Reports

Iodine-131 Fallout from Underground Tests

Abstract. *Evidence that the fallout of iodine-131 in the Midwest during May 1962 originated in Nevada is provided by data on air trajectories, radioactivity in milk, and other findings. Selective escape of radioiodine from vented explosions, and the rapid transfer of iodine from the lower atmosphere to the biosphere, place significance on even minor releases of radioactive fission products from subsurface nuclear explosions.*

The highest levels of iodine-131 fallout in the United States prior to the end of 1958 were associated with tropospheric debris from nuclear tests in Nevada (*1–4*). However, when nuclear testing was resumed in September 1961, distant Soviet atmospheric shots were blamed for the iodine-131 fallout in southeastern United States in late September 1961 (*5*, *6*) and an atmospheric test at far-away Christmas Island was given as the source of the iodine-131 fallout over a part of midwestern United States in mid-May, 1962 (*7–9*).

It is my contention, first expressed in June 1962 (*10*), that underground tests in Nevada were, in many recent instances, the principal source of iodine-131 fallout in the United States. This view is supported by subsequent specific associations of domestic underground tests with iodine-131 fallout, as follows: (i) concentrations of iodine-131 in milk at Spokane and Seattle reached 1000 pc/liter in late June 1962 and were attributed (*11*) to the vented Nevada underground test of 13 June 1962; (ii) the appreciable iodine-131 fallout in Utah during July 1962, was due in part to the underground cratering explosion (Sedan) of 6 July 1962 (*12*).

More recently, Penn and I (*13*) pointed out that the vented Nevada underground test of 15 September 1961 could reasonably be held responsible for most of the iodine-131 fallout in southeastern and southcentral United States during late September 1961. However, during the same period, surface air radioactivity originating from Soviet tests (*13*, *14*) was experienced in some areas of Canada and eastern United States. Because possible trajectories for the Nevada and Soviet sources intersect in southeastern United States, the relative importance of these two sources for the appreciable iodine-131 fallout in this region is unlikely to be resolved on a meteorological basis. However, preliminary results from radiochemical analysis of filtered samples of surface air from the affected areas indicate that the vented underground test in Nevada of 15 September 1961 was an important contributing source (*15*).

The significance of iodine-131 in fallout is due to the fact that it readily finds its way to the thyroid glands of humans and animals (*1*, *2*). The short-lived (8-day half-life) iodine-131 in thyroids is derived mainly from tropospheric fallout which deposits on the surfaces of plants which, in turn, are eaten by dairy animals. Iodine-131 in fresh milk is the main source of iodine-131 in man (*5*). Concentrations up to 0.06 microcuries of iodine-131 per liter of milk were observed in Utah during July 1962, corresponding to an infant thyroid dose of about 14 rad (*16*). Similar and higher doses have been estimated for other periods of fallout (*4*, *17*).

Published records (*7–9*) reveal a number of unusual features in the mid-May 1962 appearance of iodine-131 in milk in several midwestern states. These features are difficult or impossible to explain on the hypothesis that the iodine-131 originated from a test on Christmas Island, as advanced by Machta and others (*7–9*). However, they follow readily from the hypothesis that the iodine-131 originated from a vented underground test in Nevada. This hypothesis is supported by certain physical and chemical data which suggest that there is considerable selective venting of iodine-131 from underground tests.

Pertinent data of the U.S. Public Health Service and the St. Louis County Health Department are summarized in Table 1 and Fig. 1. The following peculiarities of this iodine-131 fallout seem difficult to account for on the hypothesis of a stratospheric cloud from Christmas Island.

1) The iodine-131 fallout was accompanied by an almost imperceptible increase in surface air radioactivity in the May 1962 instances (*9*, *14*). By contrast, the September 1961 instance in southeastern United States, and the July 1962 instance in Utah involved 100 times larger relative increases in surface air radioactivity.

