INVESTIGATING FALLOUT FROM NUCLEAR TESTING

HOT PARTICLES & THE COLD WAR

BY PIER ROBERTO DANESI

Entering the next century, more than 2000 nuclear test explosions of various sizes and varieties will have been recorded. Nearly all of them were conducted during the Cold War period ending in the 1990s.

Atmospheric nuclear tests dispersed radioactive residues into the environment. They are partitioned between the local ground (or water surface) and the tropospheric and stratospheric regions, depending on the type of test, location, and yield. The subsequent precipitation carrying the residues leads to both local and global fallout. Concentrations of certain radionuclides can result in formation of "hot particles" — tiny bits of materials containing radioactive chemical elements.

Local fallout includes large radioactive aerosols, particles which are generally deposited within about 100 kilometers of the test site. Local radioactive contamination at nuclear-weapon test sites additionally is attributed to safety trials of nuclear devices that often dispersed fissile material. This material is released in various forms, including plutonium vapour, plutonium aerosols of various sizes, plutonium oxide particulates, plutonium-coated particles, and sizeable lumps of plutonium-contaminated structural material destroyed by the test explosion.

Global fallout encompasses both tropospheric and stratospheric fallout. The first consists of aerosols that are not carried across the tropopause and that deposit with a mean residence time of up to 30 days. During this time, the residues become dispersed in the latitude band of injection, following trajectories governed by wind patterns. Stratospheric fallout arises from particles that later give rise to widespread global fallout, most of which is in the hemisphere where the nuclear test was conducted. It accounts for most of the residues of long-lived fission products.

Nuclear fallout results in the exposure of people to radioactivity through internal irradiation (due to inhalation of radioactive materials in air or ingestion of contaminated food) and external irradiation (by radioactive materials present in surface air or deposited on the ground). Extensive studies in these areas have been done.

In the case of nuclear testing, evaluations of the nature of the primary event typically include analyses of radioactive material deposited on the ground. Such studies are problematical, however, because there can be significant alteration of radionuclide composition between the time of a nuclear detonation and the time samples are collected for radiochemical analysis. A process called fractionation causes samples of radioactive residues to be unrepresentative of the detonation products.

Fractionation begins with the condensation of radioactive and inert material from the fireball. The mixture may begin to separate while condensation is still in progress because of the influence of wind, gravity and the turbulence of the atomic cloud. Further separation of the condensate occurs through various processes, including contact of the residues with inert material.

When the atomic cloud is formed, the processes of

Photos: A marker at the former nuclear test site in Maralinga, southern Australia. At the Mururoa test site in French Polynesia, an IAEA team collected soil samples for analysis of hot particles. *(Credit: Danesi/IAEA)*

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cooling, condensation, coagulation, mixing, and separation take place simultaneously but to different extents in different regions of the cloud. Furthermore, the initial radioactive products change in elemental form through processes of radioactive decay.

Hot particles. Scientific understanding of the fractionation phenomena is important for interpreting global fallout and the nuclear chemistry of the detonation process. It is also useful for evaluating contamination and ingestion hazards.

Variations in size and type of particles are accompanied by variations in radiochemical composition, according to fractionation patterns. The nature and concentrations of radionuclides, together with the size and shape of particles, are in turn the factors determining the inhalation or ingestion hazard.

Other important scientific information concerns the chemical form in which radionuclides are present in the particles. The radionuclides present in hot particles are in general relatively inert compared to ions, atoms and low molecular mass species that are more mobile and easily available.

To assess short- and long-term consequences of atmospheric fallout — and in particular the leakage of radionuclides from hot particles — detailed physico-chemical information on this source term is essential. Usually, characteristics of source terms have been restricted to inventory estimates, activity levels, or activity concentrations of deposited radionuclides. Information on the physico-chemical form is limited.

After deposition, particles are subject to weathering and the associated radionuclides are mobilised over time. The particle composition, possible structural changes, and the chemical conditions at the deposition site will influence the weathering rate. Moreover, the mobilised radionuclides can also interact with soil and sediments.

Most models assessing the transfer and consequences of radionuclide contamination assume that the radionuclides are present as ionic or low molecular weight species. This may easily lead to an overestimation of the short-term consequences of the radiological contamination.

On the other hand, if the particles are rather inert, as often the case, the transfer of radionuclides will be delayed until weathering occurs. Consequently, the assessment of the long-term consequences of radiological contamination can be underestimated.

