## Radioactive Fallout from Nuclear Weapons Tests

The sources and characteristics of fallout material, the transport of such material through the atmosphere, and radioactivity in soil, food, and man have been extensively studied under the auspices of the U.S. Atomic Energy Commission. The second AEC conference on radioactive fallout was held 3–6 November 1964 in Germantown, Maryland, for the purpose of presenting and discussing results which have been obtained since the first conference in November 1961.

Freiling, Crocker, and Adams (U.S. Naval Radiological Defense Laboratory) described a program of study of nuclear debris formation in which two prediction systems programmed for computer calculations are being used. Along with the predictions, radiological properties of nuclear debris are ascertained. Preliminary laboratory investigations of interactions of fission products with various substrate materials were described. These investigations have shown large negative deviations from Raoult's law. In another paper Crocker, Kawahara, and Freiling described correlations of radiochemical data from samples collected during nuclear tests. Differences in fractionation of nuclides in various types of detonations appear to be a matter of degree. Air bursts, coral surface bursts, and silicate bursts seem to be similar in the kind of fractionation which occurs, except for the fractionation of  $Mo^{99}$  with respect to  $Zr^{95}$  in air bursts and of Np<sup>239</sup> with respect to Zr95 in coral surface bursts. Fractionation appears to increase as particle size increases. Miller and Sartor (Stanford Research Institute) described the coordinated program of study of fallout following the surface detonation of Small Boy in Nevada in 1962 and discussed the results in relation to fallout prediction models. The field program attempted to obtain data needed for

# Meetings

describing and defining a large number of parameters. Krey and Friend (Isotopes, Inc.) described a program of study at greater distances from the Nevada Test Site after this detonation and after the nuclear explosives excavation test Sedan. Data obtained on arcs at 1010, 1600, and 2240 km from Ground Zero consisted of gross yray assays, y-ray decay measurements,  $\gamma$ -spectrum analyses, and radiochemical analyses of samples. The data showed fractionation effects for both events. Heft and Kahn (University of California, Lawrence Radiation Laboratory, Livermore) described a program of study of particles collected from their point of origin during past tests, using as an example the Sedan event.

Benson, Gleit, and Leventhal (Tracerlab) presented results of studies of physical and radiochemical fractionation characteristics of single particles from early nuclear cloud samples obtained after tests in the Pacific in 1962. Beta activity 25 days after three events was found to be directly proportional to particle volume and to be a function of nuclear yield. The negative time exponent for activity decay of individual particles ranged from 0.7 to 1.4 and was independent of particle size. The individual particles were extremely fractionated. Lockhart, Patterson, and Saunders (U.S. Naval Research Laboratory) found no obvious relation between fractionation and particle size in surface air samples at Washington, D.C., during 1963-1964. They concluded that nuclear debris from stratospheric sources becomes associated with relatively large nonradioactive particles during transit in the troposphere. Resulting particles had an "average" diameter of 0.3 to 1.0  $\mu$  and a rather restricted distribution with respect to size.

Foote (Texas Instruments, Inc.) has developed a method for the study of  $\gamma$ -ray spectra in the field. He showed the effects of rainfall and snow cover on  $\gamma$ -spectra and discussed other factors influencing the levels of nuclides measured at Dallas, Texas. Lowder, Beck, and Condon (AEC Health and Safety Laboratory) described their system of field measurements of  $\gamma$ -emitting nuclides and results obtained at more than 200 locations in the U.S. during the last several years.

