of global fallout Pu-239,240. The sampling coverage and density, however, are inadequate to show the apparently broader extent of off-site plutonium contamination from NTS.

ACKNOWLEDGEMENTS

The support and effort of the following groups and individuals were indispensable in carrying out this study:

The sampling areas were selected based on wind trajectories prepared for each event by Kosta Telegadas of the Air Resources Laboratories, National Oceanic and Atmospheric Administration, Silver Spring, MD.

Mr. Kermit Larson of the Forest Service in Ogden, UT and Dr. Theron Hutchings of the Soil Conservation Service in Salt Lake City provided the names of field personnel who designated suitable sampling areas.

The sampling phase was carried out with the field assistance and advice of Frank Jakubowski and Harold Peer of the Environmental Protection Agency, National Environmental Research Center, Las Vegas. Mr. Wayne Bliss, head of the off-site monitoring operations at EPA-LV, provided the field support and guidance.
Mass Spectrometric analyses were performed at Knolls Atomic Power Laboratory under the direction of Leonard Dietz and Frank Rourke (Mass Spectrometry Programs), at Lawrence Livermore Laboratory by Jene Dupzyk and Riley Carver (Radiochemistry Division) and at McClellan Central Laboratory under the direction of Col. William Meyers.

The analyses at HASL were carried out by N.Y. Chu, J. Feldstein and C. Sanderson.
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Mass Isotopic Composition of Global Fallout Plutonium in
Soil
IAEA Int'l Symp. on Transuranium Nuclides in the Environment,
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ST. 1/PUB/410

Plutonium Isotopic Ratios at Rocky Flats
USAEC Report HASL-257, p. I-95, July (1972)

Remote Plutonium Contamination and Total Inventories from
Rocky Flats
<table>
<thead>
<tr>
<th>No.</th>
<th>Site</th>
<th>Date</th>
<th>Core Depth</th>
<th>Area of Cores</th>
<th>Air Drilled</th>
<th>Sampled Total</th>
<th>Core Sampling Method</th>
<th>Ce-137</th>
<th>Pu-239-240</th>
<th>Pu-238</th>
<th>Mcl per kg</th>
<th>Mass Isotope Ratio</th>
<th>Mass Spectrum</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1718</td>
<td>Provo, UT</td>
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<td>620</td>
<td>29.3</td>
<td>HASL 234</td>
<td>Ge-Li Acid Extr. Complete Sol'n</td>
<td>0.6726</td>
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<td>Ge-Li Acid Extr. Complete Sol'n</td>
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<td>Duplicate Sampling</td>
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<td>25.8</td>
<td>HASL 268</td>
<td>Ge-Li Acid Extr. Complete Sol'n</td>
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<td>S1730</td>
<td>Manti-Lasal-Copper F.</td>
<td>6-14-71</td>
<td>0-30</td>
<td>620</td>
<td>21.3</td>
<td>HASL 296</td>
<td>Ge-Li Acid Extr. Complete Sol'n</td>
<td>1.0433</td>
<td>0.02131</td>
<td>0.0007326</td>
<td>160</td>
<td>3.3</td>
<td>0.11</td>
</tr>
<tr>
<td>S1731</td>
<td>Manti-Lasal-Guard S.</td>
<td>6-14-71</td>
<td>0-30</td>
<td>620</td>
<td>23.0</td>
<td>HASL 316</td>
<td>Ge-Li Acid Extr. Complete Sol'n</td>
<td>0.56510</td>
<td>0.01451</td>
<td>0.0005625</td>
<td>89</td>
<td>2.3</td>
<td>0.09</td>
</tr>
</tbody>
</table>

**TABLE A:**

Basic data for soil samples collected for plutonium from NTS.

- **HASL:** High Altitude Soils Laboratory
- **IPA:** Indoor Particle Analyzer
- **LFE:** Low Flow Extraction
- **HASL:** High Altitude Soil Laboratory
- **KAPL:** KAPL Soil Laboratory
- **Spec.:** Special

---

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<table>
<thead>
<tr>
<th>Map No.</th>
<th>Site</th>
<th>Date (cm)</th>
<th>Area (m²)</th>
<th>Air Cuts (kg)</th>
<th>Total Grav. (kg)</th>
<th>NASL (g)</th>
<th>HASL (g)</th>
<th>LFE (mg)</th>
<th>IIASL (mg)</th>
<th>Anal. Method</th>
<th>Sampled Soil %</th>
<th>Mass Isotope Ratio</th>
<th>Mass Spec.</th>
<th>Lab.</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>Milford, UT</td>
<td>6-6-74</td>
<td>0-15</td>
<td>2</td>
<td>800</td>
<td>14.6</td>
<td>HASL 371</td>
<td>Ge-Li</td>
<td>0.8824</td>
<td>72</td>
<td>0.0258</td>
<td>0.0097</td>
<td>2.1</td>
<td>0.0126</td>
</tr>
<tr>
<td>8</td>
<td>Baker, NV</td>
<td>6-6-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>12.8</td>
<td>HASL 300</td>
<td>Ge-Li</td>
<td>0.8827</td>
<td>82</td>
<td>0.0102</td>
<td>0.0028</td>
<td>9.5</td>
<td>0.0997</td>
</tr>
<tr>
<td>9</td>
<td>Elko, NV</td>
<td>6-6-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>11.6</td>
<td>HASL 267</td>
<td>Ge-Li</td>
<td>1.3957</td>
<td>120</td>
<td>0.0706</td>
<td>0.0012</td>
<td>5.0</td>
<td>0.0625</td>
</tr>
<tr>
<td>10</td>
<td>Eureka, NV</td>
<td>6-6-74</td>
<td>0-15</td>
<td>3</td>
<td>675</td>
<td>13.7</td>
<td>HASL 347</td>
<td>Ge-Li</td>
<td>1.3067</td>
<td>120</td>
<td>0.2085</td>
<td>0.0008</td>
<td>19.0</td>
<td>0.2152</td>
</tr>
<tr>
<td>11</td>
<td>Elko, NV</td>
<td>6-8-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>13.6</td>
<td>HASL 289</td>
<td>Ge-Li</td>
<td>1.1253</td>
<td>120</td>
<td>0.0216</td>
<td>0.0013</td>
<td>2.1</td>
<td>0.0275</td>
</tr>
<tr>
<td>12</td>
<td>Wendover, UT</td>
<td>6-8-74</td>
<td>0-15</td>
<td>2</td>
<td>800</td>
<td>15.6</td>
<td>HASL 367</td>
<td>Ge-Li</td>
<td>1.2502</td>
<td>110</td>
<td>0.0556</td>
<td>0.0014</td>
<td>5.0</td>
<td>0.0575</td>
</tr>
<tr>
<td>13</td>
<td>Vernon- Eureka, UT</td>
<td>6-9-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>12.1</td>
<td>HASL 277</td>
<td>Ge-Li</td>
<td>1.6956</td>
<td>150</td>
<td>0.0651</td>
<td>0.0018</td>
<td>3.9</td>
<td>0.0756</td>
</tr>
<tr>
<td>14</td>
<td>Wales, UT</td>
<td>6-9-74</td>
<td>0-15</td>
<td>2</td>
<td>800</td>
<td>8.2</td>
<td>HASL 239</td>
<td>Ge-Li</td>
<td>2.7326</td>
<td>190</td>
<td>0.0921</td>
<td>0.0029</td>
<td>4.3</td>
<td>0.0974</td>
</tr>
<tr>
<td>15</td>
<td>Vernal, UT</td>
<td>6-10-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>14.9</td>
<td>HASL 364</td>
<td>Ge-Li</td>
<td>0.7120</td>
<td>77</td>
<td>0.0141</td>
<td>0.0004</td>
<td>1.5</td>
<td>0.0182</td>
</tr>
<tr>
<td>16</td>
<td>Robertson, NV</td>
<td>6-11-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>12.0</td>
<td>HASL 314</td>
<td>Ge-Li</td>
<td>1.6123</td>
<td>140</td>
<td>0.0711</td>
<td>0.0012</td>
<td>2.7</td>
<td>0.0426</td>
</tr>
<tr>
<td>17</td>
<td>Cavez NF, UT</td>
<td>6-11-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>11.0</td>
<td>HASL 271</td>
<td>Ge-Li</td>
<td>1.7023</td>
<td>150</td>
<td>0.0712</td>
<td>0.0014</td>
<td>2.8</td>
<td>0.0735</td>
</tr>
<tr>
<td>18</td>
<td>Twin Falls, ID</td>
<td>6-12-74</td>
<td>0-15</td>
<td>10</td>
<td>620</td>
<td>10.4</td>
<td>HASL 262</td>
<td>Ge-Li</td>
<td>1.1823</td>
<td>88</td>
<td>0.0345</td>
<td>0.0004</td>
<td>2.3</td>
<td>0.0275</td>
</tr>
</tbody>
</table>

TABLE A
BASIC DATA FOR SOIL SAMPLES COLLECTED FOR PLUTONIUM FROM NTS (Cont'd)
<table>
<thead>
<tr>
<th>HASL No.</th>
<th>Site</th>
<th>mCi Pu-239,240 per km²</th>
<th>No. of Analyses</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1713</td>
<td>Univ. of UT</td>
<td>5.3±0.2 5.8±0.1</td>
<td>1 1</td>
</tr>
<tr>
<td>S1913</td>
<td>Tybo, NV</td>
<td>4.2±0.8 6.3±0.2</td>
<td>2 1</td>
</tr>
<tr>
<td>S1912</td>
<td>Cherry Creek, NV</td>
<td>11.1±0.4 8.2±1.6</td>
<td>2 2</td>
</tr>
<tr>
<td>S1910</td>
<td>Timber Mtn., NV</td>
<td>5.1±1.1 6.5±0.2</td>
<td>3 1</td>
</tr>
<tr>
<td>S1909</td>
<td>Panaca, NV</td>
<td>4.6±2.5 4.3±0.1</td>
<td>3 1</td>
</tr>
<tr>
<td>S1901</td>
<td>Baker, NV</td>
<td>9.0±0.7 9.0±0.1</td>
<td>2 2</td>
</tr>
<tr>
<td>S1899</td>
<td>Eureka, NV</td>
<td>19.3±0.3 20.0±0.6</td>
<td>2 1</td>
</tr>
<tr>
<td>S1892</td>
<td>Vernon-Eureka, UT</td>
<td>4.0±0.1 4.6±0.2</td>
<td>2 1</td>
</tr>
<tr>
<td>S1891</td>
<td>Wales, UT</td>
<td>4.3±0.1 2.4±0.4</td>
<td>2 1</td>
</tr>
<tr>
<td>S1888</td>
<td>Robertson, WY</td>
<td>3.2±0.7 4.0±0.2</td>
<td>2 1</td>
</tr>
</tbody>
</table>
**TABLE B-2. COMPARISON BETWEEN ACID EXTRACTION OF 100 and 1000 g ALIQUOTS IN SOIL Pu ANALYSIS**

<table>
<thead>
<tr>
<th>HASL No.</th>
<th>Site</th>
<th>mCi Pu-239,240 per km²</th>
<th>No. of Analyses</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>100 g</td>
<td>1000 g</td>
</tr>
<tr>
<td>S1913</td>
<td>Tybo, NV</td>
<td>3.6±0.3</td>
<td>4.7±0.1</td>
</tr>
<tr>
<td>S1912</td>
<td>Cherry Creek, NV</td>
<td>11.4±0.8</td>
<td>10.8±0.3</td>
</tr>
<tr>
<td>S1911</td>
<td>Timber Mtn., NV</td>
<td>3.5±0.2*</td>
<td>4.2±0.1</td>
</tr>
<tr>
<td>S1910</td>
<td>&quot;</td>
<td>4.8±1.5</td>
<td>5.6±0.2</td>
</tr>
<tr>
<td>S1909</td>
<td>Panaca, NV</td>
<td>3.1±0.1</td>
<td>7.5±0.2</td>
</tr>
<tr>
<td>S1908</td>
<td>St. George, UT</td>
<td>3.1±0.2</td>
<td>3.3±0.2</td>
</tr>
<tr>
<td>S1907</td>
<td>Panguitch, UT</td>
<td>2.5±0.2</td>
<td>2.4±0.1</td>
</tr>
<tr>
<td>S1906</td>
<td>W. Milford, UT</td>
<td>2.1±0.2</td>
<td>2.4±0.1</td>
</tr>
<tr>
<td>S1901</td>
<td>Baker, NV</td>
<td>9.5±0.6</td>
<td>8.5±0.4</td>
</tr>
<tr>
<td>S1902</td>
<td>&quot;</td>
<td>5.0±0.5</td>
<td>4.9±0.2</td>
</tr>
<tr>
<td>S1900</td>
<td>Ely, NV</td>
<td>5.8±0.5</td>
<td>5.1±0.2</td>
</tr>
<tr>
<td>S1899</td>
<td>Eureka, NV</td>
<td>19.0±1.7</td>
<td>19.6±0.4</td>
</tr>
<tr>
<td>S1898</td>
<td>Elko, NV</td>
<td>2.1±0.2</td>
<td>2.7±0.1</td>
</tr>
<tr>
<td>S1897</td>
<td>Wendover, NV</td>
<td>2.2±0.2</td>
<td>2.0±0.1</td>
</tr>
<tr>
<td>S1892</td>
<td>Vernon-Eureka, UT</td>
<td>3.9±0.4</td>
<td>4.1±0.1</td>
</tr>
<tr>
<td>S1893</td>
<td>&quot;</td>
<td>6.4±0.6</td>
<td>3.6±0.1</td>
</tr>
<tr>
<td>S1891</td>
<td>Wales, UT</td>
<td>4.3±0.4</td>
<td>4.3±0.2</td>
</tr>
<tr>
<td>S1890</td>
<td>Vernal, UT</td>
<td>1.5±0.2</td>
<td>1.9±0.1</td>
</tr>
<tr>
<td>S1888</td>
<td>Robertson, WY</td>
<td>2.7±0.4</td>
<td>3.7±0.1</td>
</tr>
<tr>
<td>S1889</td>
<td>&quot;</td>
<td>2.8±0.3</td>
<td>4.6±0.1</td>
</tr>
<tr>
<td>S1887</td>
<td>Cache N.F., UT</td>
<td>4.2±0.4</td>
<td>4.4±0.2</td>
</tr>
<tr>
<td>S1886</td>
<td>Twin Falls, ID</td>
<td>2.3±0.3</td>
<td>2.7±0.1</td>
</tr>
</tbody>
</table>

*Excluded 16.1 mCi/km² value.
TABLE B-3. COMPARISON BETWEEN COMPLETE DISSOLUTION OF 10 AND 100 g ALIQUOTS IN SOIL Pu ANALYSIS

<table>
<thead>
<tr>
<th>HASL No.</th>
<th>Site</th>
<th>mCi Pu-239,240 per km²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>10 g</td>
</tr>
<tr>
<td>S1912</td>
<td>Cherry Creek, NV</td>
<td>9.4±0.8</td>
</tr>
<tr>
<td>S1901</td>
<td>Baker, NV</td>
<td>9.1±0.6</td>
</tr>
<tr>
<td>S1903</td>
<td>&quot;</td>
<td>7.4±0.5</td>
</tr>
</tbody>
</table>

(Each value represents a single analysis.)
### TABLE C-1. ANALYTICAL QUALITY CONTROL ASSOCIATED WITH ANALYSES OF SOILS

<table>
<thead>
<tr>
<th>Aliq. wt. (g)</th>
<th>Lab.</th>
<th>Method</th>
<th>dpm Pu-239,240 per g</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>HASL</td>
<td>Acid extr.</td>
<td>0.0002±100%</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td>&quot;</td>
<td>0.0001±100%</td>
</tr>
<tr>
<td>1000</td>
<td>LFE</td>
<td>&quot;</td>
<td>0.0004±14%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Avg. of previous data</td>
<td>0.0002±100%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Aliq. wt. (g)</th>
<th>Lab.</th>
<th>Method</th>
<th>dpm Cs-137 per g</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>HASL</td>
<td>Ge-Li</td>
<td>0.02±100%</td>
</tr>
<tr>
<td>315</td>
<td></td>
<td>&quot;</td>
<td>0.01±100%</td>
</tr>
<tr>
<td>310</td>
<td></td>
<td>&quot;</td>
<td>0.02±100%</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Avg. of previous data</td>
<td>0.04±100%</td>
</tr>
<tr>
<td>Aliq. wt. (g)</td>
<td>Lab.</td>
<td>Method</td>
<td>dpm Pu-239,240 per g</td>
</tr>
<tr>
<td>--------------</td>
<td>------</td>
<td>--------------</td>
<td>---------------------</td>
</tr>
<tr>
<td>100</td>
<td>HASL</td>
<td>Acid extr.</td>
<td>0.011±10%</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td>&quot;</td>
<td>0.012±9%</td>
</tr>
<tr>
<td>1000</td>
<td>LFE</td>
<td>&quot;</td>
<td>0.011±4%</td>
</tr>
<tr>
<td>Avg. of previous data</td>
<td></td>
<td></td>
<td>0.012±10%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Aliq. wt. (g)</th>
<th>Lab.</th>
<th>Method</th>
<th>dpm Cs-137 per g</th>
<th>mCi Cs-137 per km²</th>
</tr>
</thead>
<tbody>
<tr>
<td>364</td>
<td>HASL</td>
<td>Ge-Li</td>
<td>0.72±3%</td>
<td>140</td>
</tr>
<tr>
<td>373</td>
<td></td>
<td>&quot;</td>
<td>0.65±7%</td>
<td>130</td>
</tr>
<tr>
<td>Avg. of previous data</td>
<td></td>
<td></td>
<td>0.78±8%</td>
<td>150</td>
</tr>
</tbody>
</table>
## Table C-3. Analytical Quality Control Associated with Analyses of Soils

N. Eastham, MA. Reference Soil, No. S1781*
Sampled Oct. 1972, 0-30 cm Composite

<table>
<thead>
<tr>
<th>Aliq. wt. (g)</th>
<th>Lab.</th>
<th>Method</th>
<th>dpm Pu-239,240 per g</th>
<th>mCi Pu-239,240 per km²</th>
<th>Mass Ratio Pu-240/Pu-239</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>HASL</td>
<td>Acid extr.</td>
<td>0.012±10%</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td></td>
<td></td>
<td>0.013±10%</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td>1000</td>
<td>LFE</td>
<td></td>
<td>0.014±3%</td>
<td>2.4</td>
<td>0.176</td>
</tr>
<tr>
<td>Avg. of previous data</td>
<td></td>
<td></td>
<td>0.013±10%</td>
<td>2.3</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Aliq. wt. (g)</th>
<th>Lab.</th>
<th>Method</th>
<th>dpm Cs-137 per g</th>
<th>mCi Cs-137 per km²</th>
</tr>
</thead>
<tbody>
<tr>
<td>384</td>
<td>HASL</td>
<td>Ge-Li</td>
<td>0.72±5%</td>
<td>130</td>
</tr>
<tr>
<td>395</td>
<td></td>
<td></td>
<td>0.75±3%</td>
<td>130</td>
</tr>
</tbody>
</table>

*Aliquots taken from original 3 kg that were pulverized in 1972.
ABSTRACT

Radon concentrations at the HASL regional site in Lloyd, New York are reported for an 8 week period during March and April 1976. The high values are associated with temperature inversions, as might be expected.

