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the Chernobyl Nuclear Reactor Accident

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RADIOACTIVE FALLOUT FROM THE CHERNOBYL NUCLEAR REACTOR ACCIDENT*

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When the Chernobyl nuclear-reactor accident was reported on April 29, 1986 it appeared that the radioactive plume would extend over the United States. The prevailing winds were known¹ to be carrying the radioactive plume over Asia and the Pacific Ocean toward the western coast of the United States. The modeling efforts of the Atmospheric Release Advisory Center (ARAC) predicted that the plume would reach the western United States May 5 in the upper atmosphere and May 10 near ground level.

At LLNL the Environmental Quality Verification Program (LLNL-EQVP) carries out an extensive environmental surveillance program which includes systematic monitoring of air filters for gamma-emitting radionuclides. Following the Chernobyl accident additional air sampling stations were added. At the Geophysical Institute at the University of Alaska aerosol samples are routinely collected for chemical determination of the character and levels of air pollution. Following the Chernobyl accident three of the air sampling stations were used to monitor the levels of radioactive fallout in these areas. At two stations the early filters were changed daily, providing a very good time profile of the fallout.

The LLNL-EQVP air samplers consist of 520 cm² Whatman 41 cellulose fiber filters through which ambient air is pumped at a rate of 400 to 700 L/min. The air filters are at a height of 122 cm above ground level. Under normal conditions twenty-two air samplers operate continuously in and around LLNL. The filters are changed weekly and counted monthly. To avoid compromising our normal sampling, two separate air samplers were installed to obtain a much better time profile of the Chernobyl-related activity. As the prevailing wind is west-southwest in May and June, one sampler was placed on the west perimeter of LLNL and another 1.5 km south to assure that the activity measured was not influenced by LLNL operations. The samplers operated at an average flow rate of 400 L/min, and the air filters were changed three times per week and gamma-counted within hours of removal from the samplers.

In Alaska, air samplers, similar to those described above but with flow rates of 700 to 1600 L/min, were already in place at Fairbanks, Poker Flat Rocket Range, and Barrow. Under normal conditions the filters are analyzed by neutron activation analysis to determine the chemical composition of the pollution aerosols. Upon report of the Chernobyl accident the filters were gamma-counted without neutron irradiation to determine the level of radioactive fallout. A sufficient number of filters were analyzed prior to the arrival of the radioactive plume to provide an upper limit on the normal activity levels. From April 29 until June 15, 1986 the filters were changed on a daily basis. After June 15 the filters were changed biweekly. The Alaskan filters were gamma-counted within days after acquisition.

In particulate samples of normal Livermore Valley air, we routinely detect ⁷-Be, ⁴⁰-K, and ¹³⁷-Cs by gamma-ray spectroscopy. The ⁷-Be and ⁴⁰-K are naturally occurring isotopes, but low levels of ¹³⁷-Cs occur primarily from long-term global fallout and fallout resuspension. Following the Chernobyl accident, other isotopes, not normally detected, were observed in the air-filter samples. These were ¹⁰³,¹⁰⁶-Ru, ¹²⁹m,¹³²-Te, ¹³¹-I, ¹³⁴,¹³⁶-Cs, ¹⁴⁰-Ba, ²¹²-Pb, and low concentrations of ⁹⁹-Mo, ¹⁴¹-Ce, ¹¹⁰m-Ag, and ¹²⁵-Sb. All of these isotopes can be attributed to reactor by-products. However, the ²¹²-Pb detected here is attributed to ²²⁸-Ra emanations from the soil. The ²¹²-Pb in the air filters is not normally detected because the decay time prior to counting is sufficient to allow the 10.6 hr half-life

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to decay away. As mentioned above, the special air filters were counted within hours of the end of the collection period.

In calculating the concentrations of the isotopes in air, we accounted for the decrease in the air flow rate caused by dust clogging, and for the decay of the short-lived isotopes on the filter during the collection period. The concentrations were decay corrected to the mid-point of the collection period but not to the time of the Chernobyl accident. This is for two reasons. We were interested in the concentrations of the isotopes when they were detected in the Livermore area and not at the time of their release from the reactor. Secondly, the release was not instantaneous but lasted for many days. Our calculations assumed a constant air concentration during the collection period. This assumption is not justified by the data, although lack of further information gives us no alternative. The isotopic concentrations reported for the Livermore area are the average of the values for the two special air sampling stations. The concentration at the two stations generally differed by less than ten percent.

The maximum concentrations during the May 7-9 Livermore collection was followed by a regular damped oscillatory concentration pattern peaking every six to seven days. However, our long collection periods (2 to 3 days) make it difficult to determine accurate radionuclide deposition times. Figure 1 shows these trends for 103-Ru, 131-I, and 137-Cs isotope concentrations vs the numerical day from January 1, 1986 at the end of the collection period. To highlight the similarities of the time profiles of the concentrations, each isotope was given a different concentration scale and plotted so that the maximum values of May 7-9 coincide on the scale. The damped oscillation of the concentrations was seen for all the isotopes.