2) The initial rise in radioiodine in St. Louis and Kansas City (Table 1 and Fig. 1), adjusted for the 1 to 2 days between grazing and the distribution of pasteurized milk, indicates fallout arrival in Missouri and Kansas areas on 8 or 9 May. A similar fallout arrival time is indicated for the Wichita milkshed area (*8*), which received the heaviest iodine-131 fallout. Although there was widespread rainfall within and east of the fallout area during the period 8 to 13 May, the iodine-131 fallout was confined to the midwest.

3) The data indicate a later arrival for fallout in Minnesota, Iowa, and Colorado. Another increase of iodine-131 in milk occurred during the 4th week of May.

It seems difficult, if not impossible,

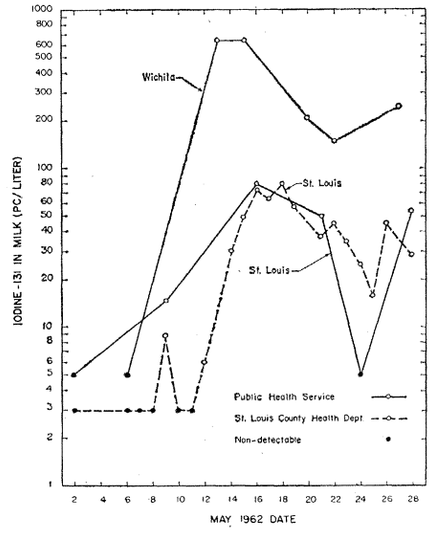

Fig. 1. Concentrations of iodine-131 in pasteurized milk from Wichita, Kansas, and St. Louis, Missouri, milkshed areas during May 1962 (*9*).

Table 1. Iodine-131 in pasteurized milk, May 1962 (*8*).

Location	I^{131} pc/lit. (sample date)			
Little Rock, Ark.		30(14th)	<10(23rd)	40(24th)
Denver, Colo.	<10(10th)	45(17th)	<10(24th)	
Des Moines, Iowa	<10(10th)	300(17th)	60(24th)	75(28th)
Wichita, Kan.	670(13th)	660(15th)	154(22nd)	250(27th)
Kansas City, Mo.	45(10th)	605(18th)	150(25th)	
St. Louis, Mo.	15(9th)	80(16th)	50(21st)	75(28th)
Minneapolis, Minn.	<10(11th)	290(18th)	170(25th)	
Cincinnati, Ohio	<10(10th)	50(17th)	20(24th)	

to believe that a stratospheric cloud source passing overhead at a height of 15 km could account for the repeated selective deposition of iodine-131 in this limited area of the Midwest in May 1962. Stratospheric penetration by thunderclouds suggested by Machta (*7*) should have led to fallout in the widespread rainfall areas to the east. The inappreciable rise in surface air radioactivity and the later fallout occurrences also are unexplained.

The phenomena, on the other hand, seem to fit well with the hypothesis of Nevada origin. Air trajectories by Penn (*18*) for low atmosphere levels following the 7 and 12 May underground tests are given in Fig. 2. Trajectories for the test on 7 May pass through and just north of the Missouri-Kansas area on 8 and 9 May, consistent with the observed arrival of iodine-131 fallout. Similarly, trajectories for the 12 May shot pass through and north of the Iowa-Minnesota area on 13 and 14 May.

The occurrence of heavy iodine-131 fallout without appreciable mixed fission products brings to mind the fallout resulting from the accident at Windscale No. 1 Pile on 10 October 1957. By analogy, the selective release of iodine-131, and the activity of its parent tellurium-131, from vented underground tests in Nevada would be equally possible. During May 1962, four underground shots were fired at the Nevada test site, on the 7th, 12th, 19th, and 25th (*19*). Of these underground shots, the one on the 19th was actually reported to have vented (*19*) and to have produced a radioactive cloud. If the other underground shots in May vented even to a limited degree, the selective deposition of iodine-131 with only limited rise in surface air radioactivity, the arrival times and locations, and the repeated rises in iodine-131 fallout are explained.