The Health and Safety Laboratory established a field station at Lloyd, New York in the latter part of 1974. This site is about 90 miles north of HASL and is meant to represent a regional condition for air pollution and for exposure to man-made and natural radioactivities. Preliminary data from this site were reported in the October 1975 Environmental Quarterly (HASL-297). The present report lists some of the radon data collected in the spring of 1976.

A continuous radon monitor was installed as part of the program of measuring radioactivity and radiation at the site. The device was designed, built and has been operated by the Instrumentation Division of HASL. A number of mechanical problems arose during the winter and only a limited amount of continuous data is available. A new instrument is under construction and it is hoped that it will
provide true continuous recording.

The monitor is basically a two-filter system which samples air at 0.1 m$^3$/min. The air is drawn through an input filter to remove radon daughter products and passes through a 170 liter chamber to allow buildup of fresh daughter products which are collected on a sampling filter. During the spring, the samples were collected for half an hour and the daughter products counted for two successive 15 minute periods. The daughter product counts are converted to radon concentrations by a computer program based on the decay of the radionuclides involved. The counter background is less than 5 pCi/m$^3$ and the sensitivity is about 10 pCi/m$^3$. The data are tabulated to the nearest pCi/m$^3$, but this exaggerates their accuracy.

Meteorological data have been collected at the New York ERDA ninety-meter meteorological tower on the site. Operation of the instruments has not been completely satisfactory and it is not possible to attempt a complete correlation of radon levels with meteorology at this time.

RESULTS AND DISCUSSION

The data for 8 weeks during March and April 1976 are recorded in the table. The individual measurements have been combined into 4 hour periods to compress the data. This combination does not
obscure any major changes although one-hour periods may prove best for correlation purposes.

The radon emanating from the ground will raise the air concentration markedly if the radon is mixed into a small volume, that is, if it is not dispersed by the winds or mixed upward in the atmosphere. It is obvious from the data that the higher radon concentrations are associated with the stable conditions of inversions and the lower concentrations are usually associated with the turbulence of superadiabatic temperature gradients. There is no obvious correlation with wind speed and direction.

The overall mean concentration during this period was about 80 pCi/m³ as compared with almost 200 pCi/m³ the previous summer. It is apparent that it will be necessary to have a year of continuous data along with the associated meteorological measurements before tests of correlation can be attempted. It might also be helpful to have corresponding measurements of radon daughters and possibly measurements of unattached fraction and radon emanation as well.
### Radon Measurements at Lloyd, New York
March 6 through April 30, 1976

<table>
<thead>
<tr>
<th>Hour</th>
<th>III 6</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10</th>
<th>11</th>
<th>12</th>
<th>III 13</th>
<th>14</th>
<th>15</th>
<th>16</th>
<th>17</th>
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#### Mean
- 68
- 120
- 72
- 74
- 48
- 54
- 44
- 76
- 41
- 65
- 42
- 30
- 47
- 130

#### Wind
- NW
- SW-NW
- SW-NE
- NE-SE
- SW-NW
- NE-SE
- NW
- NW-SW
- SW-NW
- NE
- NW
- SW
- SW

#### Wind Speed (Beaufort)
- 2
- 2
- 2
- 1
- 2
- 2
- 2
- 1-3
- 2
- 2
- 2
- 2
- 3
- 2
- 2

**Noted:**
- * indicates that fewer than 10 measurements were recorded in the period.
- f indicates weather data not available.
- - indicates radon data not available.
- I indicates temperature inversion shown for 60 meters vs. 10 meters.
- S indicates super-adiabatic conditions with temperature gradient greater than one degree per 100 meters.
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ABSTRACT

Estimates of Sr-90 intake via the total diet in New York City and San Francisco have been made since 1960 from quarterly food samplings and average consumption statistics. The dietary intakes of Sr-90 have decreased from the maximum levels attained during 1963-64, but the declines have become more gradual in recent years due to the continuing small amounts of Sr-90 deposition and the little-changing cumulative deposit of Sr-90 in soil. The annual intake in 1975 averaged 8.3 pCi/day in New York and 3.3 pCi/day in San Francisco, a slight decrease from the previous year in New York and a slight increase in San Francisco. Further gradual reductions in Sr-90 intake are anticipated during 1976.

Quarterly samplings of representative food items in New York City and San Francisco have been conducted by the Health and Safety Laboratory since 1960. Results of Sr-90 analyses and estimates of average consumption of these food items yield estimates of the total dietary intakes of Sr-90 for residents of these cities. This information has been used to study the transfer mechanisms of Sr-90 fallout through the food chain to man and to relate to observed variations in Sr-90 body burdens.

The quarterly results from this program are presented in section II of this report as the data become available. Figure 1 illustrates the quarterly Sr-90 intake results for the entire sampling period.
The average Sr-90 concentration in each diet item and estimates of Sr-90 and Ca intakes for the entire year 1975 are listed in Table 1. It is noted that the calcium intake is estimated to be 370g per year. Thus, Sr-90 intakes expressed as pCi/day or pCi/g Ca are almost equivalent.

Results for 1975

In New York City the annual Sr-90 intake during 1975 was estimated to be 3010 pCi, an average of 8.3 pCi/day. This is a decrease of 13% from the Sr-90 intake in the previous year. This change reflects decay of the accumulated deposit along with a decrease in Sr-90 deposition. In New York the annual Sr-90 deposition was 0.69 mCi/km² in 1975 and 0.93 mCi/km² in 1974. The contributions to Sr-90 intake decreased for most food items, most notably for fresh vegetables and fresh fruits.
Table 1

STRONTIUM-90 IN THE DIET DURING 1975

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<th>Diet Category</th>
<th>New York City</th>
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<td></td>
<td>kg/yr</td>
<td>gCa</td>
<td>pCi Sr-90</td>
<td>pCi Sr-90</td>
<td>kg/yr</td>
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<td>10.5</td>
<td>231</td>
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<td>56</td>
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<tr>
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<td>1.2</td>
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<td>0.9</td>
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<td>3.0</td>
<td>84</td>
<td>2.0</td>
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<td>3010 pCi</td>
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<td>8.1 pCi/gCa</td>
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<td>3.3 pCi/d</td>
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These items generally show the largest variations, due to the diversity of source regions and the uncertainty of origin of produce available for purchase at the time of sampling. Variations which appear as slight increases were noted for potatoes, canned vegetables, flour, meat, and poultry. In terms of food categories, Sr-90 intake in 1975 via dairy products, vegetables and grain products remained unchanged (within 10%) from the previous year, but fruits declined 54% and meats increased 24%. After two years in which vegetables contributed a slightly greater fraction to the total Sr-90 intake in New York than dairy products, the situation reversed in 1975. Milk and other dairy products contributed 37% of the total Sr-90 intake in New York during 1975, while vegetables accounted for 34%. The contribution from fruits decreased to 12%. Grain products contributed 14%, and the combined category of meat, fish, and eggs 3%.

In San Francisco the Sr-90 intake during 1975 was estimated to be 1190 pCi, an average of 3.3 pCi/day, compared to 2.9 pCi/day in 1974. The contributions from most foods remained relatively unchanged, however a slight increase was noted for milk. The relative contributions to the total Sr-90 intake in San Francisco during 1975 were dairy products 31%, vegetables 25%, grain products 22%, fruit 17%, and meat, fish, and eggs 5%.
Table 2 shows how the Sr-90 intake varied during each quarter in 1975 in New York and San Francisco. The numbers in parentheses are the results for 1974.

Table 2

<table>
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<th>Quarter</th>
<th>New York</th>
<th>San Francisco</th>
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<tr>
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<td>8.0 (8.3)</td>
<td>3.7 (2.7)</td>
</tr>
<tr>
<td>2</td>
<td>8.9 (10.5)</td>
<td>3.6 (3.0)</td>
</tr>
<tr>
<td>3</td>
<td>8.2 (10.3)</td>
<td>2.6 (2.6)</td>
</tr>
<tr>
<td>4</td>
<td>7.9 (9.0)</td>
<td>3.1 (3.3)</td>
</tr>
</tbody>
</table>

Yearly Avg. 8.3 (9.5) 3.3 (2.9)

Sr-90 Intake, 1960 - 1975

The estimates of annual intakes since 1960 of Sr-90 in the total diet and the major food categories in New York and San Francisco are illustrated in Figure 2. The total diet Sr-90 levels in New York have been consistently higher than in San Francisco because of less fallout deposition in the San Francisco food-producing region. The declines in Sr-90 intake in 1960-61 followed the nuclear testing moratorium in 1959-60. Maximum levels of Sr-90 in the diet were recorded in 1963-64 following the period of most active nuclear testing from September 1961 until late 1962, following which the Test Ban Treaty was signed.
The rapid declines in Sr-90 intake following the peak values in 1963-64 became more gradual after 1966-67 as uptake from the little changing cumulative deposit of Sr-90 in soil became the dominant factor contributing to Sr-90 levels in food. The resumption of atmospheric testing by the French and Chinese in 1966, resulting in a relatively constant low fallout rate of Sr-90, has also been a factor in maintaining the dietary intakes of Sr-90 at little changing levels since 1968.
The relative importance of the major food categories to the total Sr-90 intake can be seen in Figure 2. Milk, including other dairy products, has been the most important contributor to Sr-90 intake. Grain products are the second greatest contributors during periods of heavy deposition, but vegetables and, to a lesser extent, fruit become more important in later years. Meat, fish, and eggs are minor contributors.

The variations in the relative contributions of the major food categories to the total Sr-90 intake are listed in Table 3. The contribution from dairy products to the total intake in New York has declined to 30 to 40%, it previously being above 40% during periods of heavier deposition. Grain products contribute from 30% in periods of heavy deposition to 15% in later years. For vegetables the range is similar to grain products, but the greatest relative contribution has been occurring in more recent years, due in part to more rapid declines in the other food categories. In San Francisco, the milk contribution has varied from 20 to 30% of the total yearly Sr-90 intake in recent years, increased contribution having occurred during periods of heavier deposition. Grain products have usually accounted for 20 to 30% with a maximum contribution of 39% in 1964, one year after the peak milk contribution.
Vegetables have increased their contribution from 10% in 1964 to 25 to 35% the last few years. The contributions from fruit in both New York and San Francisco have behaved similarly to vegetables but with somewhat less total intake. Meat, fish and eggs have accounted for 5% or less of the total intake in New York and slightly higher relative amounts in San Francisco.
Prediction Methods

A useful model for correlating Sr-90 deposition with Sr-90 levels in milk and a procedure for using milk concentrations to predict total diet Sr-90 intake were discussed in the report HASL-246. The deposition-milk model is given by the formula:

\[ M_n = p_1 F_n + p_2 F_{n-1} + p_3 \sum_{m=1}^{n} F_{n-m} e^{-m\mu} \]

where \( M_n \) (pCi/g Ca) is the average Sr-90/Ca ratio in milk in the year \( n \), and \( F_n \) (mCi/km²) is the Sr-90 fallout deposition during the year \( n \). The proportionality constant \( p_1 \) includes the rate and deposit effects of the current year's deposition; \( p_2 \) is the lag factor for the previous year's deposition; and \( p_3 e^{-m\mu} \) is the deposit factor with exponential removal due to reduced uptake availability of the deposition in each of the preceding years. The results of least squares fits to the average yearly Sr-90 concentrations in milk in New York through 1970, 1971, 1972, 1973, and 1974, were reported previously. No parameter changes were required by the addition of the 1971, 1972, 1973 and 1974 data. Inclusion of the 1975 fallout and milk data (Figure 3) results in the same proportionality factors, which are \( p_1 = .69, p_2 = .21, p_3 = .20, \mu = .14 \).
Figure 3. Average annual Sr-90 concentration in milk in New York City

Such stability of the parameter values with fits to additional data reflects the appropriateness of the model formulation. The mean residence time of Sr-90 in soil inferred from the exponential removal term is 7.1 years after the initial deposition year. This corresponds to 11% per year removal of Sr-90 in addition to the 2.4% radioactive decay. A similar deposition-milk model has been used previously by Bartlett, Russell and Jenkins. Their results for the U. K. are in general agreement with the results reported here.
Table 4 shows the measured annual Sr-90 deposition and the Sr-90/Ca ratio in milk in New York and the milk Sr-90/Ca ratio calculated by the above model using the given proportionality factors. Also shown is the percentage of the Sr-90/Ca ratio attributable to uptake of Sr-90 from soil. Since 1965, contamination of milk has been primarily due to uptake of Sr-90 from the cumulative deposit in soil. In 1975 this source accounted for 86% of the Sr-90 contamination of milk.
The procedure for estimating total diet Sr-90 intake from milk levels is given by the following formula:\(^2\)

\[ D_i = M_i + f M_{i-1} \]

Where \( D_i \) is the quarterly Sr-90 intake in total diet (pCi), \( M_i \) is the measured Sr-90 intake in milk during the quarter \( i \) and \( f M_{i-1} \) is the estimated intake of Sr-90 in foods other than milk based on the milk levels during the preceding quarter. During periods of low fallout deposition the proportionality factor, \( f \), is 1.4. The formula gives 7.7 pCi per day in total diet in New York during 1975, somewhat lower than the measured ratio.

The application of this formula to estimate total diet Sr-90 intake from milk data and comparison with the measured total diet intake are shown in Figure 4. Using a constant proportionality factor, \( f = 1.4 \), for the entire period 1960-75 yields annual estimates of total diet intake within 25% of measured values. To maintain the correlations within 10%, the proportionality factor must be reduced somewhat during the quarters when Sr-90 deposition is greater than 3 mCi/km\(^2\) per quarter (see ref. 2). This correction is indicated by the dashed line during 1962-64 in Figure 4.

The more rapid decline of Sr-90 contamination of milk compared to other foods in recent years necessitates a further correction in
the proportionality factor. Since 1971 a value of $f = 1.8$ is appropriate. This modification is indicated by the dotted line in Fig. 4.

Milk may be used as an empirical indicator of Sr-90 intake in total diet, but it is inappropriate to assume a constant relationship through the entire fallout period.

More detailed investigation of the transfer of Sr-90 deposition to diet is possible by applying the deposition - milk model presented above to other food categories and to the Sr-Ca ratios in total diet.
Since there is a wide area of supply of the various food items, except for milk, the Sr-90 deposition has been taken to be the average for the 30° - 50° N. latitude band. The results of the correlations with New York diet data are listed in Table 5 and illustrated in Figure 5.

As can be seen in Figure 5, the Sr-Ca ratios in fruits and vegetables have been higher than in milk. Fruits and vegetables peaked at just over 50 pCi/gCa in 1964-65, while milk reached 25 pCi/gCa in 1963. Since milk provides 7 and 15 times more calcium to total diet intake than vegetables and fruits, respectively, the contribution to total Sr-90 intake from milk has, until recently, been much more important. In 1973 and 1974 in New York the Sr-Ca ratio in vegetables exceeded that in milk by a factor greater than the Ca intake difference, and vegetables became the larger contributor to Sr-90 intake.