There are several possible explanations for the concentration-time profile measured in the Livermore area. Since the radioactivity release from the Chernobyl-reactor site lasted for many days it is possible that part of the variation is due to the release pattern. However, the radioactivity was detected for several weeks after the last radioactive release so this is probably only a partial explanation. Another explanation is that the air mass containing the radioactive plume passed over the western United States several times before the level of activity was below our detection limits. Each time the air mass passed over our area more activity would be detected, at successively lower concentrations due to continuous deposition and radioactive decay. This would result in the pattern seen in Fig. 1. This explanation has been proposed by others⁷ to explain similar concentration patterns. It has also been suggested that such an oscillatory pattern is normal for surface deposition from a wide ranging plume. The variety of factors which affect the plume transport rate can produce a striated concentration pattern within the plume. Then as the plume passes over a given area the concentration-time profile of the deposited material will have an oscillatory pattern which will mimic the plume striations⁸.

In Fig. 1 we also have plotted the decay lines for the three isotopes assuming purely radioactive decay of the maximum activity measured for the time period, May 7-9. Notice that the 103-Ru and 131-I concentrations appear to show behavior that can be explained by continuous deposition and radioactive decay. However, the 137-Cs data shows a much faster removal rate than expected, and implies an accelerated deposition rate. In fact, the disappearance rate of the 137-Cs is very close to that of the 131-I. This could be possible if the major species of 131-I being transported over the western United States were CsI. The radioactive decay of the iodine would result in the break-up of the CsI and enhance the deposition of the Cs⁹.

The arrival of the radioactive plume in Alaska coincided with its detection in California. From April 29 through May 5, 1986 the levels of radionuclides on the daily air filter samples at the three sampling stations in Alaska were below the detection limits of the systems. Filters which were put in place after 17:20 YDT (Yukon Daylight Time) on May 5 began to show abnormal levels of radioactivity.

In Fig. 2 we show the time profile of the 131-I concentrations measured at one of the three air sampling stations due to the lack of space in this paper. The results from the Poker Flat station are not included because only a few samples were received and the results of these agree with the values measured at the close-by Fairbanks station. Only the 131-I

data are presented because the majority of the samples were analyzed using NaI detectors and only the 131-I peak was relatively free from interference.

The general activity levels measured in Alaska are lower than those detected in the Livermore area. This was expected from the path of the radioactive plume. More northerly areas are expected to receive less fallout. It is fortuitous that the Siberian High (a normally strong springtime atmospheric teleconnection between Alaska and Central Eurasia) had broken down several weeks prior to the Chernobyl accident. Otherwise this clockwise flow, which is known to bring industrial pollutants from Central Eurasia into the Alaskan Arctic causing the "Arctic Haze", would have greatly increased the fallout levels in Alaska.

The daily sample in Alaska presents a better time profile of the early variation in the 131-I concentration than could be obtained by the thrice weekly samples taken in Livermore. As in Fig. 1 we have included lines indicating the effect of radioactive decay on the maximum value obtained at each sampling station during the first ten days. Although we see a fairly regular oscillation in the 131-I values during the first sixty days of the sampling, the levels of 131-I do not seem to be decreasing along the decay lines as they did at Livermore. This can be explained by the unusual air flow patterns near the Arctic Circle. These air flow patterns are known to affect the transport and deposition of particulates in this region and have been studied as explanations of the Arctic air pollution. Full descriptions of these weather patterns are beyond the scope of this report. The unusually high value on July 5 (day 186) could be due to the influence of anticyclonic air flow during this time period which brought a "heat-wave" and slight precipitation to the area.

The Barrow site is 805 km north of the Fairbanks and Poker Flat sites and 265 km north of the Arctic Circle. Because this is so far north we expected and found lower 131-I concentrations in air samples. With the breakdown of the Siberian High mentioned above, the radioactive plume would follow the prevailing easterly winds and only a small portion of the activity would reach north of the Arctic Circle. The general trend of the 131-I concentration values shows three major peaks near day 130 (May 10), day 142, and day 155. Other fallout measurements in the Barrow, Alaska area, by Larsen et al., show a similar pattern where the maximum detected values occurred late in May. Unfortunately we have insufficient meteorological data to attempt to explain the air flow patterns that could have caused this variation in radioactivity.

REFERENCES

1. P. H. GUDIKSEN, R. LANGE, "Atmospheric Dispersion Model of Radioactivity Releases from the Chernobyl Event" to be published in *Nature*, UCRL-95363 (1986).
2. A. J. THOMAS, I. M. MARTIN, *Nature*, 321 (1986) 817.
3. P. H. GUDIKSEN, Private Communication, Lawrence Livermore National Laboratory, June 1986.
4. J. K. DAWSON, R. G. SOWDEN, Chemical Aspects of Nuclear Reactors, vol. 2 (1963) p. 132.
5. R. J. LARSEN, C. G. SANDERSON, W. RIVERA, M. ZARRICHEL, "A Compendium of the Environmental Measurements Laboratory's Research Projects Related to the Chernobyl Nuclear Accident", H. L. VOLCHOCK, N. CHIECO (Eds), EML-460, (1986) p. 1.

Figure 1. Time profiles of the concentrations of ^{103}Ru , ^{131}I , and ^{137}Cs in the air in Livermore, California. To highlight the similarity between the time profiles of the various isotopes the linear concentration scales of each are adjusted to overlap the maximum concentration points at Day 132. The units are in atoms/ m^3 for comparison of the various isotopes. Also shown is the predicted value for pure radioactive decay of the maximum values detected for each isotope. (ND = not detected)

Figure 2. Time profile of the ^{131}I concentrations in the air in Fairbanks, Alaska. The linear concentrations are plotted in atoms/ m^3 to correspond with the values in Figure 1. Also shown are the predicted values for the pure radioactive decay of the maximum value of the ^{131}I detected in the first 10 days after the Chernobyl accident. (ND = not detected)