Thus it seems that the occurrence of several peaks of iodine-131 fallout in the Midwest during May 1962 can be reconciled with the several underground shots in Nevada more readily than with the repeated selective deposition of stratospheric debris.

The radiochemical composition of contaminated substances in the atmosphere, which result from the venting of underground nuclear tests, depends mainly on the time-temperature history of the fireball, the time and extent of venting, and the nature of the explosion medium (*20*). The vented debris will be enriched with gaseous and volatile fission products and some of their radioactive daughter products. The chains of decay of the fission products which are of greatest interest for the evaluation of the venting process and the composition of the effluent radioactivity in the atmosphere are:

$$As^{85}(0.43\ sec) \rightarrow Se^{85}(40\ sec) \rightarrow \begin{matrix} & & Kr^{85m}(4.4\ hr) \\ & 82\% \nearrow & \downarrow \\ Br^{85}(3.0\ min) & & \\ & 18\% \searrow & Kr^{85}(10.3\ yr) \end{matrix} \quad (1)$$

$$Br^{89}(4.5\ sec) \rightarrow Kr^{89}(3.2\ min) \rightarrow Rb^{89}(15\ min) \rightarrow Sr^{89}(51\ day) \quad (2)$$

$$Br^{90}(1.4\ sec) \rightarrow Kr^{90}(33\ sec) \rightarrow Rb^{90}(2.7\ min) \rightarrow Sr^{90}(27\ yr) \rightarrow Y^{90}(64\ hr) \quad (3)$$

$$Sn^{131}(3.4\ min) \rightarrow \begin{matrix} & & Te^{131m}(30\ hr) & & \\ & 5\% \nearrow & \downarrow & \searrow & \\ Sb^{131}(23\ min) & & & & I^{131}(8.1\ day) \\ & 95\% \searrow & Te^{131}(25\ min) & \nearrow & \end{matrix} \quad (4)$$

$$Sn^{132}(2.2\ min) \rightarrow Sb^{132}(2.1\ min) \rightarrow Te^{132}(78\ hr) \rightarrow I^{132}(2.3\ hr) \quad (5)$$

$$Te^{137}(3\ sec) \rightarrow I^{137}(22\ sec) \rightarrow Xe^{137}(3.9\ min) \rightarrow Cs^{137}(27\ yr) \quad (6)$$

$$Xe^{140}(16\ sec) \rightarrow Cs^{140}(66\ sec) \rightarrow Ba^{140}(12.8\ day) \rightarrow La^{140}(40\ hr) \quad (7)$$

Goeckermann (*20*) has described the venting process and the disposition of some of the radioactive products for underground tests in Nevada tuff. For low yield shots, the maximum radius of the underground bubble is reached within 10 msec. If the cavity collapses, it takes place within tens of seconds, accompanied by a sudden release of pressure and the escape of high temperature water vapor and gases from the central cavity. The zone of rupture extends upward, sometimes to the ground surface.

All the fission products of biological importance that pass through the food chain (strontium-90, strontium-89, barium-140, iodine-131, and cesium-137) have volatile or gaseous precursors, or both. If rupture occurs, each of these activities will be enriched appreciably in the rupture zone and in the atmospheric fallout mixture. Goeckermann (*20*) gives the distribution of some of the radioactive bomb products in the glass shell resulting from the solidification of the fused tuff and in the rupture chimney. The vented material shows nearly tenfold enrichments for strontium-89, fivefold for strontium-90, and several fold for cesium-137 and barium-140.