The Sr-Ca ratios in milk and grain products declined relatively rapidly since 1964, corresponding to sharp decline in the fallout deposition rate. The Sr-Ca ratios in fruits and vegetables have declined less markedly, reflecting a stronger dependence on root uptake from the relatively constant cumulative deposit of Sr-90 in soil. The parameters from the model fits (Table 5) illustrate these dependencies. The rate dependence \( p_1 + p_2 \) is highest for grain products, which is
Table 5
PARAMETERS OBTAINED BY FITTING *0Sr/Ca RATIOS IN FOOD CATEGORIES AND TOTAL DIET IN N.Y. TO *0Sr DEPOSITION

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Dairy Products</th>
<th>Grain Products</th>
<th>Meat, Fish Products</th>
<th>Vegetables</th>
<th>Fruit</th>
<th>Total Diet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_1$ ($pCi/gCa$ per $mCi/km^2$y)</td>
<td>0.69</td>
<td>0.86</td>
<td>0.004</td>
<td>0.96</td>
<td>1.36</td>
<td>0.90</td>
</tr>
<tr>
<td>$p_5$ (&quot; )</td>
<td>0.21</td>
<td>1.90</td>
<td>0.17</td>
<td>0.37</td>
<td>0.00</td>
<td>0.54</td>
</tr>
<tr>
<td>$p_5$ (&quot; )</td>
<td>0.20</td>
<td>0.30</td>
<td>0.31</td>
<td>1.03</td>
<td>0.90</td>
<td>0.36</td>
</tr>
<tr>
<td>$\mu$ ($y^{-1}$)</td>
<td>0.14</td>
<td>0.13</td>
<td>0.28</td>
<td>0.07</td>
<td>0.03</td>
<td>0.10</td>
</tr>
</tbody>
</table>

$P_3^j$ Food category transfer coefficient ($pCi$ $y/gCa$ per $mCi/km^2$)

<table>
<thead>
<tr>
<th>Food category</th>
<th>$P_3^j$</th>
<th>$P_3^j$ Food category transfer coefficient ($pCi$ $y/gCa$ per $mCi/km^2$)</th>
<th>$W_j$ Fractional contribution to Ca in total diet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dairy Products</td>
<td>2.23</td>
<td>4.92</td>
<td>1.13</td>
</tr>
<tr>
<td>Grain Products</td>
<td>0.582</td>
<td>0.179</td>
<td>0.111</td>
</tr>
<tr>
<td>Meat, Fish Products</td>
<td>1.30</td>
<td>0.88</td>
<td>0.13</td>
</tr>
</tbody>
</table>

$P_3$ Total diet transfer coefficient ($pCi$ $y/gCa$ per $mCi/km^2$)

<table>
<thead>
<tr>
<th>$P_3$</th>
<th>4.88</th>
</tr>
</thead>
</table>

Contribution to total diet $P_3$ ($W_j P_3^j$) | 4.87 |

Figure 5. Sr-90 in food groups in New York - observed (points) and fits from deposition model (solid lines).
three times higher than for milk. The deposit dependence ($p_3$) is 4 to 5 times greater for vegetables and fruits than for milk, and the mean time for uptake availability ($\mu^{-1}$) is inferred to be 7, 14, and 33 years for milk, vegetables, and fruit, respectively.

For individuals or populations who maintain more nearly vegetarian diets, the Sr-Ca levels in diet, and thus the Sr-90 concentrations in bone would be expected to have declined less markedly than for individuals consuming diets consisting of greater quantities of meat and dairy products.

The total transfer to diet per unit deposition is given by the transfer coefficient $P_{23}$. The subscripts refer to the transfer chain

Atmosphere(1) → Earth's surface(2) → Diet(3) → Tissue(4).

The transfer coefficient is defined as the ratio of the infinite integral of concentrations in diet to the integrated deposition rate. As applied to the correlation model, assuming unit deposition in the year $n=1$, the transfer coefficient is

$$P_{23} = \frac{\sum_{n=1}^{\infty} D_n}{\sum_{n=1}^{\infty} \delta_{n,t} F_n} = p_1 + p_2 + p_3 \sum_{m=1}^{\infty} e^{-m\mu} = p_1 + p_2 + p_3 \left( \frac{e^{-\mu}}{1-e^{-\mu}} \right)$$

The units are pCi y/g Ca per mCi/km². The overall transfer to food categories, as measured by $P_{23}$, is highest for fruits (30.5) and
vegetables (15.5), due to longer-term availability and greater uptake of Sr-90 from soil. The value for grain products is 4.9, for milk 2.2, and for meat, fish, and eggs 1.1. When weighted for contribution to total Ca intake in diet, the contributions to total diet DealS, are 1.4 from vegetables, 1.3 from milk, 1.2 from fruit, 0.9 from grain products, and 0.1 from meat, fish and eggs. The total, 4.9 pCi y/gCa per mCi/km², from the separate fits to the food groups, agrees with the result from fitting the total diet Sr-Ca ratios. The fit to total diet data is shown in Figure 6. The model provides good correlation between deposition and diet levels, the correlation coefficients ranging from .93 for fruits, in which considerable scatter in the data is evident due to the diverse supply regions, to greater than .99 for total diet.

$D_n = \sum p_i (\theta_i \cdot \text{Year})^n$

Figure 6. Deposition model fit to Sr-90 in total diet in New York
Anticipated Sr-90 Intake in 1976

Atmospheric nuclear testing activity in recent years in the Northern Hemisphere has included one Chinese test of estimated total yield of 3 MT in each of 1968, 1969, and 1970. Low to intermediate yield tests were conducted by the Chinese in November 1971 and in January and March 1972. More recent, larger tests include one of 2–3 MT yield in June, 1973 and a 1 MT test in June, 1974. There were 5 French tests of varying yields in the South Pacific in 1968, 8 in 1970, 5 in 1971, 3 of low yield in 1972, 5 in 1973, and 7 of low to intermediate yield in 1974. There were no reported atmospheric tests during 1975. The larger atmospheric tests conducted during 1973 and 1974 caused a slight increase in Sr-90 deposition during 1974, however the deposition rate is now again decreasing. Dietary intake of Sr-90 remained little changed in 1975. The large contribution to diet attributed to uptake from the relatively constant cumulative deposit of Sr-90 in soil precludes large changes in Sr-90 intake from one year to the next. Based on the formula presented above, relating deposition to dietary intake, and assuming no increase in atmospheric testing programs, projected estimates of Sr-90 intakes in 1976 are 8.0±.4 pCi/day in New York and 3.0±.2 pCi/day in San Francisco.
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Ionizing Radiation: Levels and Effect.
FALLOUT $^{239,240}$Pu IN DIET

- 1974 Results -

by B. G. Bennett (HASL)

ABSTRACT

Ingestion intake of fallout $^{239,240}$Pu in New York in 1974 has been determined from a complete diet sampling. A similar sampling was conducted previously in 1972. Changes in concentrations and intake have been minor. Concentrations of plutonium in the 19 food categories ranged from .04 pCi/kg in shellfish to below the minimum detection level in canned vegetables and rice. For a very large sample of milk, the plutonium level was, this time, detectable. Annual intake in total diet is estimated to have been 1.5±.5 pCi in 1974. Combining the 1972 and 1974 results, the average intake during the period is estimated to have been 1.6±.3 pCi/ý. The record of fallout plutonium in tap water in New York from 1973 through 1975 is also reported.

Although the dose to man from fallout plutonium arises largely from inhalation intake (1), the long half-life and environmental persistence of $^{238}$Pu requires that the ingestion pathway also be investigated. A complete diet sampling, consisting of foods representing 19 separate categories, was conducted in 1974, repeating a similar sampling in 1972.(2) The foods were individually analyzed for $^{239,240}$Pu content, and the annual intake was determined from consumption estimates.
The foods were purchased during 1974 in New York as part of our regular program to determine the quarterly dietary $^{90}\text{Sr}$ intake. The foods were dry-ashed, and aliquots were taken for the $^{90}\text{Sr}$ analyses. The residual ash from the four quarterly sampling periods was composited in order to obtain 120 gram ash samples of the separate foods for $^{239,240}\text{Pu}$ determination. The large sample sizes are necessary because of the low plutonium concentrations present in the foods. The 1972 samples consisted of 100 grams of ash. Since plutonium in milk was not previously detectable, a 240 gram ash sample was formed. The food samples analyzed correspond to 3 to 47 kg quantities of the fresh foods. A total equivalent of 303 kg fresh food was used in the analyses.

PLUTONIUM CONCENTRATIONS IN FOOD

The results of the analyses for $^{239,240}\text{Pu}$ are listed in Table 1. The highest concentration (.04 pCi/kg) was found in shellfish. The sample consisted of clams and shrimp. Separate analyses had previously shown that most of the activity is in clams, (2) associated most probably with the gastrointestinal portion of these filter feeders. The meat portion of fresh fish (halibut, snapper, flounder) had a plutonium concentration 20 times less than the shellfish sample.
Other foods with higher plutonium concentrations were grain products and fresh vegetables and fruits. The concentration in chicken (meat only, no internal organs) was 70% higher than in 1972, apparently reflecting only a sampling variation. Lowest concentrations were found in milk and prepared or processed foods, such as peeled potatoes and canned foods. Below the minimum detection level (.02 dpm/sample) were canned vegetables and rice.
The concentration differences between fresh and canned vegetables and fruit indicate that external contamination is an important factor. Activity is lost through washing and processing. It was previously shown that the plutonium concentration on potato peels, which could be accounted for by the soil activity, exceeded the concentration in peeled potatoes by a factor of 60.(2)

The milk sample was a composite of 20 grams of ash from each monthly sampling of milk in New York during 1974, for a total of 240g ash, representing 33 liters of fresh milk. The activity in a 120g ash sample for 1972 was below the detection limit.(2) The concentration in the 1974 sample was 1.5 fCi/L. Sansom(3), in a tracer study, found very low transfer to milk ($10^{-6}$ of the ingested dose per liter) and suspected that external contamination may have been the actual transfer mechanism. The very low uptake of plutonium from the gastrointestinal tract in animals keeps concentrations low in milk and also in meat.

Since aliquots of the same ash were analyzed for $^{90}$Sr,(4) direct comparisons can be made of the plutonium and $^{90}$Sr occurrence. The comparisons can show excess or deficient amounts of plutonium compared to $^{90}$Sr, normalized to equal deposition amounts. The
$^{239,240}\text{Pu}/^{90}\text{Sr}$ ratio in the cumulative deposit in soil, including radioactive decay to 1974 is .035. The normalized occurrence ratio in food is thus .035 times the $^{90}\text{Sr}$ concentration divided by the $^{239,240}\text{Pu}$ concentration. The ratios are listed in Table 2. In shellfish, representing probable physical concentration primarily, rather than biological uptake, the $^{239,240}\text{Pu}$ and $^{90}\text{Sr}$ normalized occurrence are about equal. In all other foods $^{239,240}\text{Pu}$ is deficient relative to deposition amounts by factors of 10 or less in meat and fish, to which $^{90}\text{Sr}$ also shows no strong affinity, to factors of a

Table 2

<table>
<thead>
<tr>
<th>Pu Deficiency*</th>
</tr>
</thead>
<tbody>
<tr>
<td>SHELLFISH</td>
</tr>
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<td>POULTRY</td>
</tr>
<tr>
<td>MEAT</td>
</tr>
<tr>
<td>FRESH FISH</td>
</tr>
<tr>
<td>EGGS</td>
</tr>
<tr>
<td>BAKERY PRODUCTS</td>
</tr>
<tr>
<td>WHOLE GRAIN PRODUCTS</td>
</tr>
<tr>
<td>RICE</td>
</tr>
<tr>
<td>CANNED FRUIT</td>
</tr>
<tr>
<td>ROOT VEGETABLES</td>
</tr>
<tr>
<td>MACARONI</td>
</tr>
<tr>
<td>POTATOES</td>
</tr>
<tr>
<td>MILK</td>
</tr>
<tr>
<td>FLOUR</td>
</tr>
<tr>
<td>FRUIT JUICE</td>
</tr>
<tr>
<td>FRESH FRUIT</td>
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<tr>
<td>FRESH VEGETABLES</td>
</tr>
<tr>
<td>CANNED VEGETABLES</td>
</tr>
<tr>
<td>DRY BEANS</td>
</tr>
<tr>
<td>TOTAL DIET</td>
</tr>
</tbody>
</table>

*Normalized to equal cumulative deposition ($^{90}\text{Sr} \times .035/^{239,240}\text{Pu}$)
hundred or more in other foods. Overall, fallout plutonium was deficient by a factor of 80 in total diet relative to $^{90}$Sr. This is the same result that was obtained from the 1972 data.(2)

**PLUTONIUM INGESTION INTAKE**

Ingestion intake estimates of plutonium in the various foods are obtained from consumption statistics (5) and the concentration data. These results are shown in Table 3. Those items which are consumed in the

<table>
<thead>
<tr>
<th></th>
<th>CONSUMPTION (kg/y)</th>
<th>CONCENTRATION (pCi/kg)</th>
<th>INTAKE (pCi/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>MILK</strong></td>
<td>200</td>
<td>.0015</td>
<td>.30</td>
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<td><strong>MEAT</strong></td>
<td>79</td>
<td>.0025</td>
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<td>44</td>
<td>.0040</td>
<td>.18</td>
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<td><strong>FRESH VEGETABLES</strong></td>
<td>48</td>
<td>.0031</td>
<td>.15</td>
</tr>
<tr>
<td><strong>FRESH FRUIT</strong></td>
<td>59</td>
<td>.0025</td>
<td>.15</td>
</tr>
<tr>
<td><strong>TAP WATER</strong></td>
<td>1200</td>
<td>.0003</td>
<td>.15</td>
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<tr>
<td><strong>POULTRY</strong></td>
<td>20</td>
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<td><strong>WHOLE GRAIN PRODUCTS</strong></td>
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<td>.0050</td>
<td>.055</td>
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<td><strong>POTATOES</strong></td>
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<td>.046</td>
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<td><strong>SHELL FISH</strong></td>
<td>1</td>
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<td>.040</td>
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<td>34</td>
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<td><strong>ROOT VEGETABLES</strong></td>
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<td>.0019</td>
<td>.019</td>
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<td><strong>FRUIT JUICE</strong></td>
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<td>.019</td>
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<td><strong>CANNED VEGETABLES</strong></td>
<td>22</td>
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<td>&lt; .018</td>
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<td><strong>FRESH FISH</strong></td>
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<td>.014</td>
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<tr>
<td><strong>MACARONI</strong></td>
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<td>.00095</td>
<td>.0028</td>
</tr>
<tr>
<td><strong>RICE</strong></td>
<td>3</td>
<td>&lt; .00078</td>
<td>&lt; .0023</td>
</tr>
</tbody>
</table>

**TOTAL** 1.5 pCi/y
greatest amounts are the most important contributors to fallout plutonium dietary intake - meat and poultry, milk, bakery products, fresh vegetables, fresh fruit, and tap water.

The estimated dietary intake of fallout $^{239,240}$Pu during 1974 in New York is 1.5±.5 pCi. This intake is due 26% to meat, poultry, eggs and fish, 20% to dairy products, 18% to grain products, 15% to vegetables, 11% to fruit and 10% to tap water.

**PLUTONIUM IN DIET, 1972-74**

The changes in plutonium concentration of food items from 1972 to 1974 are illustrated in Figure 1. The error bars reflect the estimated standard deviations based on counting statistics. Within these uncertainties, there have been few changes in plutonium concentrations in

![Figure 1. Changes in Pu-239,240 concentrations in foods from 1972 (left) to 1974 (right). The means of each pair of determinations (points) are weighted according to indicated counting uncertainties. Values below the minimum detection level (±100% uncertainty) have no downside uncertainty indicated.](image)
foods between 1972 and 1974. Increases are noted for shellfish, poultry, and milk and decreases for bakery products, fresh fruit, root vegetables, and flour.

Assuming that the variations in plutonium concentrations are due to sampling, the estimated mean concentrations for 1972-74 have been obtained, indicated by the points in Figure 1. The means are weighted according to estimated counting uncertainties. The combined results are listed in Table 4. The mean concentrations reflect the best estimates

<table>
<thead>
<tr>
<th>Table 4</th>
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</thead>
<tbody>
<tr>
<td><strong>FALLOUT $^{239,240}$Pu DIETARY INTAKE</strong></td>
</tr>
<tr>
<td><strong>New York 1972-74</strong></td>
</tr>
<tr>
<td><strong>CONSUMPTION (kg/y)</strong></td>
</tr>
<tr>
<td>SHELLFISH</td>
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<tr>
<td>BAKERY PRODUCTS</td>
</tr>
<tr>
<td>WHOLE GRAIN PRODUCTS</td>
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<td>FRESH FRUIT</td>
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<td>MILK</td>
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<tr>
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</tr>
<tr>
<td>CANNED FRUIT</td>
</tr>
<tr>
<td>TAP WATER</td>
</tr>
</tbody>
</table>

**TOTAL 1.6 ± .3 pCi/y**
of current levels of fallout plutonium in diet. Estimated dietary intake from this concentration data is 1.6±.3 pCi/y. Contributing to the annual intake are grain products, 30%; meat, poultry, eggs and fish, 21%; vegetables, 17%; fruit, 17%; tap water, 9%; and dairy products, 6%.

PLUTONIUM IN TAP WATER

Tap water has been sampled weekly and analyzed quarterly for fallout plutonium since 1973 in New York by the Health and Safety Laboratory. The sample size is 300 liters. The results of these measurements are shown in Figure 2. The concentrations range from 0.08 to 0.6 fCi/L. The plutonium levels in tap water have shown a seasonal variation, with highest levels in springtime, corresponding to peak deposition and runoff into the watershed. The annual means have been 0.22±0.04 in 1973, 0.30±0.03 in 1974, and 0.25±0.02 fCi/L in 1975. The mean for the entire period 1973-75 is 0.26±0.02 fCi/L.