It is then apparent that unless the role of hot particles is taken into account, model predictions can be affected by significant uncertainties.

The presence of hot particles may also render invalid some assumptions made when dealing with soil and sediment contamination. This includes the frequent assumption that the scaling of surface or mass activity concentrations to units of various magnitudes (for example, from Bq/cm^2 to Bq/m^2 or Bq/km^2 and vice versa) is a legitimate operation.

The following examples illustrate specific approaches for studying hot particles at nuclear-weapon test sites.

STUDIES IN SOUTH AUSTRALIA

From 1953-63, the United Kingdom conducted a programme of nuclear-weapon testing at Maralinga and Emu in South Australia. At these sites, now completely rehabilitated, nine nuclear explosions and several hundred smaller-scale trials were performed.

While the major explosions had a yield between one and 27 kilotons, the minor trials only involved the burning and explosive dispersal of uranium, plutonium, and short-lived radionuclides. The environmental consequences of these trials were extensively studied by the Australian Radiation Laboratory (ARL) and have been the subject of a number of publications.

The most significant plutonium contamination at Maralinga resulted from a series of twelve safety trials in which 22 kg of plutonium (and a similar quantity of uranium-235) were released into the environment. The material was dispersed by conventional explosives and little or no nuclear reactions took place. Plutonium was ejected vertically to altitudes up to 800 meters and was dispersed by the prevailing winds over considerable distances. This caused the contamination of considerable portions of land. Plutonium could be found many kilometers away from the detonation points.

ARL identified plutonium in basically three forms — as superficially coated on materials such as pieces of metals, plastic, wire, and lead bricks that were part of the experimental assemblies; as tiny fragments or

particles, not always visible to the eye but easily detectable by a gamma monitor; and as very finely dispersed material consisting of contaminated soil particles and recondensed plutonium particles in the same size range as the soil itself.

Samples of this material were collected and separated by sieving down to a diameter of 45 micrometers to determine mass and activity concentrations. Results showed that the greatest mass was generally associated with the 250-500 micrometers fraction; however, most of the activity (about 41%) was concentrated in the fraction below 45 micrometers, which contained only 5% of the total mass.

The lowest fraction was further fractionated into seven aerodynamic sizes ranging from 45 micrometers to less than three micrometers. In this way, the inhalable fraction — defined as that with a size less than seven micrometers — was also identified.

Particles were also identified in an 800-gram sample of soil (having an activity of 25 Bq of americium-241), broken down by a process of binary separations into discrete particles. All the sample's activity was found to be contained in the separated 54 hot particles. The activity of the individual particles was estimated to be in the range 0.1 to 2.0 Bq of americium-241 and the average diameter was about 20 micrometers.

The study also identified a large number of sub-millimeter hot particles which were analysed by high resolution gamma-ray spectrometry (to determine the ratio of plutonium-239 to americium-241) and for biological uptake. Five sub-millimeter particles with activities ranging from 30 Bq to 5 kBq were also analysed by proton-induced X-ray emission spectroscopy to gain information on their elemental composition and homogeneity.

In these particles, which were several hundreds of microns in diameter, plutonium and uranium were found homogeneously distributed on the surface. The major elements identified were aluminum (1.8%), potassium (2.3%), calcium (1%), iron (23%), lead (1.9% to 35%), uranium (2.9% to 0.8%) and plutonium (19%). Dissolution studies in simulated lung fluid indicated that the particles had no significant solubility.

STUDIES IN FRENCH POLYNESIA

Other investigations of hot particles were conducted in French Polynesia by an expert team as part of the international Study on the Radiological Situation at the Atolls of Mururoa and Fangataufa, completed in 1998. From July 1966 to September 1974, forty-one atmospheric nuclear tests were conducted there. In addition, five safety trials were conducted on the surface at the northern tip of the Mururoa atoll, in areas generally referred to as the Colette region.

The safety trials were conducted to investigate the behaviour of the core of nuclear devices under simulated faulty detonation conditions. The core was destroyed by a conventional explosive detonation with the consequential dispersion during each test of about 3.5 kg of plutonium-239 in the form of finely divided plutonium and plutonium oxide. Although extensive cleanup operations were done in 1982-87, plutonium hot particles were left on the surface of the Colette region and in the adjacent sand bank in the lagoon.