Perkins, Thomas, and Nielsen (Hanford Laboratories, General Electric Co.) described a recently developed multidimensional  $\gamma$ -ray spectrometer for direct analysis of many nuclides produced by fallout and cosmic-rays: Be<sup>7</sup>, Na<sup>22</sup>, Mn<sup>54</sup>, Co<sup>66</sup>, Y<sup>88</sup>, Zr<sup>95</sup>-Nb<sup>95</sup>, Ru<sup>106</sup>, Sb<sup>124</sup>, Sb<sup>125</sup>, Cs<sup>134</sup>, Cs137, and Ce144. This relatively inexpensive method is potentially quite valuable for fallout studies because it permits analyses which formerly required lengthy radiochemistry. Haines and Musgrave (University of Arkansas) discussed their studies of the ratios of tritium to hydrogen in the atmospheric hydrogen and of the chemical forms in which atmospheric tritium is found. They also discussed the variations (and their causes) in T/H ratios and the sources of tritiated methane.

Results of several fundamental investigations of the long-term behavior of particles were reported. Friedlander (California Institute of Technology) and Pasceri (Allegheny Ballistics Laboratory) described a simple method for measuring size distribution of particles less than a few tenths of a micron in diameter by diffusion from room air to a rotating disk. Measurements made by this method and the theory involved were discussed. In another paper based on earlier theoretical and experimental considerations of Friedlander and Swift, Friedlander presented his theory of self-preserving size distributions for the case of Brownian diffusion. From these considerations he offered an explanation for regularities in the size distribution of the atmospheric aerosol. Liu and Whitby (University of Minnesota) discussed their studies of electric charging of solid methylene blue particles ranging from 0.02 to 2.0  $\mu$  in diameter at pressures ranging from 0.01 to 1.0 atmospheres. They found the measured particle charge was generally higher than that predicted and that this difference increased with decreasing pressure. Owe Berg, Gaukler, and Squier (Aerojet-General Corp.) presented results of studies of wetting of solids by water under conditions simulating those of washout of nuclear debris by water spray or rain. They found that wetting is a formation of hydrogen bonds between  $H_2O$  in the water drop and OH on the surface of the solid, a process requiring reorientation of bonds in the drop.

Several of the papers presented included other data on particle characteristics as well as on mechanisms of transport of particles in the atmosphere.

#### Atmospheric Transport of Fallout Material

The sessions of the conference devoted to discussions of the transport of nuclear debris from the source to various receptors were characterized by a feeling that, though a great deal of work has been done in this area, much more is necessary before any definitive statements can be made concerning (i) the exact distribution of global fallout, (ii) how and why that distribution changes in time and space, or (iii) the preparation of quantitative forecasts of fallout under varied meteorological conditions.

This need for further research was emphasized by Machta (U.S. Weather Bureau), who reported on the status of global fallout predictions. For tropospheric sources, the details of the individual trajectories and precipitation scavenging play a major role, while the statistical behavior of the stratosphere controls the long-term transport and dilution of material at higher levels. There is a trend toward the use of computers for prediction of fallout, but computer results give only the mean field and not the local "hot spots."

Newell and Miller (Massachusetts Institute of Technology) discussed the complications of stratospheric circulation. They concluded that the lower stratosphere is driven by the troposphere and is also coupled with the upper stratosphere. Thus there is no simple circulation in the stratosphere but rather a complicated one which is interrelated with the layers above and below it.

Several papers dealt with observations made in the stratosphere. Loysen (HASL) and Drevinsky and Pecci (U.S. Air Force Cambridge Research Laboratories) presented material on the size and distribution of particle sizes in the stratosphere. They determined that most of the activity is carried by particles less than 0.2  $\mu$  in radius. Feely and Bazan (Isotopes, Inc.) reported that the vertical distribution of Sr<sup>90</sup> observed in the stratosphere showed little stability. In addition, during 1963 large quantities of debris deposited in the stratosphere during 1961 and 1962 reached the southern polar stratosphere; this was explained by seasonal changes in the coefficient of turbulent exchange in the tropical stratosphere.