Figure 2. $^{239,240}$Pu in Tap Water in New York.
During the same period 1973-75, $^{90}$Sr in New York tap water was 0.49 pCi/L and $^{137}$Cs was 0.044 pCi/L. The Pu/Sr and Pu/Cs ratios for tap water are thus 0.00053 and 0.0059, respectively. Since the corresponding ratios for decayed cumulative deposition are 0.033 and 0.021, $^{239,240}$Pu is deficient in tap water relative to $^{90}$Sr occurrence by a factor of 60 and relative to $^{137}$Cs by a factor of 3.5. As was shown before (2), plutonium behaves more similarly to cesium with respect to the amounts which become removed to sediments.

CONTRIBUTION TO BODY BURDEN

Uptake of plutonium from the gastrointestinal tract is estimated to range from $3 \times 10^{-5}$ to $10^{-6}$. On this basis, the 1.6 pCi/y ingestion intake contributes, at most, $5 \times 10^{-5}$ pCi/y to the body burden. During 1972-74, inhalation intake averaged 0.2 pCi/y, of which approximately 20%, or $4 \times 10^{-2}$ pCi/y, contributes to the initial lung and body burden. As deposition and air activity of fallout plutonium decrease, the ratio of inhalation intake to ingestion intake declines. Currently, however, the inhalation pathway is more important by a factor of 1000 compared to the ingestion pathway in contributing to body burden.
ACKNOWLEDGEMENT

The plutonium analyses in food were performed by Norton Chu and the tap water analyses by Jerry Feldstein, Chemists, Health and Safety Laboratory.

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ICRP Publication 19 (1972)
ABSTRACT

The stratospheric inventories of Sr-90, Zr-95, Cs-137 and Pu-239+240 are reported up to July 1975. The stratospheric burden of Sr-90 dropped to 28 kCi. The ratios of the Cs-137 inventories to those of Sr-90 are close to the theoretical production value of 1.44.

This is the tenth in a series of reports (1-9) which updates the Sr-90, Zr-95, Cs-137, and Pu-239+ (indicating Pu-239 and Pu-240) stratospheric inventories. The method of calculation has been described earlier (6).

CONCENTRATION CONTOURS AND INVENTORIES

The stratospheric concentration contours for Sr-90 and Zr-95 for October 1974, April 1975, and July 1975 are illustrated in Figures 1 through 6. These contours are developed from data obtained by ERDA's aircraft and balloon sampling programs in the stratosphere. If the balloon launches did not occur in the same month as the aircraft mission, the balloon data were corrected to the aircraft sampling dates by the appropriate nuclide decay and by a stratospheric half-residence time of 10 months (10). Because of budgeting and other compelling considerations, the aircraft sampling program
as it was structured terminated after the April 1974 mission. The missions, formerly flown by the Air Force, extended latitudinally from 75°N to 51°S with a slight gap between 10°S to 16°S and were completed within 9 days. The National Aeronautics and Space Administration conducts the missions which now range latitudinally from 75°N to 10°S and are completed within 30 days (11).

The concentration contours for Cs-137 and Pu-239+ have also been prepared but are not presented because of their similarity to the Sr-90 distributions. Due to the low levels, our estimates of the Pu-238 concentrations in the stratosphere are highly uncertain and have, therefore, been discontinued. The stratospheric inventories of Cs-137 and Pu-239+ in addition to the inventories of Sr-90 and Zr-95, are given in Table 1. The accuracy of these inventories is estimated to be about ±25% based on ongoing studies. Figure 7 illustrates the hemispheric and total stratospheric burdens of Sr-90 from 1963 to July 1975. The stratospheric burden of Sr-90 in the Northern Hemisphere for July 1975 is lower than at any time since the large nuclear test series in 1961 and 1962.

INVENTORY RATIOS

The ratios of several of the nuclide inventories in Table 1 are presented in Table 2. The 1974 and 1975 mean Cs-137/Sr-90
ratios and the inclusive mean Cs-137/Sr-90 ratio since 5/8/70 show
good agreement with the theoretical production ratio of 1.44 (12).

ATMOSPHERIC TESTS

The Chinese and the French each conducted an intermediate-
sized nuclear test during 1974. The Sr-90 injected into the
stratosphere can be calculated for the Chinese test which occurred
on June 17, 1974 in the Northern Hemisphere. The Sr-90 inventory
prior to the test (April 20, 1974 – 88 kCi) is corrected for strato-
spheric fallout to the sampling date, October 17, 1974, using a
stratospheric half-residence time of 10 months (10). The decay-
corrected inventory, which represents the Sr-90 remaining in the
stratosphere from earlier detonations at sampling time, is sub-
tracted from the measured inventory. The difference is then cor-
rected back to the test date with a 10 month half-residence time to
give the Sr-90 injected by the test.

The Sr-90 injection is also estimated from a similar calcu-
lation using the Zr-95 inventory. In addition to the 10 month
half-residence time, the Zr-95 data is corrected for radioactive
decay. The Sr-90 injected into the stratosphere can be estimated
by dividing the Zr-95 injected at shot time by the Zr-95/Sr-90
theoretical production ratio of 226 (12). The results of these
calculations, with a mean of 45 kCi, are summarized in Table 3.

Debris from the French test of September 15, 1974 can be seen penetrating the Northern Hemisphere stratosphere in the April 1975 concentration contour of Zr-95 (Figure 4). The ratios for Ce-144/Sr-90 (13) and Zr-95/Cs-137 are higher by factors of 2 and 4 respectively in the equatorial stratosphere. An estimation of the Zr-95/Sr-90 ratio in the April 1975 equatorial stratosphere was made to confirm that this is French debris. The mean concentrations of Sr-90 and Zr-95 in April 1975 were calculated for the samples in Figure 4 with concentrations of Zr-95 ≥100. Similar mean concentrations were calculated for samples in this equatorial stratospheric region before the French debris became evident (October 1974). The October 1974 concentrations were corrected to the April 1975 sampling date using the appropriate radioactive decay and a 10 month stratospheric half-residence time. The results, summarized in Table 4, show good agreement between the observed and the theoretical Zr-95/Sr-90 ratio decayed from the date of the French test, indicating that this is French debris.
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USAEC Report HASL-281, April (1974), I-130 to I-142

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with Observations of Radioactive Debris
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12) Harley, N., I. Fisenne, L.D.Y. Ong and J. Harley  
Fission Yield and Fission Product Decay  
USAEC Report HASL-164, October (1965), pp. 251-261

13) Leifer, R., L. Toonkel and M. Schonberg  
Project Airstream  
USERDA Report HASL-302, April (1976), pp. II-7 to II-84
Table 1

NORTHERN HEMISPHERE
STRATOSPHERIC INVENTORIES, kCi

<table>
<thead>
<tr>
<th>DATE</th>
<th>Sr-90</th>
<th>Zr-95</th>
<th>Cs-137</th>
<th>Pu-239+</th>
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<tbody>
<tr>
<td>10/17/74</td>
<td>87</td>
<td>2480</td>
<td>140</td>
<td>1.6</td>
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<td>4/28/75</td>
<td>46</td>
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<tr>
<td>7/27/75</td>
<td>28</td>
<td>29</td>
<td>43</td>
<td>.58</td>
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</tbody>
</table>

Table 2

NORTHERN HEMISPHERE
NUCLIDE INVENTORY RATIOS

<table>
<thead>
<tr>
<th>DATE</th>
<th>Cs-137/Sr-90</th>
<th>Pu-239+/Sr-90</th>
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</thead>
<tbody>
<tr>
<td>10/17/74</td>
<td>1.6</td>
<td>.018</td>
</tr>
<tr>
<td>1974 mean</td>
<td>1.7±1a</td>
<td>.020±.003a</td>
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<tr>
<td>4/28/75</td>
<td>1.5</td>
<td>.020</td>
</tr>
<tr>
<td>7/27/75</td>
<td>1.5</td>
<td>.021</td>
</tr>
<tr>
<td>1975 mean</td>
<td>1.5±0</td>
<td>.021±.001</td>
</tr>
<tr>
<td>Inclusive mean</td>
<td>1.5±.2b</td>
<td>.020±.003c</td>
</tr>
</tbody>
</table>

a: Included ratios from ref. 9
b: Since 5/8/70
c: Since 7/27/72

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Table 3

STRATOSPHERIC INJECTION OF Sr-90 (kCi)
FROM CHINESE TEST OF JUNE 17, 1974

<table>
<thead>
<tr>
<th>SAMPLING MISSION</th>
<th>ESTIMATE DERIVED FROM</th>
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<td>Sr-90 ANALYSES</td>
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<tr>
<td>10/17/74</td>
<td>38</td>
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</table>

Mean : 45
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<tr>
<th></th>
<th>Sr-90</th>
<th>Zr-95</th>
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<tbody>
<tr>
<td>Mean Measured Concentration - 4/28/75</td>
<td>18</td>
<td>140</td>
</tr>
<tr>
<td>Mean Measured Concentration - 10/17/74</td>
<td>21</td>
<td>460</td>
</tr>
<tr>
<td>Concentration Corrected to 4/28/75</td>
<td>14</td>
<td>38</td>
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<tr>
<td>Estimated Injection from French Test - 4/28/75</td>
<td>4.1</td>
<td>100</td>
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<tr>
<td>Observed Ratio - 4/28/75</td>
<td>24</td>
<td></td>
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<tr>
<td>Theoretical Production Ratio Corrected to 4/28/75</td>
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<td>21</td>
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Fig. 1. CONCENTRATION OF Sr-90 IN STRATOSPHERIC AIR (pCi/100 SCM) October 1974
Fig. 2. CONCENTRATION OF Zr-95 IN STRATOSPHERIC AIR (pCi/100 SCM) October 1974
Fig. 3. CONCENTRATION OF Sr-90 IN STRATOSPHERIC AIR
(pCi/100 SCM) April 1975
Fig. 4. CONCENTRATION OF Zr-95 IN STRATOSPHERIC AIR
(pCi/100 SCM) April 1975

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Fig. 5. CONCENTRATION OF Sr-90 IN STRATOSPHERIC AIR (pCi/100 SCM) July 1975
Fig. 6. CONCENTRATION OF Zr-95 IN STRATOSPHERIC AIR
(pCi/100 SCM) July 1975
STRATOSPHERIC INVENTORY of Sr-90

- Total Stratosphere
- Northern Hemisphere
- Southern Hemisphere

Large Atmospheric Tests in Respective Hemisphere

Fig. 7
TABLE OF RADIONUCLIDES

Isabel M. Fisenne (HASL)

The following table is a revised listing of radionuclides of interest in the HASL environmental program. The half-life values are currently used at HASL and are subject to change as new data become available.

The three major references consulted in developing this tabulation are:

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   Laboratoire de Méterologie des Rayonnements Ionisants, Gif-sur-Yvette, France, 1975
The following table is a revised (6/15/76) listing of radionuclides of interest in the HASL environmental program. The half-life values are currently used at HASL and are subject to change as new data become available.

<table>
<thead>
<tr>
<th>MUCLIDE</th>
<th>RADIOACTIVE DAUGHTER</th>
<th>HALF-LIFE</th>
<th>OTHER UNITS</th>
<th>MUCLIDE</th>
<th>RADIOACTIVE DAUGHTER</th>
<th>HALF-LIFE</th>
<th>OTHER UNITS</th>
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<tr>
<td>Mission Products</td>
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<td></td>
<td>Reactor Corrosion and Activation Products</td>
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<tr>
<td>85Kr</td>
<td></td>
<td>3919.1</td>
<td>10.73y</td>
<td>16N</td>
<td>7.14s</td>
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<td>89Sr</td>
<td>39</td>
<td>50.55</td>
<td></td>
<td>24Na</td>
<td>15.0h</td>
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<tr>
<td>90Sr</td>
<td>39</td>
<td>10446.2</td>
<td>28.6y</td>
<td>51Cr</td>
<td>27.7</td>
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<tr>
<td>90Y</td>
<td>39</td>
<td>2.67</td>
<td>64.1h</td>
<td>54Mn</td>
<td>312.5</td>
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<tr>
<td>95Zr</td>
<td>41Nb</td>
<td>65.</td>
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<td>56Mn</td>
<td>2.585h</td>
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<td>103Ru</td>
<td>45Rh</td>
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<td>59Fe</td>
<td>45.1</td>
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<tr>
<td>106Ru</td>
<td>45Rh</td>
<td>368.2</td>
<td>1.01y</td>
<td>58Co</td>
<td>70.78</td>
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<tr>
<td>115mCd</td>
<td>48</td>
<td>44.6</td>
<td></td>
<td>60Co</td>
<td>1921.2</td>
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<tr>
<td>125Sb</td>
<td>125mTe</td>
<td>1011.7</td>
<td>2.77y</td>
<td>63Ni</td>
<td>3.66X10^4</td>
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<tr>
<td>129I</td>
<td>51Te</td>
<td>5.73X10^9</td>
<td>1.57X10^7y</td>
<td>64Cu</td>
<td>12.71h</td>
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<td>131I</td>
<td></td>
<td>8.04</td>
<td></td>
<td>65Zn</td>
<td>244.1</td>
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<td>133I</td>
<td>54Se</td>
<td>5.25</td>
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<td>102Rh</td>
<td>45Rh</td>
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<td>134Cs</td>
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<td>752.78</td>
<td>2.061y</td>
<td>102mRh</td>
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<td>136Cs</td>
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<td>12.98</td>
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<td>45Rh</td>
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<td>137Cs</td>
<td>137mBa</td>
<td>11021.1</td>
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<td>140Ba</td>
<td>140La</td>
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<td>49Ag</td>
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<td>141Ce</td>
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<td>1.675</td>
<td>40.2h</td>
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<td>144Ce</td>
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<td>147Nd</td>
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The following table is a revised (6/15/76) listing of radionuclides of interest in the HASL environmental program. The half-life values are currently used at HASL and are subject to change as new data become available.

<table>
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<th>HALF-LIFE</th>
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<tr>
<td>Cosmogenic</td>
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<td>3 H</td>
<td>4492.6</td>
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<td>7 Be</td>
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<td>14 C</td>
<td>2.09x10^6</td>
<td>5730y</td>
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<tr>
<td>22 Na</td>
<td>950.4</td>
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<tr>
<td>Terrestrial</td>
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<tr>
<td>40 K</td>
<td>4.60x10^11</td>
<td>1.26x10^9y</td>
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<tr>
<td>210 Pb</td>
<td>8145.1</td>
<td>22.3y</td>
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<td>226 Ra</td>
<td>5.84x10^5</td>
<td>1600y</td>
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<td>228 Th</td>
<td>3.624</td>
<td>3.05m</td>
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<td>208 Pb</td>
<td>1058.5</td>
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<td>236 U</td>
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<td>228 Th</td>
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<td>242 Pu</td>
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<td>243 Am</td>
<td>1.64x10^12</td>
<td>4.49x10^9y</td>
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<td>240 Pu</td>
<td>2.70x10^6</td>
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<td>243 Am</td>
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<td>239 Np</td>
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<td>242 Pu</td>
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<td>240 Pu</td>
<td>8.91x10^6</td>
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<tr>
<td>244 Cm</td>
<td>6614.7</td>
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PART II

HASL FALLOUT PROGRAM DATA
1. Fallout Deposition

1.1 Monthly Precipitation

1.11 $^{90}$Sr and $^{89}$Sr in Monthly Deposition at World Land Sites

Precipitation and dry fallout are collected over monthly periods at stations in the United States and overseas. The samples are analyzed for $^{90}$Sr and prior to 1971, for $^{89}$Sr whenever possible. A description of the sampling network and available data for each site are given in the Appendix, Section A.

1.12 Other Isotopes at Selected Sites

At a number of stations in the United States, monthly deposition collections were analyzed for radiostrontium and other nuclides of interest. Multinuclide analyses were discontinued as of July 1967 and the complete data reported in HASL-193, p. II-4 thru II-25.

Plutonium analyses of monthly deposition were carried out at New York City, Melbourne, Seattle, Honolulu, Salisbury, Durban, and Rio de Janeiro. This program has been terminated and the complete data are given in HASL-237 on pages II-4 thru II-13.

1.2 $^{90}$Sr and $^{89}$Sr Deposition at Atlantic Ocean Weather Stations

Measurements of radiostrontium in precipitation and dry fallout collections at four U. S. Coast Guard Stations in the North Atlantic Ocean were carried out for comparison with land stations in the same latitude band. This program was terminated in the Spring of 1972. A description of the stations and available data are given on pp II-4 thru II-17 of HASL-274.

2. Surface Air Sampling Program

The Health and Safety Laboratory has been collecting surface air particulate samples at stations in the Western Hemisphere since January 1963. The filters are analyzed for a number of fission and activation product radionuclides as well as stable lead. A description of the program and available data are given in the Appendix, Section B.
3. **Project Airstream**  
The Health and Safety Laboratory measures radioactivity in the lower stratosphere employing the WB-57F aircraft as a sampling platform. The aircraft are now flown by the National Aeronautics and Space Administration. The missions are scheduled three times a year and the coverage extends from 75°N to 10°S latitude in the Western Hemisphere. Air filter samples are collected from 12 to 20 km altitude and analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-7 to II-84 of HASL-302, April 1, 1976.