The extent to which iodine and tellurium are released into the atmosphere by vented underground shots is of special interest in connection with the problem of iodine-131 fallout. The amount of iodine-131 which is formed directly by fission is very small. Most of the iodine-131 accumulates from the decay of its precursors in the mass-131 decay chain, No. (4). Most of the mass-131 chain is in the form of antimony-131 and tellurium-131 immediately after the nuclear explosion. Chamberlain (*21, 22*) showed that at Windscale, tellurium-132 and iodine-131 were selectively vented in nearly the same ratio that applied in the pile at the time of escape. The chemical form of the escaping tellurium is unknown, although elemental tellurium, Te_2, is a likely possibility. The initial yield of tellurium-131 for uranium-235 fission ranges from about 0.13 for thermal neutrons (*23*) to an experimental value of 0.44 for 14 Mev neutrons (*24*). Intermediate chain yield values should apply for fission spectrum neutrons on uranium-235 and plutonium-239. Thus the radioactive mixture vented immediately after underground nuclear explosions can release up to about one-fourth of the mass-131 chain into the atmosphere, as tellurium-131 principally. Following the initial venting process, additional pressurized vapor and gas may continue to escape for hours, resulting in the further release of tellurium-131 and iodine-131 into the atmosphere. One possible chemical form of the escaping iodine-131 is the monochloride vapor (ICl, boiling point 97.4°C), in view of the relatively high natural abundance of chlorine. The iodine-131 also may be partially converted to methyl iodide

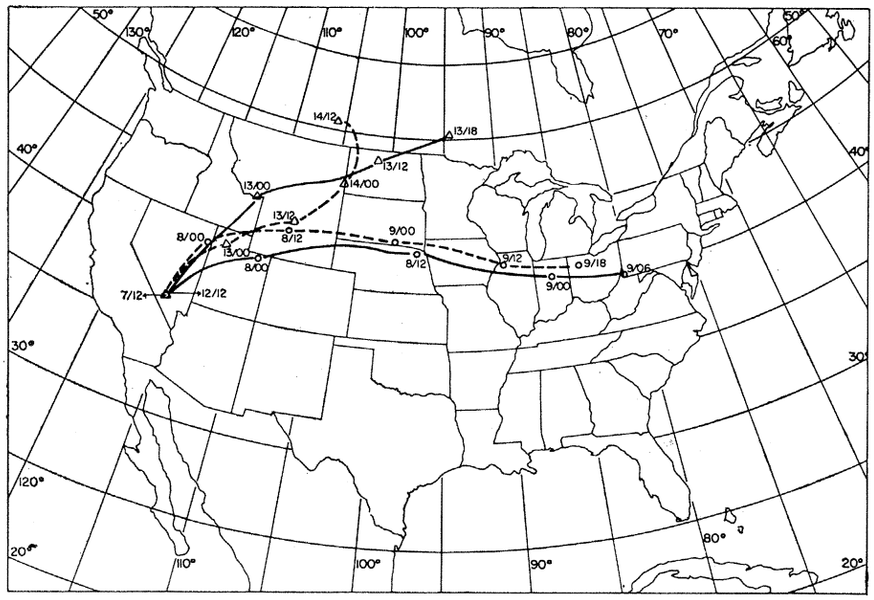

Fig. 2. Lower troposphere trajectories by Penn (*18*), originating from the Nevada test site at 1200 GMT, 7 May 1962 (circles), and 1200 GMT, 12 May 1962 (triangles). Dashed lines designate 4 km altitude, and solid lines, 6 km altitude trajectories.

and other alkyl iodides because of thermal and radiation-induced reactions with organic material during the venting process and subsequently because of photochemical interaction with trace organic constituents in the atmosphere (*25*).

The Windscale accident and vented underground explosions show only a limited parallel in behavior with respect to the venting process. Both have provided a chemical environment which has resulted in the selective venting of volatile and gaseous products, including radioiodine (*20–22, 26*). However, the composition of the atmospheric contamination mixture will vary widely for reactor accidents and vented shots due to differences in radioisotope content, chemical environment, and details of the venting process. Because the long-lived radioisotopes accumulate in reactors, there will be an extensive enrichment of cesium-137 and other long-lived volatile products in the effluent of reactor accidents. These considerations provide a basis for distinguishing the origin of the radioactive product mixture.