Tracer studies of specific radionuclides in the stratosphere were discussed by other researchers. Kalkstein, Thomasian, and Nikula (U.S. Air Force Cambridge Research Laboratories) showed that the concentration of Cd<sup>109</sup> from the Starfish test of 1962 increased at high latitudes during the Southern Hemisphere spring of 1963. These increases in concentration were coincident with warmings of the polar stratosphere in the Southern Hemisphere. Increased concentrations occurred in the Northern Hemisphere late in the winter of 1963-1964 but did not reach the levels observed in the Southern Hemisphere. Salter (HASL) discussed the origin and concentrations of nuclides found in stratospheric air in the Southern Hemisphere from 1961 and 1962 nuclear tests. He reported that Cd109 from the Starfish test of 1962 first appeared during December 1962 at 34°S in samples collected at 3200 m. Gudiksen, Jones, Schell, Swanson, Erickson, and Fairhall (University of Washington) observed a maximum in the ratio of Mn<sup>54</sup> to Cs<sup>137</sup> during April and May 1963 as a result of nonuniform mixing of the stratosphere with respect to Mn<sup>54</sup>. They also observed a direct correlation between the stratospheric and tropospheric activity levels near the tropopause. The observations also showed a rapid diffusion and no fractionation across the tropopause. Kuroda, Kauranen, Palmer, Menon, and Fry (University of Arkansas) reported that their analyses of Sb124 strongly indicates that the horizontal transport of material in the stratosphere follows the pattern outlined by Newell in which the eddy motions, rather than mean motions, are the main transport process up to 25 km. The radiochemical techniques developed by Kuroda et al. have helped to define better some of the debris sources. In 1963 concentrations of C<sup>14</sup> in the troposphere increased during summer, while lower levels were found during fall and winter. The lower values are attributed to an exchange of atmospheric CO<sub>2</sub> with that dissolved in the oceans. Those facts were concluded from measurements made over the Pacific Northwest and reported by Young, Erickson, and Fairhall (University of Washington).

One area of research in which a great deal of progress has been made is in the study of the transport of material from the stratosphere to the troposphere. Danielsen (Pennsylvania State University) discussed the results of observations made from airplanes during the spring of 1963 which substantiated the concept of a folding tropopause. As the tropopause folds, stratospheric air and radioactive debris are extruded into the troposphere. This extruded air is characterized by a high potential vorticity and, as a result, areas of high radioactivity can be located by analyses of the wind and temperature fields. Reiter and Mahlman (Colorado State University) described a case study near the jet stream where 6  $\times$ 10<sup>11</sup> metric tons of air sank from the tropopause to the surface in a period of about two days. The subsiding stratospheric air was responsible for fallout in excess of 320 pc/m<sup>3</sup> as observed over the south central U.S. 24-27 November 1962. Mahlman tried to relate the spring fallout maximum to an increase in cyclonic activity, but concluded that other processes must be involved. However, he was able to relate fallout peaks over short periods to the occurrence of cyclogenesis in the upper troposphere.

A large effort has been made to determine the role of rainshowers and thundershowers in washing out the troposphere. Huff (Illinois State Water Survey) showed that the variation in radioactive rainout from place to place is very large. A single measurement may have an average error of 20 to 25 percent if it is assumed to represent the mean rainout over an area of 25 to 30 km<sup>2</sup>. Rainfall at a given point is not correlated highly with radioactive rainout at that point; however, the average of data from a number of points over an area indicates a strong association between rainfall and rainout. Hall (University of Oklahoma) presented case studies of convective activity in Oklahoma which showed the temporal and areal distribution of radioactive debris. A minimum of  $\beta$ activity was found to be associated with maximum rainfall rates for mature storms, whereas in dissipating storms the maximum  $\beta$  activity occurred with the maximum rainfall. Additional case studies were contributed by Gatz and Dingle (University of Michigan). From their cases it was inferred that with heavy rainfall in convective storms, both radioactivity

SCIENCE, VOL. 147

and pollen entered the storm system at low levels via convective updrafts. Because of a lack of persistent organized updrafts, light rain showed no rapid decrease of radioactivity. Kruger, Hosler, and Miller (Hazleton-Nuclear Science Corp.) reported on other rainfall case studies in California, Oklahoma, Pennsylvania, and Hawaii.