4. **High Altitude Balloon Sampling Program**  
Balloon borne filtering devices are used to collect nuclear debris at altitudes from 20 - 27 km. Balloon launchings are conducted annually at Fairbanks, Alaska, 65°N and the Panama Canal Zone (9°N), and three times a year at Alamogordo, N. M. (33°N). Filters are analyzed for a number of radionuclides. A more complete description of the program and available data are given on pages II-9 to II-26 of this report.

5. **Radiostrontium in Milk and Tap Water**  
Strontium-90 levels in fresh milk distributed in New York City and tap water sampled at the Health and Safety Laboratory, have been measured on a monthly basis since 1954. Cesium-137 has been analyzed in tap water since 1965. These data are summarized in tabular and graphical form in the Appendix, Section C, of this report.

6. **Strontium-90 in Diets**  
Quarterly estimates of the annual dietary intake of Sr-90 of New York City and San Francisco residents have been made based on analyses of foods purchased at these cities every three months since 1960. Sampling in Chicago was discontinued in 1967. The program is described and the most recent data reported on pages II-4 to II-6 of this report. The data through 1975 are evaluated beginning on p. I-93 of this report.

7. **Strontium-90 in Human Bone**  
Specimens of human vertebrae from New York City and San Francisco have been made available for Sr-90 analysis since 1961. The results for 1974 are evaluated beginning on page I-21 of HASL-297, October 1, 1975.

Human vertebral specimens are also received, through the World Health Organization, from countries where western-type diets are not typical. The Sr-90 data for samples received in 1975 are reported on pages II-7 and II-8 of this report.
6. HASL Diet Studies: 4th Quarter 1975

Results of the measurements of the $^{90}\text{Sr}$ content of foods purchased in New York City and San Francisco during the fourth quarter of 1975 are given in the following table. Also listed are estimates of the total diet intake of $^{90}\text{Sr}$ based on these measurements and on recent consumption statistics compiled by the U. S. Department of Agriculture.¹

The estimates of dietary intake of $^{90}\text{Sr}$ are a continuation of the HASL Tri-City diet studies which were started in March of 1960. Results of the earlier measurements along with those made during the fourth quarter of 1975 are shown graphically in the figure on page II-6. More detailed discussion of the results for the entire sampling program through 1975 is presented in Part I of this report.² A description of the sampling methods and philosophy of the HASL diet studies is given in HASL-147.³

REFERENCES

1. Food Consumption of Households in the United States - Spring 1964

2. Bennett, B. G.
   Strontium-90 in the Diet - Results through 1975

3. Rivera, J. and Harley, J.H.
   HASL Contributions to the Study of Fallout in Food Chains
   USAEC Report HASL-147, July (1964)
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YEARLY INTAKE 370g
DAILY INTAKE 8.0 pCi/gCa

NEW YORK CITY - NOVEMBER

SAN FRANCISCO - DECEMBER
DAILY INTAKE OF STRONTIUM-90

Picocuries Sr90 per gram Ca

- New York City
- Chicago
- San Francisco

In the work of the United Nations Scientific Committee on the Effects of Atomic Radiation, the major effort in evaluating fallout from weapons tests is directed towards estimating the dose commitment. A major contributor to bone dose is strontium-90 but there have not been adequate data for many regions in the world. The Committee decided that it would be useful to have even limited data for portions of the world not otherwise covered. It was agreed that some estimate could be made based on a small number of samples of adult vertebral bone and they requested the World Health Organization to assist in procuring samples.

Dr. E.I.I. Komorov of WHO co-ordinated this program through 1971. Dr. W. Seelentag was the WHO representative in charge through 1975, and the current representative is Dr. V. B. Vouk.

Samples for 1969 were received from Senegal, Jamaica, Chile and Venezuela. Samples for 1970 were received from Senegal, Uganda, Jamaica, Venezuela, Indonesia and Thailand. Samples for 1971 were received from Indonesia and Thailand. The samples were analyzed by the Health and Safety Laboratory and the results for these three years were presented in HASL-245, pp II-4 through II-7, October 1, 1971. Dr. Eduardo Penna-Franca of Brazil analyzed human specimens of vertebrae under his own program. Data for samples collected in 1969 were included in the tabulation in HASL-245.

No samples were collected in 1972, but samples were received from Thailand, Indonesia, and India in 1973. These data were reported in HASL-281, pp II-7 and 8.

Samples were received from Suva, Fiji Islands, New Guinea and Nepal in 1974 and from Nepal in 1975. These data were reported in HASL-297, pp II-7 and 8.

Additional specimens were made available from Nepal in 1975 and a set was also received from India. These data are given on the following page.
<table>
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<th>Age (yr)</th>
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<th>pCi 88Sr per g Ca</th>
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<td>22</td>
<td>M</td>
<td>1.8</td>
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</table>

*duplicate analysis
4. HIGH ALTITUDE BALLOON SAMPLING PROGRAM

by Robert Leifer (HASL)
Lawrence E. Toonkel (HASL)
Mindy Schonberg (HASL)

The U. S. Energy Research and Development Administration's program for measuring upper atmospheric nuclear debris collected by balloon-borne filtering devices has been in continuous operation since 1956. This report contains results from samples collected in calendar year 1975. Previous reports on this program are listed in references 1 through 25.

Launch Sites and Collection Parameters

For the period of this report, balloon flights were usually made at altitudes of 21, 24 and 27 km at the locations given in Table 4a by the flight organization indicated. On some occasions due to mechanical difficulties the exact altitude was missed or the flight failed completely. Information pertaining to the collection of the balloon samples is provided by the Air Resources Laboratories of the National Oceanic and Atmospheric Administration where flight data prepared by the balloon operations organizations are summarized and evaluated. Altitude data are obtained from barometric readings on the balloon gondola and refer to pressure altitude in the ICAO Standard Atmosphere.
The entire sample is collected within ±0.6 km of the predominant altitude unless annotated with the symbol, @. This symbol indicates that:

1. The altitude varies greater than the allowed ±0.6 km, or
2. The altitude is estimated or uncertain because of flight operational difficulties, or
3. The volume assigned to the filter is uncertain or estimated also because of the flight operational difficulties.

**Sampling Units**

Collections at the lower altitudes are made with the "Direct Flow Sampler", referred to as Unit D7. This system utilizes one square foot of I.P.C. No. 1478 filter paper together with a Westinghouse motor and a Torrington 704 blower. A discussion of this sampling unit has been presented by Wood (26). A modification to the sampling door to provide a better seal has been developed by the Air Resources Laboratories. This modified unit, referred to as D7-M, was flight tested many times prior to September 1970 but then replaced the D-7 after that time. Because duplicate sampling units are flown on the same balloon for research conducted at the National Center for Atmospheric Research (NCAR), the individual samples are identified as D7M1 and D7M2.
Samples at the higher altitudes are collected by an Air Ejector pump, referred to as unit AE. This system employs two square feet of I.P.C. No. 1478 filter paper. The air is drawn through the filter by the aspirator action of escaping nitrogen gas released downstream of the filter. This sampler was developed by the Applied Science Division, Litton Systems, Inc. under Contract AT(ll-1)-401 to the U. S. Atomic Energy Commission (27).

A N A L Y S I S

The filters are forwarded to the Health and Safety Laboratory for analyses where they are coded with a HASL number. When samples are split for duplicate analysis, each half is assigned a separate HASL number. Two types of analyses are made: a non-destructive gamma measurement at HASL, and an individual radionuclide assay at a contractor laboratory.

Gamma Measurement

All samples are compressed and sealed into a 6 cm diameter by 3.5 cm depth aluminum can. The samples are then analyzed spectro-metrically for Be-7, Zr-95, Cs-137, and Ce-144 by lithium drifted germanium diode systems. These systems have been described earlier in reports on Project Aistream (29). In brief, the detectors are
closed-ended coaxial diodes of approximately 50 cm$^3$. The diode responses are stored in 3000 channels of a computer based pulse height analyzer. A computer program resolves and reduces these spectra by least squares fitting.

Radiochemical Analysis

Each filter collected by a successful flight is sent to a contractor laboratory and analyzed radiochemically. Because the Chinese and French conducted large atmospheric nuclear tests in recent years the stratospheric concentrations of both short and long-lived weapon related nuclides are of interest. These include Sr-89, Sr-90, Pu-238 and Pu-239. Pu-238 is of additional interest because about 17 kilo curies of this isotope were released in the upper atmosphere by the re-entry burn-up of the SNAP-9A power source in April 1964 (28). In fiscal year 1975 the samples were also analyzed for Pb-210 to complement the Airstream program in the stratospheric study of nuclides of terrestrial origin. In the Airstream program a positive bias of 15% was observed for Pb-210. Investigation and resolution of this problem has delayed the analysis of balloon samples as well. Thus, for this report only about half the Pb-210 analyses are available.
Quality Control Program

To evaluate the analytical performance of the contractor laboratories, HASL submits coded blank, duplicate and standard samples for analysis. A blank is an appropriate sized piece of unexposed IPC filter paper taken from the roll of paper currently used by the flight organizations. Duplicate samples are fractions of a single filter which are divided in half or filters from two samplers carried aloft on the same balloon. A standard is prepared by evaporating weighed aliquots of various tracer solutions onto a regulation size blank of IPC paper. These calibrated solutions are available from a number of sources (i.e., Radiochemistry Center, IAEA, Nuclear Chicago, etc.) and are recalibrated at HASL. Generally, the agreement between HASL's measurement and the reported value is very good. Because the handling and analysis of the balloon samples are very similar to Airstream and because the balloon program is small in terms of number of samples, the results of the Airstream quality control program are used in conjunction with the few balloon quality control samples for quality determinations.

RESULTS

Radiochemical Analyses

The radiochemical concentrations for samples collected in 1975 are given in Table 4b. The results of the quality control program are given in Table 4c. The concentrations in Table 4b are expressed in
units of picocuries per $10^3$ standard cubic meters of air (pCi/KSCM) at collection. To convert pCi/KSCM to dpm per $10^3$ standard cubic feet multiply by 0.0629. Most filters sample between 50 and 300 SCM depending upon altitude of collection. The volume of air filtered is computed at 1013 millibars and $15^\circ$C such that 1 SCM = 1.225 kilograms of air.

One standard deviation of the counting error for all data in Table 4b is less than ±20% unless annotated with these symbols:

A - One standard deviation of the counting error is between ±20-50%.

B - One standard deviation of the counting error is between ±51-100%.

* - Activity is not detectable. This designation is applied to data when one standard deviation of the counting error is greater than 100%.

? - The datum is considered suspect because:

1. The magnitude of the concentration is inconsistent with adjacent samples in space and time; or

2. The relative activity of the nuclide is inconsistent with other nuclides in the same sample.

The nuclide activity of each sample is corrected for the normal radiochemical parameters such as chemical yield and detection efficiency.
Quality Control

The blank data in Table 4c indicate that little or no contamination is introduced by the normal handling and radiochemical analyses of the filter. The inherent Sr-90 content of unexposed IPC-1478 paper can represent a significant part of the total Sr-90 activity collected at altitudes ≥27 km.

The percent deviation between duplicate samples in Table 4c represents the difference divided by the mean and expressed as a percent. In all cases the percent deviation between duplicates is within twice the counting uncertainty associated with the individual samples. In many cases the percent deviation is less than 10%. The plutonium analyses were lost on sample 3662.

The plutonium results of the standard samples in Table 4c indicate good analytical accuracy. The strontium and lead standards data suggest a bias of about -15 and +15 percent, respectively. This behavior reflects continuing problems. The lead problem was traced to Bi-210 contamination and should be absent in future analysis.
The gamma spectrometry of the balloon samples is identical to the method used in the spectral analysis of Project Airstream samples. The accuracy of this method has been discussed earlier (29) and the error has been shown to be less than ±15% for Zr-95, Cs-137 and Ce-144 in most cases.

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4) Ibid, HASL-149, p. 54, October (1964)

5) Ibid, HASL-140, p. 166, October (1963)

6) High Altitude Sampling Program (Project Ash Can)
   USAEC Report HASL-127, p. 151, July (1962)


12) Ibid, HASL-182, p. II-6, July (1967)

13) Ibid, HASL-184, p. II-26, January (1968)

21) Ibid, HASL-249, p. II-120, April (1972)
22) Ibid, HASL-259, p. II-161, October (1972)
23) Ibid, HASL-274, p. II-95, July (1973)

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Air Ejector Particle Sample, A Progress Report
Litton Systems, Inc., Report No. 2584, September 1, 1964

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29) Leifer, R., L.E. Toonkel and M. Schonberg
Project Airstream
USERDA Report HASL-302, pp II-7 - II-84, April (1976)
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<th>Location</th>
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<th>Flight Organization</th>
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<tr>
<td>Albrook Air Force Base, Panama Canal Zone</td>
<td>9°N</td>
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### TABLE 4b

**Stratospheric Radionuclide Concentrations**

**Balloon Samples Collected During May 1975**

**Latitude, 65°N Eielson Air Force Base, Alaska**

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<th>Altitude (km)</th>
<th>Flight Day</th>
<th>Hasl Number</th>
<th>Collection Unit</th>
<th>Analytical Laboratory</th>
<th>Volume (SCM)</th>
<th>PC/KSCM</th>
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**Radionuclides**

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*A: One standard deviation of counting error is >20% to 50% of count.*

*B: One standard deviation of counting error is >50% to 100% of count.*

*:* Standard deviation greater than data value
### TABLE 4b

**STRATOSPHERIC RADIONUCLIDE CONCENTRATIONS**

**Balloons samples collected during April 1975**

**Latitude, 33N Holloman Air Force Base, New Mexico**

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**Plutonium-239**

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</table>

**A:** One standard deviation of counting error is >20% to 50% of count.

**B:** One standard deviation of counting error is >50% to 100% of count.

***: Standard deviation greater than data value

**?: Data suspect
TABLE 4b

STRATOSPHERIC RADIONUCLIDE CONCENTRATIONS

BALLOON SAMPLES COLLECTED DURING JULY 1975
LATITUDE, 33° HOLLoman AIR FORCE BASE, NEW MEXICO

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| BERYLLIUM-7   | 8540 | 8180 | 8860 | 8020 |
| STRONTIUM-89  | 70.1B | 1388 | *   | *   |
| STRONTIUM-90  | 167  | 165  | 117  | 49.2 |
| ZIRCONIUM-95  | 147  | 47.3A | *   | 18.1B |
| CESIUM-137    | 249  | 247  | 171  | 63.7 |
| CERIUM-144    | 1110 | 1130 | 605  | 239 |
| PLUTONIUM-238 | *   | *   | .403A | * |
| PLUTONIUM-239 | 3.86 | 3.71 | 2.91 | .694A |

A: ONE STANDARD DEVIATION OF COUNTING ERROR IS >20% TO 50% OF COUNT.
B: ONE STANDARD DEVIATION OF COUNTING ERROR IS >50% TO 100% OF COUNT.
*: STANDARD DEVIATION GREATER THAN DATA VALUE
### Table 4b

**Stratospheric Radionuclide Concentrations**

*Balloon samples collected during October 1975, latitude 33N, Holloman Air Force Base, New Mexico*

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**Radionuclides:**

- Beryllium-7: 3860
- Strontium-89: *
- Strontium-90: 140
- Zirconium-95: *
- Cesium-137: 223
- Cerium-144: 605
- Plutonium-238: 3518
- Plutonium-239: 2.34A

**Notes:**

- A: One standard deviation of counting error is >20% to 50% of count.
- B: One standard deviation of counting error is >50% to 100% of count.
- *: Standard deviation greater than data value.
### TABLE 4b

**STRATOSPHERIC RADIONUCLIDE CONCENTRATIONS**

**BALLOON SAMPLES COLLECTED DURING NOVEMBER 1975**  
**LATITUDE, 33N HOLOMAN AIR FORCE BASE, NEW MEXICO**

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**PC/KSCM**

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A: ONE STANDARD DEVIATION OF COUNTING ERROR IS >20% TO 50% OF COUNT.  
*: STANDARD DEVIATION GREATER THAN DATA VALUE
TABLE 4b

STRATOSPHERIC RADIONUCLIDE CONCENTRATIONS

BALLOON SAMPLES COLLECTED DURING MARCH 1975
LATITUDE, 09N  ALBROOK AIR FORCE BASE, CANAL ZONE

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PC/KSCM

| BERYLLIUM-7   | 3140| 2580| 2340|
| STRONTIUM-89  | 484 | *   | *   |
| STRONTIUM-90  | 240 | 238 | 136 |
| ZIRCONIUM-95  | 1270| 55.3A| 29.4A|
| CESIUM-137    | 375 | 350 | 267 |
| CERIUM-144    | 2890| 1790| 819 |
| LEAD-210      | 9.39| 7.93| 4.33|
| PLUTONIUM-238 | .258A| .422A| .698A|
| PLUTONIUM-239 | 5.13 | 3.89 | 2.79 |

A: ONE STANDARD DEVIATION OF COUNTING ERROR IS >20% TO 50% OF COUNT.
*: STANDARD DEVIATION GREATER THAN DATA VALUE.
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<td>*</td>
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<td>+3.0</td>
<td>+16</td>
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<tr>
<td>3662</td>
<td>4/9/75</td>
<td>10500±3</td>
<td>42.5±29</td>
<td>200±1</td>
<td>226±10</td>
<td>341±5</td>
<td>1630±4</td>
<td>Lost</td>
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<td>16</td>
<td>13</td>
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<td></td>
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<td>40</td>
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<td>3672</td>
<td>7/20/75</td>
<td>8540±3</td>
<td>70.1±74</td>
<td>167±3</td>
<td>147±17</td>
<td>249±7</td>
<td>1110±3</td>
<td>Lost</td>
<td>*</td>
<td>3.86±10</td>
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<tr>
<td>3675</td>
<td>7/20/75</td>
<td>8180±3</td>
<td>138±75</td>
<td>165±8</td>
<td>47.3±42</td>
<td>247±7</td>
<td>1130±5</td>
<td>.403±36</td>
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<tr>
<td>% deviation between duplicates</td>
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<td>65</td>
<td>1.2</td>
<td>103</td>
<td>0.8</td>
<td>1.8</td>
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<td>4.0</td>
</tr>
</tbody>
</table>

*: Standard Deviation Greater than Data Value
Part III

DATA FROM SOURCES OTHER THAN HASL

Numerous environmental studies are conducted by other organizations in the United States and abroad. Some of these are sent to the editors for dissemination in these HASL Quarterly Reports. Submitted data are reproduced essentially as received and no interpretation by HASL is attempted.

