

14. The signal from 4 liter  $\text{min}^{-1}$  (STP) of the  $\text{N}_2$  used, equivalent to  $p\text{NO} = 0.13 \times 10^{-9}$  atm, has been subtracted from reported in situ stripping data. If this signal is a water-soluble NO mimic rather than NO (no evidence suggests this), the subtraction is unwarranted. The correction is  $< 3$  percent of the maximum  $p$  NO reported.
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27 March 1979; revised 22 June 1979

## Radioactive Plume from the Three Mile Island Accident: Xenon-133 in Air at a Distance of 375 Kilometers

**Abstract.** *The transit of an air mass containing radioactive gas released from the Three Mile Island reactor was recorded in Albany, New York, by measuring xenon-133. These measurements provide an evaluation of Three Mile Island effluents to distances greater than 100 kilometers. Two independent techniques identified xenon-133 in ambient air at concentrations as high as 3900 picocuries per cubic meter. The local  $\gamma$ -ray whole-body dose from the passing radioactivity amounted to 0.004 millirem, or 0.004 percent of the annual dose from natural sources.*

We observed the passage of radioactive  $^{133}\text{Xe}$ , released from the Three Mile Island reactor, through the Albany, New York, area from 29 March through 2 April 1979. After the announcement of the reactor accident and possible releases of fission products into the atmosphere, air samples were collected in Albany and were analyzed for  $^{133}\text{Xe}$ , which has a half-life of 5.3 days. We also monitored  $^{133}\text{Xe}$  directly in ambient air throughout the entire transit period by observing the 81-keV  $\gamma$ -ray line with a planar intrinsic Ge detector located in a low-background steel chamber. To our knowledge, these measurements provided the only evaluation of Three Mile Island effluents at distances greater than 100 km.

Gas analyses of 1- to 3- $\text{m}^3$  samples of air were performed in two stages: cryogenic and chromatographic separation of Xe, followed by analysis of the  $\beta$ -decay spectrum (maximum energy, 346 keV) by internal gas-proportional counting in low-background systems (1). Aged compressed-air samples were processed through the gas separation system as blanks between Albany air samples. A low residual activity found after the processing of the higher-activity samples did not substantially reduce the sensitivity.

Ambient laboratory air was monitored by an intrinsic Ge diode with an area of 500  $\text{mm}^2$  and a resolution of 630 eV (full width at half-maximum at 81 keV). This instrument was in a low-background steel chamber (3.3 m square; 2.4 m high; wall thickness, 14.5 cm) in which outside air was exchanged about ten times an hour. During the entire period when  $^{133}\text{Xe}$  was recorded, the thin window was covered by a pressed pellet of lake sedi-

ments with a thickness of 0.76  $\text{g cm}^{-2}$ , which reduced the counting efficiency for  $^{133}\text{Xe}$  by about 15 percent. After the transit the detector was calibrated under the same conditions with a virtual point source of  $^{133}\text{Xe}$ . The net count rate in the 81-keV photopeak was measured as a function of the angular and radial position of the source over the entire field of view. Integration yielded an overall efficiency of  $1.03 \times 10^{-4}$  cpm  $\text{pCi}^{-1} \text{m}^3$ . The air volume effectively seen by the detector was 10.2  $\text{m}^3$ .

Errors reported are  $\pm 2$  standard deviations from the root-mean-square counting statistics of sample measure-

ments and background determinations. An additional uncertainty of  $\pm 5$  percent is introduced by the calibration. Detection limits are three times the background standard deviation.

Air samples collected on 30 March showed high  $^{133}\text{Xe}$  concentrations:  $3120 \pm 160$   $\text{pCi m}^{-3}$  at 1500 Eastern standard time (EST),  $3530 \pm 180$  at 1900, and  $3900 \pm 200$  in a 10-hour sample (from 1545 to 0145 the next morning). By 31 March at 0900 the concentration had fallen to  $39 \pm 4$   $\text{pCi m}^{-3}$ . Samples collected on 3 April at 1600 and on 4 April at 1500 contained  $11 \pm 4$  and  $5 \pm 2$   $\text{pCi m}^{-3}$ , respectively.