Surface measurements of radioactivity were reported by several groups. Lockhart, Patterson, Saunders, and Black (U.S. Naval Research Laboratory) gave a summary of the measurements of gross  $\beta$  and Sr<sup>90</sup> activity measured in air at ground level along the 80th meridian (W) during 1957-1962. The data collected during this program have helped to substantiate a number of concepts regarding atmospheric mixing processes and the residence times of radioactive particulate matter in the atmosphere. Volchok (HASL) brought measurements from the 80th meridian up to date by discussing the recent observations and changes instituted by HASL. The ratios of Sr<sup>89</sup> to Sr<sup>90</sup> indicated that the nuclear debris in the Northern Hemisphere was in general well mixed in comparison with the older debris in the Southern Hemisphere. Volchok showed that a symmetry between the hemispheres does exist if one assumes a 4-month offset in time. Other groundlevel measurements were presented by Gustafson, Brar, and Muniak (Argonne National Laboratory). They found a variation in the concentrations of various nuclides, especially Mn54 and Cs137 and their ratios. These ratios are indicative of the source of the debris.

The observed changes in the time of the spring maximum during 1961-1964 were attributed to changes in general atmospheric circulation patterns. They concluded that there is a large stratospheric source which may lead to a substantial spring maximum for the next several years. Roser and Cullen (Universidade Catolica, Rio de Janeiro) reported on their observations in Brazil. The fallout at the tropical stations has a strong correlation with precipitation, whereas data from the stations further south show the typically peaked yearly distribution which results from a spring maximum. The most interesting fact reported was that the coastal and oceanic stations show significantly higher levels of fallout than do the inland stations.

Several reports on tritium were pre-26 FEBRUARY 1965 sented. Bolin and Erikkson (University of Stockholm) discussed the occurrence of the nuclides deuterium, tritium, and O<sup>18</sup> in natural waters and their relations to the general circulation of water. Because few measurements of concentrations of these nuclides are available, the observed patterns had to be explained on the basis of standard meteorological measurements of humidity, precipitation, and evaporation. Thatcher, Cameron, and Payne (International Atomic Energy Agency) presented data showing the variability of tritium concentrations in precipitation. They noted that the deposition values for tritium and Sr<sup>90</sup> have consistencies which may permit estimation of tritium concentration in unsampled areas. Precipitation in the Southern Hemisphere averaged 15 tritium units in 1963 as compared to thousands of tritium units for the Northern Hemisphere. Of the 168 kg of tritium produced by nuclear tests in 1963, 43 to 88 kg had been deposited by the end of the year, the amount depending on whether oceanic exchange was included in the measurement.

List (U.S. Weather Bureau) *et al.* reported on the results of the worldwide soil sampling program, which indicate that about 7.6 Mc of  $Sr^{90}$  had been deposited on the earth's surface by the end of 1963. The amount deposited increases as latitude increases and correlates positively with amount of precipitation.

The sampling of close-in fallout and, especially, the sampling of nuclear clouds by aircraft for global fallout material were reported by Ferber (U.S. Weather Bureau). He found that less than 1 percent of the total radioactivity in a nuclear cloud is present in the stem and less than 0.1 percent of the material stabilizes in the lower half of the cloud. About one third of the total debris from Dominic I initially stabilized in the troposphere. The techniques used to obtain the samples can also be used to get good inventory data.

A special case of local fallout was discussed by Knox (University of California, Lawrence Radiation Laboratory, Livermore). He presented a numerical simulation model which was developed for the prediction of fallout from nuclear detonations that produce craters. Calculations of the fallout show that the model gives good estimates of the  $\gamma$ -dose rate at 1 hour after detonation. A maximum error of a factor of

2 to 3 occurs in the  $\gamma$ -dose rate versus distance along the hot line of the pattern. Some of the papers mentioned earlier, as well as the following section, are related to field studies of such a detonation.