The release of radioactive debris into lower levels of the troposphere by vented underground shots, cratering shots, and nuclear reactor accidents results in a pattern of fallout behavior which enhances the significance of fallout for small amounts of radioactivity. Unlike tropospheric fallout, which is deposited over a wide latitudinal band completely around the earth with a mean atmospheric life of about 1 month, this "weather layer" fallout, enriched in the volatile and gaseous products, drifts downwind in surface air and can be deposited in high concentrations over areas of only tens of thousands of square miles within several days.

The Windscale accident (*22*) involved the emission of an estimated 27,000 curies of iodine-131 from the 130-m high stack of the pile. About 5000 curies, nearly 20 percent of the total, were deposited within the 0.1 $\mu c/m^2$ (microcuries per square meter) isopleth in northwestern England. It was concluded (*22*) that diffusion of iodine-131 gas or vapor from the lower atmosphere down onto the surface of vegetation was the main mechanism of deposition, although patchy, light rainfall could have contributed in some areas. Booker (*27*) indicated that about 80 percent of the total iodine-131 fallout per square meter was deposited on the vegetation. He also showed that the average concentration of iodine-131 in milk, in microcuries per liter, corresponds to about one-eleventh of the iodine-131 deposit in $\mu c/m^2$ on vegetation. The Windscale fallout of about 5000 curies of iodine-131 over an area of 16,000 km^2 in northwestern England thus corresponds to an average concentration of iodine-131 in milk of about 23,000 pc/lit. This single iodine-131 fallout occurrence results in an estimated thyroid dose averaging 2 rad to adults and 17 rad to infants who consumed 1 liter per day of milk from the affected area.

By comparison, the iodine-131 fallout in Utah during July 1962, corresponded to an average infant thyroid dose of 1 rad. The very irregular distribution of the Utah fallout, leading to estimated infant thyroid doses up to 14 rad in some areas (*16*) illustrates a significant feature of fallout from clouds in the lower troposphere. Compared to Windscale, the average fallout in Utah was about one-seventeenth as high as that around Windscale, and covered an area nearly fourteen times as large. Roughly 5000 curies of iodine-131 was deposited in each instance. This amount is only about 3 percent of the iodine-131 produced by 1 kiloton of fission energy yield.

In the Plowshare cratering explosion (Sedan) in Nevada on 6 July 1962, a 100-kiloton thermonuclear device was used for which the fission yield was about 30 kilotons. The corresponding iodine-131 production amounts to about 5 megacuries, 1000 times the estimates for the July 1962 fallout in Utah or the fallout in Windscale. The physical and chemical properties of iodine-131 in the atmosphere are too poorly known to predict its distribution in nearby areas and downwind. The relatively minor iodine-131 fallout attributed to the Sedan shot gives us no basis for complacency about similar shots at other times and for other environments.

Of the 51 announced Nevada tests from 15 September 1961 to 24 August 1962, a total of ten produced radioactive clouds: on 15 September 1961 and on 5 March, 14 April, 19 May, 13 June, and 6, 7, 11, 14, and 17 July, 1962 (*26*, *28*). An additional 15 of the 51 are reported (*29*) to have vented to a limited degree, on 10 and 29 October, and 22 December 1961 and on 18 and 30 January, 9, 15, 19 (two), and 24 February, 1 and 8 March, 27 and 28 June, and 27 July. To this list must be added the vented Plowshare shot (Project Gnome) at Carlsbad, New Mexico, on 10 December 1961 and the 7 May 1962 Nevada underground shot which seems to account for the subsequent iodine-131 fallout in the Kansas-Missouri area (Table 1 and Fig. 1). This summary does not include those which may have vented among the 23 additional underground tests in Nevada during the period from August 1962 to June 1963, inclusive (*26*).