Ambient air analyses showed the same pattern of  $^{133}\text{Xe}$  concentrations: no counts above background ( $< 360$   $\text{pCi m}^{-3}$ ) before 29 March, but  $1390 \pm 290$   $\text{pCi m}^{-3}$  in a sample spanning 29 March (1230 EST) to 30 March (1500) and  $1060 \pm 180$   $\text{pCi m}^{-3}$  in a sample spanning 30 March (1530) to 2 April (0830). No excess counts were recorded after 2 April. The peak concentrations were more than three orders of magnitude higher than those normally present in ambient air. In 1974 the concentration of  $^{133}\text{Xe}$  from all sources including routine releases from nuclear reactors was 2.6  $\text{pCi m}^{-3}$  for the Albany area (1).

The results (plotted in Fig. 1) indicate that the air mass containing  $^{133}\text{Xe}$  arrived in the Albany area after 1230 on 29 March and before 1500 on 30 March. A more precise arrival time could not be determined, since the diode measurements were integrated over a 24-hour pe-

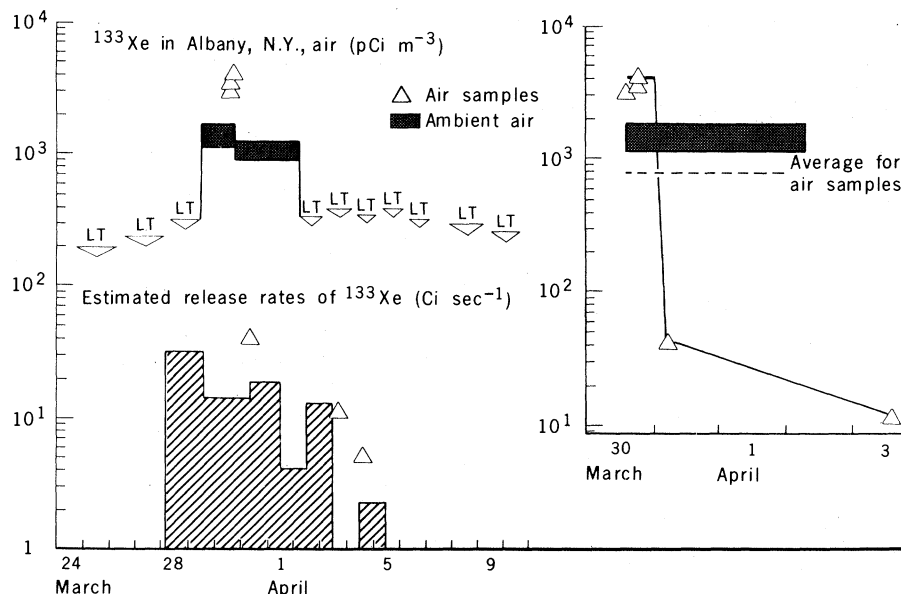


Fig. 1. Xenon-133 activity (picocuries per cubic meter of air) in Albany, New York, for the end of March and early April 1979. The lower trace shows the time-averaged estimates of releases (curies per second) from the Three Mile Island reactor (2). The inset shows detailed values for air samples (gas counting) and concurrent average values for ambient air (Ge diode). Abbreviation: LT, less than.

riod. The trailing edge is more sharply defined from the measurement of the air samples. The time behavior shown in the inset of Fig. 1 from the widely spaced air samples must closely represent the actual trailing edge of the passing air mass. This is evident from a comparison of the calculated time-averaged activity of 780 pCi m<sup>-3</sup> for this period from air samples and the observed averaged activity of 1060 pCi m<sup>-3</sup> from the diode measurement. The observed peak value for the <sup>133</sup>Xe activity at ground level was 3900 pCi m<sup>-3</sup> for the air sample taken late on 30 March. Peak concentrations of <sup>133</sup>Xe for the period before 30 March could have been higher, depending on the actual arrival time of the air containing <sup>133</sup>Xe. The average value from the diode measurement for the 24-hour interval preceding 30 March was 1390 pCi m<sup>-3</sup>.

To describe the air mass transport in more detail, we collated the available data on release rates from the reactor and the regional meteorological conditions. Measured release rates for <sup>133</sup>Xe from the reactor were not available. Average release rates were estimated indirectly by the Nuclear Regulatory Commission, using thermoluminescence dosimeters in the vicinity of the reactor (2). These rates are plotted in Fig. 1. No releases occurred before 0400 EST on 28 March.