As part of the international Study, the residual contamination in the terrestrial environment of Mururoa and Fangataufa, including the Colette region, was assessed by a team of scientists. A sampling campaign was organized and coordinated by the IAEA's Seiberdorf Laboratories and conducted in the summer of 1996. This was followed by extensive radiochemical measurements on about 300 samples. *(See article, page 24.)*

The analysis of the residual contamination in the Colette region identified the presence of plutonium-containing hot particles. Twenty relatively large particles, ranging in size from 200 micrometers to one millimeter, were separated from coral debris and crushed coral rocks. They were measured by high resolution gamma spectrometry to evaluate their activity and the ratio of plutonium-239 to americium-241. The activity of plutonium-239 was found to fall in the range 5 to 300 kBq, although a particle with a level of about one MBq was also found. The americium-241 activity in the particles was in the range 0.2 to 5.6 kBq. Six of these hot particles, with diameters ranging from 200 to 500 micrometers, were also studied by optical microscopy and micro X-ray fluorescence. The observations revealed that some particles had a smooth,

glassy compact nature while others appeared rough and could even be conglomerates of smaller particles.

In addition to plutonium, the particles were found to contain other elements. They included uranium and neptunium at concentrations 10 to 100 times lower than plutonium; calcium (indicating they had a calcium carbonate matrix); iron (indicating their metallic nature); chlorine (most likely from the sea salt); and traces of manganese, nickel, chromium, cobalt, and titanium (probably reflecting the composition of the steel containers of the fissile material used for the safety trials).

To estimate the distribution of plutonium in coral debris, a 1053-gram sample was also sieved in seven size fractions. The activity of plutonium and americium in the various fractions were measured by high resolution gamma spectrometry. The results showed that 99.9% of the mass and 95.8% of the activity were present in particles larger than 250 micrometers.

Nevertheless it must be pointed out that the study did not exclude the presence of particles smaller than 10 micrometers (containing the inhalable fraction) with plutonium-239 activities of several hundred becquerels. *In vitro* dissolution studies in serum simulant of six hot particles showed that the quantity of plutonium solubilized was in all cases less than 0.07% of that initially present in the particles. This indicated dissolution characteristics similar to those of the particles from the Maralinga nuclear test site.

FUTURE NEEDS

The investigation of hot particles is highly relevant to correct evaluations of radiation hazards at sites which were contaminated by nuclear-weapon testing. Studies so far, however, have led to more questions than final answers. They indicate that the information on hot particles and particle fractionation obtained through radiochemical, chemical and physical analyses of debris is still too small and too scattered when viewed against the diversified nature of particles produced and the number of variables requiring investigation. Therefore, the complex phenomena which control the formation, the chemical and radiochemical composition, and the physical and morphological properties of hot particles and their behaviour in the natural environment are still not fully understood.

It is apparent that the phenomena leading to the formation of hot particles and their behaviour in the ecosystems are complex. Thus, any generalization must be advanced with caution. It is believed that further progress in this area will require multi-disciplinary teams of scientists. They should include physical chemists, specialists in non-destructive microanalytical techniques, radiochemists, nuclear physicists, and health physicists.

It also must be pointed out that hot particles of various sizes and composition containing actinides, fission or activation products are released to the environment from other sources besides nuclear testing. For example, they were produced by the Windscale pile fire in 1957, the B-52 aircraft crash at Thule in 1968, the satellite Cosmos crash in Canada in 1978, and the Chernobyl accident in 1986. Hot particles have also been released to the environment at nuclear facilities contributing to the production of fissile material for nuclear-weapon programmes.

In general, any facility for the processing of nuclear material is known to release small, but detectable, amounts of radioactive and non-radioactive isotopes to the immediate environment. These releases, which are often insignificant from the radiation hazard point of view, can be in the form of waste streams, aerosols, or particles and can be found at some distance downwind or downstream of the point of release.

The same modern analytical techniques used to investigate hot particles from nuclear testing can be applied to studying other sources of environmental radioactivity. They enable scientists to measure extremely small amounts of chemical elements and isotopes present in released radioactive materials, thereby providing information about the process which formed them.

Hopefully in years ahead, more studies on hot particles will be carried out at sites that have been contaminated by nuclear-weapon testing and various types of nuclear accidents, and at sites near nuclear installations. The work will benefit from advances in instrumental analytical techniques. It will further lead to greater understanding of the role that hot particles play in evaluating radiation hazards, and in providing information on the type and purpose of the facilities that generated them. ❐