### Radionuclides in Soil, Food, and Man

Several groups undertaking field studies of the distribution and cycling of radionuclides in the environment with respect to the food chain and man presented results pertaining to several parameters involved. Ward, Johnson, and Stewart (Colorado State University) discussed coefficients for transfer of Cs137 from precipitation to forage plants and from forage plants and other feeds to milk. They found that, of the total amount of Cs137 which had fallen in precipitation on an area of 1 m<sup>2</sup>, the percentage detected in forage from that area varied from about 80 percent for pasture plants collected after a rain to about 10 percent for alfalfa hay cut late in summer. In cows which were fed rations varying in content of crude fiber, hay, or pasture plants, they found that the Cs137 secreted per liter of milk was related to crude fiber content of the rationfor example, secretion into milk could be doubled if the ration were low in crude fiber. Hawthorne (University of California, Los Angeles) discussed his studies of radionuclide movement on a farm at St. George, Utah, specifically pointing out sample heterogeneity which to some extent has been found by the Colorado State group. Pelletier, Whipple, and Wedlick (University of Michigan) described a portion of an empirical model of radionuclides in milk and the various parameters investigated during a field program. Air and precipitation concentrations, or deposition, of several nuclides were presented, and relationships found were used to predict deposition.

Martin (University of California, Los Angeles) discussed results of studies of  $Sr^{89}$  and  $I^{131}$  in native animals and plants in the close-in fallout field from the *Sedan* event. Deterministic, exponential models formulated from the data obtained were satisfactory in explaining relationships between initial depositions of the nuclides on plants and the concentrations found subsequently in bone and thyroid of rabbits. A stochastic model for such relationships was discussed by Turner (University of California, Los Angeles). Using concentrations of  $I^{131}$  found in thyroids of herbivores after the Sedan event, together with data from the literature, he showed that frequency distributions approximate log normal distributions in the cases chosen. He discussed the implications for human health of the possibility that such skewed distributions are more common than normal ones.

Hawley and Markee (Idaho Operations Office, AEC) discussed their field program for studies of the effect on the food chain of the deposition of radioiodine through controlled releases of radioiodine to the atmosphere. In two release experiments deposition velocities on grass were similar when account was taken of grass density. The half-time for radioiodine on grass was found to be 4 to 6 days, which agrees with findings of others. Peak activity in milk in the two tests was found after about two days, with large differences among individual cows. The concentration of radioiodine in milk was found to decrease with a half-time of about one day. From human ingestion of milk obtained in one test, a thyroid uptake of 19 percent was found.

Tamplin and Minkler (Lawrence Radiation Laboratory) described their information-integration system for collection, collation, and analysis of literature on biospheric transport of radionuclides. They mentioned the necessity, in view of the increasing amount of literature, for such a system, manned by scientific personnel, for efficient use of technical information. Perhaps the preceding papers point out the need for such a system, as does the paper of Thompson and Lengemann (Cornell University) on dietary intake of radionuclides. These investigators have attempted to evaluate concentrations of fallout nuclides in the diet of populations, using existing data from surveys of consumption and radionuclides. They discussed the possibility of using the concentration of Sr<sup>90</sup> in urine as an indicator of concentration in the diet, a method suggested by laboratory experiments with human subjects. They found that the average ratio of Sr<sup>90</sup> to Ca in the urine was about the same as that in the food eaten. Ratios for a single case of controlled ingestion of food, collected on Rongelap which received heavy fallout during tests in 1954, were reported by Hardy and Rivera (HASL) and Conard (Brookhaven National Laboratory). Whereas the urine was the principal route of removal of Cs137, Sr90 was removed mainly in the feces. Whole body counting showed reasonably good agreement with excretion data for retention of Cs137. Relations between concentrations of Sr<sup>90</sup> and stable strontium in the diet to concentration in bone in children were reported by Beninson, Ramos, and Touzet (Argentine Comision Nacional de Energia Atomica), who developed a preliminary model for estimating factors which determine the amount of strontium which will be retained by children of various ages. Dietary intake and retention of Sr<sup>90</sup> were discussed by Rivera in terms of prediction and evaluation of population doses in fallout situations.