Of this total of 75 continental tests, three were surface explosions, several were shallow underground cratering explosions, and the remainder were underground shots. Those which resulted in

atmospheric contamination either produced relatively low-lying clouds or vented small amounts of radioactive vapors and gases into the lower layers of the troposphere. Considering the small amounts of iodine-131 required to explain the highest observed levels of iodine-131 in fallout and the apparent ease in the selective venting of tellurium and iodine, there has been no lack of sources in the continental United States to explain most, if not all, of our iodine-131 fallout.

Prior to September 1961, when nuclear testing was resumed, the fallout of iodine-131 resulted principally from sources in the upper troposphere. The heavy fallout of radioactive debris over Troy, New York, on 25 April 1963, about 36 hours after the Simon explosion in Nevada (*4*), was largely due to a radioactive cloud with its base at an estimated altitude of 10 km. The fallout in southeastern United States in late September 1961 was similarly due to debris, initially in the middle or upper troposphere, carried down into surface layers of the troposphere by unusual meteorological conditions (*13*). Most radioactive debris in clouds in the upper troposphere remains there for an average of about 1 month. Thus short-lived radioactive products like iodine-131 decay substantially before falling out and are widely dispersed at low intensity.

For underground explosions and low-yield surface tests the fate of the radioactive products is less predictable. Radioactive products pass into the lowest layers of the troposphere where they can be deposited over a smaller geographical area within a few days. This increases the concentration of fallout and renders more hazardous the effects of short-lived radioactive products like iodine-131. Even underground tests which are largely contained below ground with only a limited release of radioactive gases and vapors cannot be overlooked as sources of iodine-131 fallout. Selective venting must explain the appreciable fallout of iodine-131 without corresponding increase in surface air radioactivity as in midwestern United States during May 1962.

Control of iodine-131 fallout will be more effective if we control its sources rather than the distribution and consumption of fresh dairy products. Better containment of underground tests will result from placing the shots at greater scaled depths (*30*), from avoiding areas of faulty soil structure, and from greater care in sealing access tunnels. Moreover, less damage will be caused by venting explosions if all tests are carried out during winter under favorable meteorological conditions so that iodine-131 is not deposited where dairy animals are at pasture.

The high frequency of venting of radioactive products from previous underground tests suggests that either there was no serious attempt to contain them, or that containment is difficult and uncertain. Atmospheric radioactivity from vented underground explosions may be detectable in the lower atmosphere at large distances and may be distinguished from that resulting from atmospheric tests or reactor accidents.

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30. The scaled depth for underground explosions is defined by the product, $D \times W^{-1/3}$, with D, the actual depth in feet, and W, the energy release in kilotons. The scaled depth for containment is about 500 for bedded tuff in Nevada but varies widely with soil material and soil structure.
31. I am indebted to Samuel Penn of the Air Force Cambridge Research Laboratories, Bedford, Mass., for computing the tropospheric air trajectories, Fig. 2, and for helpful discussion on the meteorological aspects of this paper.

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Cholinesterase Inhibition in Spider Mites Susceptible and Resistant to Organophosphate

Abstract. *Evidence is provided that organophosphate resistance in a strain of spider mites is due to decreased sensitivity of its cholinesterase to organophosphates. The cholinesterase activity of the susceptible strain in vitro was three times that of the resistant strain of mites.*

Cholinesterase and acetylcholine are known to occur in the red spider mite, *Tetranychus urticae* Koch (*1*). Inhibition of this enzyme is generally assumed to be the mode of action in organophosphate poisoning. For this reason the cholinesterase of the mite was studied in vitro and a strain resistant to several organophosphates was also studied, because resistance may theoretically be caused by a difference in some characteristic of the cholinesterase of susceptible and resistant strains.

The strains of *T. urticae* used were described by Helle (*2*). The resistant strain was obtained by repeated backcrossing with the susceptible strain,