Regional meteorological conditions were examined by using forward (from Middletown, Pennsylvania) and backward (from Albany) air trajectories provided by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (3). The backward trajectories were calculated for a mean transport layer between 300 and 1500 m above the terrain. Forward trajectories were calculated for the same mean transport layer and also for transport at heights corresponding to 95, 90, and 85 kPa (10<sup>2</sup> kPa = 1 bar).

For the first release period on 28 March, the meteorological conditions at Middletown were rather stagnant, with medium- to low-speed winds gradually shifting from northwesterly to northeasterly to easterly and finally to southwesterly. From 29 to 31 March, southwesterly winds prevailed at increased speed. The mean transport layer forward trajectories for this period passed 80 to 160 km south of Albany. Backward trajectories for 29 to 31 March show that the mean transport through Albany originated predominantly from regions to the west and northwest of Harrisburg, Pennsylvania. Forward transport at the 95-

kPa level did, however, indicate a plume passage in the Albany area on 29 March.

In summary, the meteorology indicates that air arriving at Albany on 29 March contained radioactive gas released from the Three Mile Island reactor on 28 March, which had been dispersed rather widely around the point of origin and then moved northeastward at low levels. The most probable transit time appears to have been 18 to 24 hours for an approximate actual travel distance of about 500 km.

The whole-body dose to an individual in the Albany area from exposure to  $\gamma$ -rays and x-rays from the passing <sup>133</sup>Xe, calculated from the average activity values, was 0.004 mrem (4). This is about 0.004 percent of the annual whole-body dose from natural sources.

A search for airborne <sup>131</sup>I showed no measurable activity, even though the air was analyzed by a highly sensitive  $\beta/\gamma$  coincidence counting method (5). Albany air processed through a charcoal cartridge impregnated with triethylenediamine during the 24-hour period of highest <sup>133</sup>Xe activity did not contain <sup>131</sup>I at or above  $8 \times 10^{-4}$  pCi m<sup>-3</sup>.

Nor did we observe a measurable increase of <sup>85</sup>Kr in air. This is not surprising, considering the long half-life of <sup>85</sup>Kr (10 years), the lower fission yield, and the sizable atmospheric background concentration from atmospheric weapon testing and routine releases by the nuclear industry. The <sup>85</sup>Kr measured in the sample from 30 March (1500 EST)

amounted to 12.6 pCi m<sup>-3</sup>, which is within the range of 10.9 to 18.4 pCi m<sup>-3</sup> encountered in samples of Albany air for the period 1975 to 1979.

In conclusion, the elevated <sup>133</sup>Xe concentrations observed in Albany on 29 and 30 March 1979 could be attributed to releases from the Three Mile Island reactor accident. The <sup>133</sup>Xe concentrations normally present in Albany air due to routine releases from nuclear reactors are lower by more than three orders of magnitude (1). The dose received from the passing radioactivity was found to be extremely small when compared to the dose from natural sources.

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- 7 August 1979; revised 19 November 1979

## Lead Isotope Identification of Sources of Galena from Some Prehistoric Indian Sites in Ontario, Canada

**Abstract.** *Lead isotopic compositions of 12 galenas from five late Archaic-initial Woodland grave and habitation sites in southern Ontario have made it possible to determine the "most likely" source areas for the galenas. For one of the oldest sites (Finlan), the most likely source is in the southwestern Upper Mississippi Valley (Wisconsin-Illinois-Iowa) mineral district. The seven Finlan galenas exhibit a range of isotopic ratios; three of the largest specimens have substantial isotopic variations (up to 1.8 percent in the ratio of lead-207 to lead-206) on a scale of a few centimeters. This suggests that the lead isotopic zoning ascribed to the Upper Mississippi Valley area is not sufficiently well defined to enable us to determine if all the Finlan samples were derived from a single mineral deposit. Galenas from the other sites (Constance Bay, Hind, Bruce Boyd, and Picton) most probably originated in the southeastern Ontario-northwestern New York area. Isotopic differences among the Constance Bay, Hind, and Bruce Boyd galenas, on the one hand, and the Picton galena, on the other, suggest that at least two distinct sources in that region were exploited.*

Among the objects found at some of the grave and habitation sites of prehistoric North American Indians of the late Archaic and initial Woodland cultures of southern Ontario and the eastern

and central United States are pieces of the mineral galena (PbS). It is not known why the Indians collected this mineral, and why it is found at grave sites, but its ubiquitous distribution suggests that it