1. National Tsing Hua University - Hsinchu, Taiwan
   Institute of Nuclear Science, Health Physics Section
   Environmental Radioactivity Surveys for Nuclear Power Plants in North Taiwan: Report for Year 1975
   by Pao-Shan Weng, Chun-Nan Hsu and Hung-Tsan Chang
   NTHU - HPS Report 5

2. Radioecological and Environmental Research Division
   Argonne National Laboratory
   Cesium-137 in Various Chicago Foods (collection month April 1976)
   by J. O. Karttunen

III - 1
ENVIRONMENTAL RADIOACTIVITY SURVEYS FOR NUCLEAR POWER PLANTS IN NORTH TAIWAN

Report for the Year 1975

PAO-SHAN WENG, CHUN-NAN HSU and HUNG-TSAN CHANG

Health Physics Section
Institute of Nuclear Science
National Tsing Hua University
Hsinchu, Taiwan 300
Republic of China

February 1, 1976
ABSTRACT

This report is the continuation of environmental radioactivity monitoring for the nuclear power plants in North Taiwan which are under construction. The gamma scan for ash samples from conventional power plant is included for comparison. A separation method for iron from yttrium has been developed by back-extraction with perchloric acid while the soil samples were analyzed with tri-n-butyl phosphate for $^{90}\text{Sr}$. 

III - 4
INTRODUCTION

The report presents the results of the environmental monitoring program at Chienhua power reactor site for the year 1975. The purposes of this program are to detect variations in the radioactive content of the power reactor construction site and environs and to compare the radioactivities thus obtained with other nuclear establishments in North Taiwan.

Samples were collected from Chienhua site and from locations approximately 5, 10, and 20 km from Chienhua. The sampling locations are given in Figs. 1, 2, 3, 4, and 5. The location 90 km (Hsinchu) from Chienhua is referred to as reference site.

Most of the samples were analyzed for total alpha and beta activity by direct counting after suitable preparation of the sample. The counting rates were converted to picocuries or microcuries by applying corrections measured for $^{230}$Th (for alpha particles) and $^{40}$K (for beta particles). Some of the samples were also analyzed for specific elements and nuclides. The analytical and counting procedures were similar to those used in 1973.

All results are expressed with one standard deviation derived from counting statistics or multiple experimental observations.

RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

1. Background Gamma Monitoring

Measurement stations consisting of thermoluminescent dosimeters CaSO$_4$: Dy, have been distributed in and near the reactor site as shown in Figs. 1 and 2 in an attempt to obtain representative results. The results are summarized in Table 1. Several monitoring posts were lost during the mid-monitoring period.

2. Airborne Particulates

Gummed papers and low volume air samplers have been used for accumulated particulate matter. The results are presented in Tables 2 and 3.
Fig. 1. The TLD Monitoring Stations at Chienhua Power Reactor Site.

Fig. 2. The TLD Monitoring Stations beyond Chienhua Power Reactor Site.
Fig. 3. The Sampling Stations at Chienhua Power Reactor Site.

Fig. 4. The Sampling Stations beyond Chienhua Power Reactor Site.
3. Rain Samples

The gross beta activity in precipitation collected on the Chienhua site and the neighbor villages are given in Table 4.

4. Surface Water Samples

The total beta activity of surface waters taken from Chienhua site are summarized in Table 5.

5. Ground Water

Chemical analysis was used to determine the $^{89}\text{Sr}$ concentrations in ground and surface water. The results are summarized in Tables 6 and 7.

6. Vegetation and Vegetables

Total beta and $^{89}\text{Sr}$ and gamma scan were made for vegetation and vegetable samples. The results are summarized in Table 8.
Table 1. Results of Background Gamma Monitoring from February 4, 1975 to December 12, 1975

<table>
<thead>
<tr>
<th>Station No.</th>
<th>Location</th>
<th>Average Exposure (mR/month)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>2/4/75-5/5/75</td>
</tr>
<tr>
<td>1</td>
<td>Meteorological Tower</td>
<td>12.14±0.16</td>
</tr>
<tr>
<td>2</td>
<td>Near Dinning Room</td>
<td>11.18±0.25</td>
</tr>
<tr>
<td>3</td>
<td>East of Reactor Valley</td>
<td>12.67±0.12</td>
</tr>
<tr>
<td>4</td>
<td>East of Reactor Valley</td>
<td>11.60±0.12</td>
</tr>
<tr>
<td>5</td>
<td>Survey Team Hill</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>West of Reactor Valley</td>
<td>12.27±0.22</td>
</tr>
<tr>
<td>7</td>
<td>Upper End of Reactor Valley</td>
<td>11.76±0.37</td>
</tr>
<tr>
<td>8</td>
<td>Near Reactor Building</td>
<td>11.78±0.43</td>
</tr>
<tr>
<td>9</td>
<td>Resettlement Village Primary School</td>
<td>12.06±0.09</td>
</tr>
<tr>
<td>10</td>
<td>Near Barracks</td>
<td>13.18±0.12</td>
</tr>
<tr>
<td>11</td>
<td>Touku</td>
<td>12.70±0.25</td>
</tr>
<tr>
<td>12</td>
<td>Shihmen Junior Middle School</td>
<td>11.90±0.12</td>
</tr>
<tr>
<td>13</td>
<td>Shihpeng Hill</td>
<td>10.12±0.67</td>
</tr>
<tr>
<td>14</td>
<td>Shihmen Primary School (Second Campus)</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>Shihmen Hill</td>
<td>11.22±0.31</td>
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<tr>
<td>16</td>
<td>Laomei Primary School</td>
<td>12.04±0.09</td>
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<tr>
<td>17</td>
<td>Shanchi Village</td>
<td>13.58±0.12</td>
</tr>
<tr>
<td>18</td>
<td>Old Tea Factory</td>
<td>12.58±0.12</td>
</tr>
<tr>
<td>19</td>
<td>Threshing Floor</td>
<td>11.74±0.43</td>
</tr>
<tr>
<td>20</td>
<td>Pond Bank</td>
<td>12.23±0.16</td>
</tr>
<tr>
<td>21</td>
<td>Near Telegraph Poles</td>
<td>11.97±0.06</td>
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<tr>
<td>22</td>
<td>Chinshan Transformer Station</td>
<td>10.83±0.09</td>
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<tr>
<td>23</td>
<td>Hopingtau Transformer Station</td>
<td>12.06±0.12</td>
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<tr>
<td>24</td>
<td>Old Pond</td>
<td>11.04±0.09</td>
</tr>
<tr>
<td>25</td>
<td>Tiaosih Village</td>
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<td>26</td>
<td>Office Near Meteorological Tower</td>
<td>12.98±0.06</td>
</tr>
<tr>
<td>27</td>
<td>Chaoli Village</td>
<td>12.41±0.09</td>
</tr>
<tr>
<td>28</td>
<td>Chaoliwei</td>
<td>12.74±0.12</td>
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<td>29</td>
<td>Shanchih Junior Middle School</td>
<td>12.07±0.12</td>
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<td>30</td>
<td>Tansui Transformer Station</td>
<td>11.53±0.06</td>
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<td>31</td>
<td>Tienmu Transformer Station</td>
<td>11.46±0.09</td>
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<td>32</td>
<td>Chungshan Transformer Station</td>
<td>13.05±0.12</td>
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<td>Sungshan Community No. 1</td>
<td>12.23±0.06</td>
</tr>
<tr>
<td>34</td>
<td>Sungshan Community No. 2</td>
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<tr>
<td>35</td>
<td>Sungshan Community No. 3</td>
<td></td>
</tr>
<tr>
<td>36</td>
<td>Chiu-Chiung Lin</td>
<td>11.37±0.06</td>
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Table 2. Total Beta Activity of Gummed Paper Samples Collected at Shihmen, Fu-Kuei-Chiao, Chienihua, Chinshan, and Hsinchu Stations

<table>
<thead>
<tr>
<th>Sampling Period</th>
<th>Shihmen</th>
<th>Fu-Kuei-Chiao</th>
<th>Chienhu</th>
<th>Chinshan</th>
<th>Hsinchu</th>
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<tbody>
<tr>
<td>1/1–1/31</td>
<td>(8.5±9.8) × 10⁻²</td>
<td>(7.1±4.6) × 10⁻²</td>
<td>(6.7±6.1) × 10⁻²</td>
<td>(9.7±11.6) × 10⁻²</td>
<td>(3.2±2.0) × 10⁻²</td>
</tr>
<tr>
<td>2/1–2/28</td>
<td>(5.8±6.0) × 10⁻²</td>
<td>(5.0±4.1) × 10⁻²</td>
<td>(5.8±5.7) × 10⁻²</td>
<td>(9.4±6.7) × 10⁻²</td>
<td>(2.7±3.3) × 10⁻²</td>
</tr>
<tr>
<td>3/1–3/31</td>
<td>(7.1±9.9) × 10⁻²</td>
<td>(6.4±3.9) × 10⁻²</td>
<td>(7.7±5.1) × 10⁻²</td>
<td>(9.0±7.5) × 10⁻²</td>
<td>(5.0±3.3) × 10⁻²</td>
</tr>
<tr>
<td>4/1–4/30</td>
<td>(6.8±7.9) × 10⁻²</td>
<td>(4.7±4.1) × 10⁻²</td>
<td>(8.9±9.3) × 10⁻²</td>
<td>(6.5±8.7) × 10⁻²</td>
<td>(3.4±2.8) × 10⁻²</td>
</tr>
<tr>
<td>5/1–5/31</td>
<td>(5.6±5.2) × 10⁻²</td>
<td>(5.6±4.3) × 10⁻²</td>
<td>(1.8±2.6) × 10⁻¹</td>
<td>(5.3±6.0) × 10⁻²</td>
<td>(2.0±2.7) × 10⁻²</td>
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<tr>
<td>6/1–6/30</td>
<td>(3.5±2.8) × 10⁻²</td>
<td>(2.7±2.2) × 10⁻²</td>
<td>(4.7±3.3) × 10⁻²</td>
<td>(6.3±4.6) × 10⁻²</td>
<td>(4.2±9.4) × 10⁻²</td>
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<td>7/1–7/31</td>
<td>(3.6±2.8) × 10⁻²</td>
<td>(1.6±2.2) × 10⁻²</td>
<td>(4.6±3.9) × 10⁻²</td>
<td>(9.6±16.8) × 10⁻²</td>
<td>(2.5±1.5) × 10⁻²</td>
</tr>
<tr>
<td>8/1–8/31</td>
<td>(2.5±2.3) × 10⁻²</td>
<td>(1.9±1.8) × 10⁻²</td>
<td>(2.2±1.2) × 10⁻²</td>
<td>(6.5±8.1) × 10⁻²</td>
<td>(1.7±1.3) × 10⁻²</td>
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<td>9/1–9/30</td>
<td>(2.9±1.5) × 10⁻¹</td>
<td>(2.0±2.5) × 10⁻²</td>
<td>(2.0±1.5) × 10⁻²</td>
<td>(3.0±1.9) × 10⁻²</td>
<td>(1.8±1.3) × 10⁻²</td>
</tr>
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<td>10/1–10/31</td>
<td>(3.6±2.9) × 10⁻¹</td>
<td>(3.4±1.8) × 10⁻²</td>
<td>(4.0±2.1) × 10⁻²</td>
<td>(2.4±2.6) × 10⁻²</td>
<td>(3.0±2.0) × 10⁻²</td>
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<td>11/1–11/30</td>
<td>(2.3±2.4) × 10⁻¹</td>
<td>(2.6±2.1) × 10⁻²</td>
<td>(1.9±2.0) × 10⁻²</td>
<td>(3.7±3.2) × 10⁻²</td>
<td>(2.7±1.9) × 10⁻²</td>
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<tr>
<td>12/1–12/3</td>
<td>(4.0±4.0) × 10⁻¹</td>
<td>(4.2±4.9) × 10⁻²</td>
<td>(4.0±3.2) × 10⁻²</td>
<td>(3.9±3.7) × 10⁻²</td>
<td>(2.8±2.2) × 10⁻²</td>
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Table 3. Total Beta Activity of Low Volume Air Sampler Filters Collected at Chinshan and Chienhua