The relationships among measured levels of Cs137 in various samples of interest in the Chicago area were discussed by Gustafson, Brar, and Muniak (Argonne National Laboratory). From these studies, along with the field experiments mentioned earlier and the studies in Norway, a considerably better basis for prediction and evaluation of amounts and distribution of Cs137 in fallout is evident. Strømme (Norwegian Radium Hospital) and Madsus (Norsk Hydros Institute) reported that a significant relationship was found between the concentrations of Cs137 in milk during one year and the amount of precipitation during the preceding year along the west coast of Norway but that no such relationship was found in eastern Norway. As an extension of these studies, Cs137 and potassium body burdens of 22 schoolboys were measured during 1963 and 1964.

The proceedings of the conference will be published as report TID-7701 in the AEC symposium series and will be available in a few months from the Office of Technical Services, U.S. Department of Commerce.

EUGENE W. BIERLY ALFRED W. KLEMENT, JR. Fallout Studies Branch, U.S. Atomic Energy Commission, Washington, D.C.

#### Paleomagnetism

Paleomagnetism was the subject of the second conference of a group of American and Japanese scientists meeting under the aegis of the U.S.-Japan Cooperative Science Program in Berkeley, California, 12–13 November 1964. Two years of work since the Tokyo meeting were reviewed.

Japanese scientists working in Japan and the United States reported on the magnetic properties of ferrimagnetic oxides. Akimoto (Tokyo) reported on the system  $Fe_2TiO_4$ - $FE_3O_4$ ; neutron diffraction clearly shows that titanium remains exclusively in six-coordinated positions at all compositions. Anisotropy and magnetostriction constants at low temperature show remarkable variations with temperature and composition, and generally increase with titanium content. Akimoto thinks that the effect is mostly due to  $Fe^{2++}$ ; the higher stability and natural remanence of basaltic rock, as compared with granites in which the spinel phase generally contains very little titanium, may be explained in this way.

Ozima (Tokyo) reported on the memory effects associated with the magnetic transition at about 120°K where the signs of the anisotropy coefficients change. He showed that there is very little memory of isothermal remanent megnetization (IRM). However thermal remanent magnetization (TRM) and particularly the partial TRM acquired within 100 degrees of the Curie point, has an extraordinary recovery ratio after cooling below the transition point. This clearly shows that IRM and TRM are acquired by different mechanisms, and suggests, as was proposed by Verhoogen, that TRM depends mainly on magnetostrictive effects. Ozima suggested that the loss of IRM by cooling below 120°K may be used as a simple and practical method for "magnetic cleaning" of rock specimens. Kobayashi and co-workers at Pittsburgh made similar experiments on the magnetite transition and the Morin transition in  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>; both transitions are completely suppressed in material with an average grain size of 0.1  $\mu$ ; and in magnetite the memories of saturated remanence acquired below the transition point and at room temperature in the same direction are reciprocal in the sense that they destroy each other. Kobayashi also found that in hematite the reversible susceptibility decreases when the IRM carried by the specimen increases. However, the susceptibility is independent of the magnitude of TRM and anhysteretic remanent magnetization until saturation is reached, showing that TRM is independent of domain configuration. Nagata (Tokyo) reported on the effect of uniaxial pressure on the acquisition of remanence; Kume and Koizumi (Osaka) had studied the acquisition of remanence during the  $\alpha \rightarrow \gamma$  and  $\gamma - \alpha$  transitions in Fe<sub>2</sub>O<sub>3</sub>.

Investigations of the direction and intensity of the earth's field in the last