<table>
<thead>
<tr>
<th>Sampling Period</th>
<th>Gross $\beta$ Activity (pCi/m$^3$)</th>
<th>Sampling Period</th>
<th>Gross $\beta$ Activity (pCi/m$^3$)</th>
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<tbody>
<tr>
<td></td>
<td>Chinshan</td>
<td></td>
<td>Chienhua</td>
</tr>
<tr>
<td>1/3-1/10</td>
<td>B</td>
<td>12/30-1/6</td>
<td>B</td>
</tr>
<tr>
<td>1/10-1/18</td>
<td>B</td>
<td>1/6-1/13</td>
<td>$(2.6 \pm 1.4) \times 10^{-1}$</td>
</tr>
<tr>
<td>1/18-1/25</td>
<td>B</td>
<td>1/13-1/20</td>
<td>$(5.1 \pm 1.5) \times 10^{-1}$</td>
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<tr>
<td>1/25-2/1</td>
<td>$(7.5 \pm 1.5) \times 10^{-1}$</td>
<td>1/20-1/27</td>
<td>$(7.1 \pm 15.3) \times 10^{-2}$</td>
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<tr>
<td>2/1-2/10</td>
<td>B</td>
<td>1/27-2/3</td>
<td>B</td>
</tr>
<tr>
<td>2/10-2/17</td>
<td>$(9.1 \pm 16.4) \times 10^{-8}$</td>
<td>2/3-2/10</td>
<td>B</td>
</tr>
<tr>
<td>2/17-2/24</td>
<td>$(4.9 \pm 1.65) \times 10^{-1}$</td>
<td>2/10-2/17</td>
<td>$(4.9 \pm 1.7) \times 10^{-1}$</td>
</tr>
<tr>
<td>2/24-3/1</td>
<td>$(1.9 \pm 0.2) \times 10^{0}$</td>
<td>2/17-2/24</td>
<td>B</td>
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<td>3/1-3/8</td>
<td>$(2.7 \pm 1.5) \times 10^{-1}$</td>
<td>2/24-3/2</td>
<td>B</td>
</tr>
<tr>
<td>3/3-3/15</td>
<td>B</td>
<td>3/3-3/10</td>
<td>B</td>
</tr>
<tr>
<td>3/22-4/2</td>
<td>B</td>
<td>3/17-3/24</td>
<td>$(4.8 \pm 1.6) \times 10^{-1}$</td>
</tr>
<tr>
<td>4/2-4/10</td>
<td>$(1.8 \pm 13.7) \times 10^{-8}$</td>
<td>3/24-3/31</td>
<td>$(1.1 \pm 1.5) \times 10^{-1}$</td>
</tr>
<tr>
<td>4/10-4/17</td>
<td>B</td>
<td>3/31-4/7</td>
<td>B</td>
</tr>
<tr>
<td>4/17-4/24</td>
<td>B</td>
<td>4/7-4/14</td>
<td>$(5.0 \pm 15.6) \times 10^{-3}$</td>
</tr>
<tr>
<td>4/24-5/1</td>
<td>B</td>
<td>4/14-4/21</td>
<td>B</td>
</tr>
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<td>5/1-5/8</td>
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<td>4/21-4/28</td>
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<td>5/8-5/15</td>
<td>B</td>
<td>4/28-5/5</td>
<td>B</td>
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<tr>
<td>5/15-5/22</td>
<td>$(1.9 \pm 1.7) \times 10^{-1}$</td>
<td>5/5-5/12</td>
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</tr>
<tr>
<td>5/22-5/29</td>
<td>$(9.1 \pm 1.7) \times 10^{-1}$</td>
<td>5/12-5/19</td>
<td>$(5.2 \pm 1.7) \times 10^{-1}$</td>
</tr>
<tr>
<td>5/29-6/5</td>
<td>$(1.0 \pm 0.2) \times 10^{0}$</td>
<td>5/19-5/26</td>
<td>$(1.3 \pm 1.7) \times 10^{-1}$</td>
</tr>
<tr>
<td>6/5-6/12</td>
<td>B</td>
<td>5/26-6/2</td>
<td>$(9.0 \pm 1.7) \times 10^{-1}$</td>
</tr>
<tr>
<td>6/12-6/19</td>
<td>B</td>
<td>6/2-6/9</td>
<td>B</td>
</tr>
<tr>
<td>6/19-6/26</td>
<td>$(1.1 \pm 0.2) \times 10^{0}$</td>
<td>6/9-6/16</td>
<td>$(5.0 \pm 16.7) \times 10^{-2}$</td>
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<td>Value</td>
<td>Date Range</td>
<td>Value</td>
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</tr>
<tr>
<td>6/26-7/3</td>
<td>B</td>
<td>6/16-6/23</td>
<td>B</td>
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<td>7/3-7/10</td>
<td>B</td>
<td>6/23-6/30</td>
<td>B</td>
</tr>
<tr>
<td>7/10-7/17</td>
<td>B</td>
<td>6/30-7/7</td>
<td>(1.8±1.7)x10⁻¹</td>
</tr>
<tr>
<td>7/17-7/24</td>
<td>(2.8±1.7)x10⁻¹</td>
<td>7/7-7/14</td>
<td>B</td>
</tr>
<tr>
<td>7/24-7/31</td>
<td>B</td>
<td>7/14-7/31</td>
<td>B</td>
</tr>
<tr>
<td>7/31-8/7</td>
<td>B</td>
<td>7/21-7/28</td>
<td>(1.3±0.2)x10⁰</td>
</tr>
<tr>
<td>8/7-8/14</td>
<td>B</td>
<td>7/28-8/4</td>
<td>B</td>
</tr>
<tr>
<td>8/14-8/21</td>
<td>B</td>
<td>8/4-8/11</td>
<td>B</td>
</tr>
<tr>
<td>8/21-8/28</td>
<td>B</td>
<td>8/11-8/18</td>
<td>B</td>
</tr>
<tr>
<td>8/28-9/4</td>
<td>B</td>
<td>8/18-8/25</td>
<td>B</td>
</tr>
<tr>
<td>9/4-9/11</td>
<td>B</td>
<td>8/25-9/1</td>
<td>B</td>
</tr>
<tr>
<td>9/11-9/18</td>
<td>B</td>
<td>9/1-9/8</td>
<td>(2.2±1.7)x10⁻¹</td>
</tr>
<tr>
<td>9/18-9/25</td>
<td>B</td>
<td>9/8-9/15</td>
<td>(2.6±16.9)x10⁻¹</td>
</tr>
<tr>
<td>9/25-10/2</td>
<td>(9.8±16.9)x10⁻²</td>
<td>9/15-9/22</td>
<td>(6.1±1.7)x10⁻¹</td>
</tr>
<tr>
<td>10/2-10/9</td>
<td>B</td>
<td>9/22-9/29</td>
<td>(1.5±0.2)x10⁰</td>
</tr>
<tr>
<td>10/9-10/16</td>
<td>B</td>
<td>9/29-10/6</td>
<td>(6.2±1.7)x10⁻¹</td>
</tr>
<tr>
<td>10/16-10/23</td>
<td>B</td>
<td>10/6-10/13</td>
<td>(5.4±16.8)x10⁻¹</td>
</tr>
<tr>
<td>10/23-10/30</td>
<td>B</td>
<td>10/13-10/20</td>
<td>B</td>
</tr>
<tr>
<td>10/30-11/6</td>
<td>B</td>
<td>10/20-10/27</td>
<td>(1.0±1.7)x10⁻¹</td>
</tr>
<tr>
<td>11/6-11/13</td>
<td>B</td>
<td>10/27-11/3</td>
<td>(1.4±1.7)x10⁻¹</td>
</tr>
<tr>
<td>11/13-11/20</td>
<td>B</td>
<td>11/3-11/10</td>
<td>B</td>
</tr>
<tr>
<td>11/20-11/27</td>
<td>B</td>
<td>11/10-11/17</td>
<td>(3.7±16.3)x10⁻²</td>
</tr>
<tr>
<td>11/27-12/4</td>
<td>B</td>
<td>11/17-11/24</td>
<td>(6.4±16.4)x10⁻²</td>
</tr>
<tr>
<td>12/4-12/11</td>
<td>B</td>
<td>11/24-11/30</td>
<td>(3.1±1.9)x10⁻¹</td>
</tr>
<tr>
<td>12/11-12/18</td>
<td>B</td>
<td>12/1-12/8</td>
<td>(1.1±1.6)x10⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>12/8-12/15</td>
<td>(6.1±16.4)x10⁻²</td>
</tr>
<tr>
<td></td>
<td></td>
<td>12/15-12/22</td>
<td>(2.3±1.7)x10⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>12/22-12/29</td>
<td>(3.7±1.7)x10⁻¹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>12/29-1/5</td>
<td>B</td>
</tr>
</tbody>
</table>

B = background
Table 4. Total Beta Activity of Rain Water Samples Collected at Chinshan Village, Chienhua Village, Shihmen Village, and Hsinchu City

| Sampling Period | Chienhua Village Precipitation | | | Hsinchu City Precipitation | | | | | Chinshan Village Activity | | | Shihmen Village Activity |
|-----------------|-------------------------------|----------------|----------------|----------------|----------------|---------------|----------------|---------------|---------------|----------------|----------------|
|                 | Gross Beta Activity           | Gross Beta Activity | | Gross Beta Activity | | | Gross Beta Activity | | | Gross Beta Activity | | |
|                 | cm | mCi/km² | pCi/liter | cm | mCi/km² | pCi/liter | cm | mCi/km² | pCi/liter | pCi/liter | pCi/liter |
| 1/1-1/31        | 10.2 | 3.6±0.2 | 34.9±2.2 | 13.3 | 1.4±1.8 | 10.7±13.2 | 48.9±2.1 | 46.7±1.6 |
| 2/1-2/28        | 10.8 | 0.3±0.2 | 3.2±1.5 | 10.7 | 0.9±0.9 | 8.4±8.2 | 10.3±1.7 | 1.9±1.5 |
| 3/1-3/31        | 29.6 | 1.3±0.4 | 4.5±1.4 | 28.2 | 9.4±7.4 | 33.2±26.1 | 9.9±2.4 | 10.9±2.4 |
| 4/1-4/30        | 15.0 | 0.6±0.3 | 3.7±2.2 | 20.0 | 5.2±8.4 | 26.1±42.0 | 74.6±2.4 | 5.2±1.4 |
| 5/1-5/31        | 42.1 | 5.5±1.3 | 13.1±3.1 | 38.8 | 3.6±3.6 | 9.4±9.4 | 1.8±2.9 | 4.6±2.9 |
| 6/1-6/30        | 47.9 | 2.2±1.4 | 4.5±2.9 | 38.1 | 3.3±3.1 | 8.6±8.1 | 1.4±2.8 | 0.7±2.8 |
| 7/1-7/31        | 22.5 | 6.2±0.7 | 22.7±3.3 | 31.0 | 2.6±3.4 | 8.4±10.9 | 1.2±2.8 | 1.9±2.9 |
| 8/1-8/31        | 12.8 | 0.4±0.2 | 2.8±1.6 | 39.0 | 1.5±2.0 | 3.8±5.0 | 0.3±1.5 | 3.4±1.6 |
| 9/1-9/30        | 9.3  | B | B | 19.0 | 0.6±0.5 | 3.3±2.7 | 0.5±1.7 | 2.3±1.7 |
| 10/1-10/31      | 11.4 | 0.1±0.2 | 1.1±1.8 | 7.8  | 0.2±0.0 | 2.1±0.5 | 0.2±1.8 | B |
| 11/1-11/30      | 10.5 | 0.1±0.2 | 0.6±1.8 | 4.0  | 0.2±0.2 | 6.0±4.2 | B | 0.9±1.8 |
| 12/1-12/31      | 17.5 | 0.9±0.3 | 5.1±1.9 | 11.6 | 1.0±0.9 | 9.0±7.9 | 4.6±1.9 | 3.0±1.9 |

B = background
Table 5. Total Beta Activity of Surface Waters at Chienhua Reactor Site

<table>
<thead>
<tr>
<th>Sampling Station</th>
<th>Gross Beta Activity pCi/liter</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st Qtr. 74</td>
</tr>
<tr>
<td>Upstream, No. 15</td>
<td>99.2±2.7</td>
</tr>
<tr>
<td>Downstream, No. 15</td>
<td>84.9±2.6</td>
</tr>
<tr>
<td>Pond, Resettlement Village</td>
<td>9.0±1.5</td>
</tr>
<tr>
<td>Pond, Chien-Tzu-Lu</td>
<td>4.6±1.4</td>
</tr>
<tr>
<td>Drinking Water, Shihmen</td>
<td></td>
</tr>
<tr>
<td>Drinking Water, Settlement Village</td>
<td>13.5±1.6</td>
</tr>
<tr>
<td>Drinking Water, Tanshui</td>
<td>70.7±2.4</td>
</tr>
<tr>
<td>Sea Water, Shihmen No. 8</td>
<td>2.1±1.8</td>
</tr>
<tr>
<td>Sea Water, Tiaoshih</td>
<td>2.1±1.8</td>
</tr>
<tr>
<td>Sea Water, Coolant Discharge</td>
<td>2.2±1.8</td>
</tr>
<tr>
<td>Sea Water, TPC Bridge No. 19</td>
<td>1.9±1.7</td>
</tr>
<tr>
<td>Sea Water, Survey Team No. 20</td>
<td>2.6±1.9</td>
</tr>
<tr>
<td>Sea Water, Chaoli No. 13</td>
<td>1.8±1.7</td>
</tr>
<tr>
<td>Sea Water, Penshui</td>
<td>1.9±1.7</td>
</tr>
</tbody>
</table>

B = background

Table 6. Total Beta Activity in Ground Water, pCi/liter

<table>
<thead>
<tr>
<th>Sampling Station</th>
<th>Sampling Date</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2/4</td>
</tr>
<tr>
<td>Chienhua</td>
<td>9.0±1.5</td>
</tr>
<tr>
<td>Chinshan</td>
<td>13.7±1.6</td>
</tr>
</tbody>
</table>

B = background

III - 14
Table 7. *Sr Concentration in Water at Chienhua Reactor Site

<table>
<thead>
<tr>
<th>Sampling Station</th>
<th>Activity in pCi/liter</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2nd Qtr. 75</td>
</tr>
<tr>
<td>Upstream, No. 15</td>
<td>1.10±0.54</td>
</tr>
<tr>
<td>Downstream, No. 15</td>
<td>0.36±0.57</td>
</tr>
<tr>
<td>Pond, Resettlement Village</td>
<td>0.88±0.57</td>
</tr>
<tr>
<td>Pond, Chien-Tzu-Lu</td>
<td>0.29±0.57</td>
</tr>
<tr>
<td>Drinking Water, Shihmen</td>
<td>0.50±0.64</td>
</tr>
<tr>
<td>Drinking Water, Resettlement Village</td>
<td></td>
</tr>
<tr>
<td>Drinking Water, Tanshui</td>
<td>0.76±0.60</td>
</tr>
<tr>
<td>Rain Water, Shihmen</td>
<td>B</td>
</tr>
<tr>
<td>Rain Water, Chinshan</td>
<td>B</td>
</tr>
<tr>
<td>Rain Water, Chinshan</td>
<td>2.13±0.63</td>
</tr>
<tr>
<td>Rain Water, Kuosheng</td>
<td>0.42±0.97</td>
</tr>
<tr>
<td>Ground Water, Chienhua</td>
<td>1.13±0.66</td>
</tr>
<tr>
<td>Ground Water, Chinshan</td>
<td>0.47±0.51</td>
</tr>
</tbody>
</table>

B = background
Table 8. The Activity of Vegetation and Vegetables Samples Taken Around the Chienhua Power Reactor Site (Potassium Included)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sampling Station</th>
<th>Members of Plant</th>
<th>Gross Beta Activity pCi/g (ash)</th>
<th>$^{40}$Sr pCi/g Ca (ash)</th>
<th>$^{40}$Sr pCi/g Ca (ash)</th>
<th>$^{40}$Sr pCi/g Ca (ash)</th>
</tr>
</thead>
<tbody>
<tr>
<td>New Tea</td>
<td>Chienlu Village 9 Lin</td>
<td>Leaves</td>
<td>379±11 335±16 310±18</td>
<td>46.6 28.7±1.1 616±24</td>
<td>45.3±0.8</td>
<td>45.3±0.8</td>
</tr>
<tr>
<td></td>
<td>Shih-Peng-Shan</td>
<td></td>
<td>343±11 372±17 260±16</td>
<td>58.3 36.7±1.2 630±21</td>
<td>37.6±1.0</td>
<td>37.6±1.0</td>
</tr>
<tr>
<td></td>
<td>Chiu-Chiung-Lin</td>
<td></td>
<td>352±11 303±16 255±16</td>
<td>59.3 30.6±1.6 516±27</td>
<td>80.2±1.3</td>
<td>80.2±1.3</td>
</tr>
<tr>
<td></td>
<td>Nei-Chung-Men</td>
<td></td>
<td>335±11 319±16 351±18</td>
<td>49.6 22.0±1.0 444±20</td>
<td>37.8±1.0</td>
<td>37.8±1.0</td>
</tr>
<tr>
<td>Old Tea</td>
<td>Chienlu Village 9 Lin</td>
<td>Leaves</td>
<td>431±18 400±12 349±17 286±17</td>
<td>74.5 60.5±1.3 812±17</td>
<td>52.2±0.9</td>
<td>52.2±0.9</td>
</tr>
<tr>
<td></td>
<td>Shih-Peng-Shan</td>
<td></td>
<td>693±22 463±12 388±17 301±17</td>
<td>80.0 74.0±1.6 925±20</td>
<td>59.2±1.3</td>
<td>59.2±1.3</td>
</tr>
<tr>
<td></td>
<td>Chiu-Chiung-Lin</td>
<td></td>
<td>654±21 525±13 351±17 317±18</td>
<td>82.9 80.6±1.5 972±18</td>
<td>78.5±1.2</td>
<td>78.5±1.2</td>
</tr>
<tr>
<td></td>
<td>Nei-Chung-Men</td>
<td></td>
<td>721±22 585±14 356±17 355±19</td>
<td>90.5 28.7±1.6 649±18</td>
<td>56.0±1.3</td>
<td>56.0±1.3</td>
</tr>
<tr>
<td>Sweet Potato</td>
<td>Chien-Tzu-Lu</td>
<td>Root</td>
<td>266±7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rice</td>
<td>Shihmen</td>
<td>Grains*</td>
<td>134±6</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sweet Potato</td>
<td>Shin-Peng-Shan</td>
<td>Leaves</td>
<td>324±12</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Without shell
7. Silt

The silt samples were taken from the sea bottom at 20 m depth along the coast of the reactor site area. The sampling stations are shown in Fig. 5. The gross alpha and beta activities and $^{90}\text{Sr}$ content are summarized in Table 9.

8. Soil

Soil samples consist of soil taken from the surface to a depth of 30 cm below the surface at location of interest as shown in Figs. 3 and 4. The total beta activity and $^{90}\text{Sr}$ content in soil are summarized in Table 10. The total alpha activity in soil is summarized in Table 11. Table 12 shows activity in soil samples without chemical treatment prior to counting. The gamma scan of soil samples is shown in Fig. 6, while the background of the counting system is shown in Fig. 7. All gamma emitters appearing in Figs. 6 and 7 are naturally occurring radioisotopes and belong to thorium series such as $^{212}\text{Pb}(\text{ThC'})$, $^{228}\text{Ac}(\text{MsTh2})$, $^{208}\text{Tl}(\text{ThC''})$, and $^{212}\text{Bi}(\text{ThC})$, uranium series such as $^{214}\text{Pb}(\text{RaB})$ and $^{214}\text{Bi}(\text{RaC})$, and $^{40}\text{K}$. The gamma scan for other types of samples is shown in Figs. 8 through 19. The gamma scan for ash samples from conventional power plant is also included in Figs. 20 and 21 for comparison.

### Table 9. The Activity of Sea Bottom Silt Samples

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Sampling Date</th>
<th>Gross Activity pCi/g (dry)</th>
<th>$^{90}\text{Sr}$ pCi/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Alpha (Untreated)</td>
<td>Untreated</td>
</tr>
<tr>
<td>1</td>
<td>7/4</td>
<td>$0.1\pm0.8$</td>
<td>$9.0\pm3.9$</td>
</tr>
<tr>
<td>2</td>
<td>7/4</td>
<td>$10.2\pm3.9$</td>
<td>$1.9\pm1.1$</td>
</tr>
<tr>
<td>3</td>
<td>7/4</td>
<td>$12.0\pm4.0$</td>
<td>$2.1\pm0.4$</td>
</tr>
<tr>
<td>4</td>
<td>7/4</td>
<td>$2.2\pm0.9$</td>
<td>$12.5\pm4.0$</td>
</tr>
<tr>
<td>5</td>
<td>7/4</td>
<td>$1.8\pm0.9$</td>
<td>$12.2\pm4.0$</td>
</tr>
</tbody>
</table>

B = background
Table 10. Total Beta Activity and $^{90}$Sr Content in Soil Samples within 20 km of the Chienhua Power Reactor Site

<table>
<thead>
<tr>
<th>No.</th>
<th>Soil Sampling Station</th>
<th>Gross Beta Activity (With Chemical Treatment)</th>
<th>**Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>January</td>
<td>April</td>
</tr>
<tr>
<td>1</td>
<td>Chinshan Transformer Station</td>
<td>4.2±1.1 (1.3±0.4) $\times 10^4$</td>
<td>3.9±1.1 (1.3±0.4) $\times 10^4$</td>
</tr>
<tr>
<td>2</td>
<td>Southwest Boundary No. 4</td>
<td>3.5±1.0 (1.1±0.3) $\times 10^4$</td>
<td>4.4±1.1 (1.4±0.4) $\times 10^4$</td>
</tr>
<tr>
<td>3</td>
<td>Chien-Tzu-Lu</td>
<td>3.7±0.9 (1.2±0.3) $\times 10^4$</td>
<td>3.0±0.8 (9.6±2.6) $\times 10^4$</td>
</tr>
<tr>
<td>4</td>
<td>Shih-Pong-Shan No. 11</td>
<td>3.0±0.9 (1.0±0.3) $\times 10^4$</td>
<td>4.6±1.0 (1.5±0.3) $\times 10^4$</td>
</tr>
<tr>
<td>5</td>
<td>Shihmen Primary School Second Campus No. 12</td>
<td>3.6±1.0 (1.2±0.3) $\times 10^4$</td>
<td>5.0±1.2 (1.6±0.4) $\times 10^4$</td>
</tr>
<tr>
<td>6</td>
<td>Chinshan Park</td>
<td>3.9±0.9 (1.3±0.3) $\times 10^4$</td>
<td>3.6±0.8 (1.2±0.3) $\times 10^4$</td>
</tr>
</tbody>
</table>
Table 11. Total Alpha Activity in Soil Samples within 20 km of the Chienhua Power Reactor Site

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Soil Sampling Station</th>
<th>January</th>
<th>April</th>
<th>July</th>
<th>October</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Chinshan Transformer Station</td>
<td>2.0±0.7</td>
<td>1.6±0.7</td>
<td>2.5±0.8</td>
<td>1.47±0.88</td>
</tr>
<tr>
<td>2</td>
<td>Southwest Boundary No. 4</td>
<td>1.6±0.7</td>
<td>1.1±0.7</td>
<td>2.4±0.8</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Chien-Tzu-Lu</td>
<td>2.3±0.7</td>
<td>1.5±0.7</td>
<td>1.5±0.7</td>
<td>1.97±0.98</td>
</tr>
<tr>
<td>4</td>
<td>Shih-Pong-Shan No. 11</td>
<td>1.5±0.7</td>
<td>1.8±0.7</td>
<td>1.0±0.7</td>
<td>15.80±1.30</td>
</tr>
<tr>
<td>5</td>
<td>Shihmen Primary School Second Campus No. 12</td>
<td>2.1±0.7</td>
<td>2.0±0.7</td>
<td>2.6±0.8</td>
<td>11.47±1.19</td>
</tr>
<tr>
<td>6</td>
<td>Chinshan Park</td>
<td>0.9±0.6</td>
<td>2.1±0.7</td>
<td>2.5±0.8</td>
<td>0.45±0.84</td>
</tr>
</tbody>
</table>

Table 12. Soil Sample Activity without Chemical Treatment prior to Counting (Potassium Contained)

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Soil Sampling Station</th>
<th>Gross Beta Activity (Without Chemical Treatment) pCi/g</th>
<th>January</th>
<th>April</th>
<th>July</th>
<th>October</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Chinshan Transformer Station</td>
<td></td>
<td>19.2±5.6</td>
<td>10.9±3.9</td>
<td>6.0±3.6</td>
<td>18.7±4.4</td>
</tr>
<tr>
<td>2</td>
<td>Southwest Boundary No. 4</td>
<td></td>
<td>21.9±5.7</td>
<td>15.0±4.0</td>
<td>11.3±5.8</td>
<td>23.2±4.4</td>
</tr>
<tr>
<td>3</td>
<td>Chien-Tzu-Lu</td>
<td></td>
<td>26.9±5.9</td>
<td>10.5±3.9</td>
<td>23.8±6.1</td>
<td>31.1±4.6</td>
</tr>
<tr>
<td>4</td>
<td>Shih-Pong-Shan No. 11</td>
<td></td>
<td>32.5±6.0</td>
<td>29.5±4.2</td>
<td>23.8±6.1</td>
<td>20.1±4.4</td>
</tr>
<tr>
<td>5</td>
<td>Shihmen Primary School Second Campus No. 12</td>
<td></td>
<td>24.5±5.8</td>
<td>28.0±4.2</td>
<td>19.9±6.0</td>
<td>35.8±4.6</td>
</tr>
<tr>
<td>6</td>
<td>Chinshan Park</td>
<td></td>
<td>19.8±5.7</td>
<td>9.9±3.9</td>
<td>17.2±5.9</td>
<td>9.8±4.2</td>
</tr>
</tbody>
</table>
Fig. 6. Gamma-ray Spectrum of Soil Samples.

Fig. 7. Gamma-ray Spectrum of Blank Background.
Fig. 8. Gamma—ray Spectrum of Sea Bottom Silt.

Fig. 9. Gamma—ray Spectrum of Sea Water.
Fig. 10. Gamma-ray Spectrum of Aquatic Biota.

Fig. 11. Gamma-ray Spectrum of Fish Ash.
Fig. 12. Gamma-ray Spectrum of Rain Water.

Fig. 13. Gamma-ray Spectrum of Old Tea Ash.
Fig. 14. Gamma-ray Spectrum of New Tea Ash.

Fig. 15. Gamma-ray Spectrum of Drinking Water.
Fig. 16. Gamma—ray Spectrum of Surface water.

Fig. 17. Gamma—ray Spectrum of Ground Water.
Fig. 18. Gamma-ray Spectrum of Rice Ash.

Fig. 19. Gamma-ray Spectrum of Na$_2$HPO$_4$•12H$_2$O Background.

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Fig. 20. Gamma—ray Spectrum of Shen Ao Power Plant Bottom Ash.

Fig. 21. Gamma—ray Spectrum of Shen Ao Power Plant Fly Ash.
9. Fish

The gross beta analysis and a gamma scan were made. An annual $^{90}$Sr analysis was also performed. The results are summarized in Table 13.

10. Chicken and Duck

The gross beta analysis of chicken, ducks, and their eggs were performed and the results are listed in Table 14.

II. Milk

Milk samples taken from the dairy farm at Shanchi indicate the background activity only as shown in Table 15.
Table 13. Gross Activity and \(^{89}\)Sr Concentration in Fish (Chienhua Area)

<table>
<thead>
<tr>
<th>Fish Specimen</th>
<th>Sampling Date</th>
<th>Calcium Content(\text{mg/g (ash)})</th>
<th>Gross Beta(\text{pCi/g (ash) pCi/kg (wet)})</th>
<th>(^{89})Sr(\text{pCi/g (ash) pCi/kg (wet) pCi/g Ca pCi/g (ash)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wu Tse</td>
<td>8/1</td>
<td>0</td>
<td>50.1±6.5 1252±164</td>
<td>0.57±0.33 14.3±8.1</td>
</tr>
<tr>
<td>Chin Hsien Yu</td>
<td>8/1</td>
<td>242</td>
<td>47.0±6.5 2386±328</td>
<td>0.80±0.32 40.6±16.3</td>
</tr>
<tr>
<td>Pao Tai Yu</td>
<td>8/1</td>
<td>172</td>
<td>104.7±7.8 2010±150</td>
<td>0.66±0.24 12.8±4.6</td>
</tr>
<tr>
<td>Pai Tu Yu</td>
<td>8/1</td>
<td>206</td>
<td>54.3±6.7 2666±327</td>
<td>0.27±0.24 13.1±11.8</td>
</tr>
<tr>
<td>Heh Kou Yu</td>
<td>11/28</td>
<td>207</td>
<td>53.0±8.0 1802±274</td>
<td>0.46±0.40 15.7±13.7</td>
</tr>
<tr>
<td>Hung Shih Pan Yu</td>
<td>11/28</td>
<td>301</td>
<td>13.0±7.0 806±435</td>
<td>0.53±0.36 32.2±21.9</td>
</tr>
</tbody>
</table>

B = background
### Table 14. The Activity of Chicken and Duck Samples Taken Around the 600 MW(e) Reactor Site

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sampling Station</th>
<th>Sampling Date</th>
<th>Gross Beta Activity</th>
<th>²⁰Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>pCi/g (ash) pCi/g (ash)</td>
<td>pCi/kg (ash)</td>
</tr>
<tr>
<td>Chicken</td>
<td>Chienlu Village 9 Lin</td>
<td>5/7</td>
<td>55.0±6.6 1647±198</td>
<td></td>
</tr>
<tr>
<td>Duck</td>
<td>Chiu-Chiung-Lin</td>
<td>5/7</td>
<td>44.1±6.3 1994±286</td>
<td>47.0±6.4 1606±219</td>
</tr>
<tr>
<td>Duck Egg</td>
<td>Chienlu Village 9 Lin</td>
<td>8/1</td>
<td>49.0±6.5 2007±267</td>
<td>51.0±6.5 2055±262</td>
</tr>
<tr>
<td></td>
<td>Chiu-Chiung-Lin</td>
<td>11/6</td>
<td>89.0±9.0 951±93</td>
<td></td>
</tr>
</tbody>
</table>

### Table 15. The Activity in Fresh Milk Samples Taken from Shanchih and Hsinchu Dairy Farms.

<table>
<thead>
<tr>
<th>Sampling Period (month)</th>
<th>Sampling Location</th>
<th>²⁰Sr</th>
<th>¹³⁷Cs</th>
<th>¹³¹I</th>
<th>¹³¹Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>pCi/liter</td>
<td>pCi/g Ca</td>
<td>Ca%</td>
<td>pCi/liter</td>
</tr>
<tr>
<td>1-12</td>
<td>Hsinchu</td>
<td>6.4±2.6</td>
<td>4.6±1.9</td>
<td>0.12</td>
<td>11.6±3.3</td>
</tr>
<tr>
<td>7</td>
<td>Shanchih</td>
<td>3.8±2.3</td>
<td>2.4±1.5</td>
<td>0.13</td>
<td>14.6±2.2</td>
</tr>
<tr>
<td>8</td>
<td>Shanchih</td>
<td>1.9±1.4</td>
<td>1.1±0.8</td>
<td>0.15</td>
<td>18.5±4.9</td>
</tr>
<tr>
<td>9</td>
<td>Shanchih</td>
<td>4.1±3.7</td>
<td>4.0±3.6</td>
<td>0.14</td>
<td>40.2±2.3</td>
</tr>
<tr>
<td>10</td>
<td>Shanchih</td>
<td>1.6±1.1</td>
<td>1.2±0.9</td>
<td>0.07</td>
<td>14.2±2.9</td>
</tr>
<tr>
<td>11</td>
<td>Shanchih</td>
<td>7.5±2.7</td>
<td>5.8±2.8</td>
<td>0.11</td>
<td>19.9±3.0</td>
</tr>
<tr>
<td>12</td>
<td>Shanchih</td>
<td>4.4±1.8</td>
<td>2.4±0.9</td>
<td>0.16</td>
<td>12.4±2.1</td>
</tr>
</tbody>
</table>

Remark: 1 liter of fresh milk = 1.2 kg.

### ACKNOWLEDGMENTS

The information contained in this report was developed during the course of work under the Contract 62 HP2 with the Taiwan Power Company. The gamma scan was performed at the Institute of Nuclear Energy Research and assisted by Mrs. Li-Yu Kao Chung of Taiwan Power Company. The routine sample analysis was assisted by Mr. Chao-Chung Liu and Mr. Tsung-Chun Chang.
Since April 1961, the $^{137}$Cs and potassium content of the Chicago portion of Tri-City Diet Sampling Program has been determined$^{(1-5)}$ in bulk food samples by gamma-ray spectrometry using a 4" x 4" NaI (TI) crystal. Each variety of food (all fresh vegetables, all fresh fruits, etc.) is composited before measurement, and each sample is counted 400–1000 minutes. From these measurements composite daily and yearly food intakes are obtained. The results for the April 1976 semi-annual sampling are tabulated in Tables I and II.

*Work performed under the auspices of the U. S. Energy Research and Development Administration.
TABLE I

Cesium-137 in Chicago Diets

April 1976

<table>
<thead>
<tr>
<th></th>
<th>kg/yr</th>
<th>Potassium g/kg</th>
<th>137Cs pCi/kg</th>
<th>Potassium g/yr</th>
<th>137Cs pCi/yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>White bread</td>
<td>37</td>
<td>1.1</td>
<td>8.2</td>
<td>41</td>
<td>303</td>
</tr>
<tr>
<td>Whole wheat bread</td>
<td>11</td>
<td>2.0</td>
<td>9.0</td>
<td>22</td>
<td>99</td>
</tr>
<tr>
<td>Eggs</td>
<td>16</td>
<td>1.4</td>
<td>&lt;4*</td>
<td>22</td>
<td>32</td>
</tr>
<tr>
<td>Fresh vegetables</td>
<td>43</td>
<td>2.6</td>
<td>6.3</td>
<td>112</td>
<td>271</td>
</tr>
<tr>
<td>Root vegetables</td>
<td>17</td>
<td>2.5</td>
<td>4.3</td>
<td>42</td>
<td>73</td>
</tr>
<tr>
<td>Milk</td>
<td>221</td>
<td>1.3</td>
<td>4.1</td>
<td>287</td>
<td>906</td>
</tr>
<tr>
<td>Poultry</td>
<td>17</td>
<td>3.0</td>
<td>4.3</td>
<td>51</td>
<td>73</td>
</tr>
<tr>
<td>Fresh fish**</td>
<td>8</td>
<td>3.8</td>
<td>20</td>
<td>30</td>
<td>160</td>
</tr>
<tr>
<td>Flour</td>
<td>43</td>
<td>1.3</td>
<td>&lt;4</td>
<td>56</td>
<td>86</td>
</tr>
<tr>
<td>Macaroni</td>
<td>3</td>
<td>1.5</td>
<td>&lt;4</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>Meat</td>
<td>73</td>
<td>3.3</td>
<td>16</td>
<td>241</td>
<td>1168</td>
</tr>
<tr>
<td>Dried beans</td>
<td>3</td>
<td>14</td>
<td>17</td>
<td>42</td>
<td>51</td>
</tr>
<tr>
<td>Fresh fruit</td>
<td>68</td>
<td>2.3</td>
<td>&lt;4</td>
<td>156</td>
<td>136</td>
</tr>
<tr>
<td>Potatoes</td>
<td>45</td>
<td>5.1</td>
<td>15</td>
<td>230</td>
<td>675</td>
</tr>
<tr>
<td>Canned fruits</td>
<td>26</td>
<td>0.8</td>
<td>&lt;4</td>
<td>21</td>
<td>62</td>
</tr>
<tr>
<td>Canned fruit juices</td>
<td>19</td>
<td>1.7</td>
<td>4.0</td>
<td>32</td>
<td>76</td>
</tr>
<tr>
<td>Canned vegetables</td>
<td>20</td>
<td>1.2</td>
<td>&lt;4</td>
<td>24</td>
<td>40</td>
</tr>
</tbody>
</table>

Total/yr       | 1414  | 4207           |
Total/day      | 3.9   | 12             |

* Samples containing <4 pCi/kg (approximately our limit of detection for this type of sample) are assumed to have 2 pCi/kg.

** It is assumed in arriving at the average that nine times more ocean fish is consumed than freshwater fish.
### TABLE II
Cesium-137 in Chicago Diets (Infants)
April 1976

<table>
<thead>
<tr>
<th></th>
<th>kg/yr</th>
<th>Potassium g/kg</th>
<th>$^{137}Cs$ pCi/kg</th>
<th>Potassium g/yr</th>
<th>$^{137}Cs$ pCi/yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Evaporated milk</td>
<td>137</td>
<td>3.2</td>
<td>11</td>
<td>438</td>
<td>1507</td>
</tr>
<tr>
<td>Formula milk</td>
<td>37</td>
<td>1.5</td>
<td>&lt;4*</td>
<td>56</td>
<td>74</td>
</tr>
<tr>
<td>Cereals</td>
<td>8</td>
<td>10</td>
<td>8.9</td>
<td>80</td>
<td>71</td>
</tr>
<tr>
<td>Fruits</td>
<td>23</td>
<td>1.0</td>
<td>&lt;4</td>
<td>23</td>
<td>46</td>
</tr>
<tr>
<td>Meats</td>
<td>17</td>
<td>1.8</td>
<td>12</td>
<td>31</td>
<td>204</td>
</tr>
<tr>
<td>Vegetables</td>
<td>23</td>
<td>1.3</td>
<td>&lt;4</td>
<td>30</td>
<td>46</td>
</tr>
<tr>
<td><strong>Total/yr</strong></td>
<td></td>
<td></td>
<td></td>
<td><strong>658</strong></td>
<td><strong>1948</strong></td>
</tr>
<tr>
<td><strong>Total/day</strong></td>
<td></td>
<td></td>
<td></td>
<td><strong>1.8</strong></td>
<td><strong>5.3</strong></td>
</tr>
</tbody>
</table>

* Samples containing <4 pCi/kg (approximately our limit for detection for this type of sample) are assumed to have 2 pCi/kg.
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