

11. J. H. Mazur, R. Gronsky, J. Washburn, *Inst. Phys. Conf. Ser. No. 67* (1983), p. 77.
12. A. Bryant, D. P. E. Smith, C. F. Quate, *Appl. Phys. Lett.* **48**, 832 (1986).
13. G. F. A. van de Walle, H. van Kempen, P. Wyder, P. Davidsson, *ibid.* **50**, 22 (1987).
14. S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981), pp. 513–565.
15. N. Osakabe, Y. Tanishiro, K. Yagi, G. Honjo, *Surf. Sci.* **109**, 353 (1981).
16. T. Hsu, *Ultramicroscopy* **11**, 167 (1983).
17. We thank D. P. E. Smith, M. Richter, and J.

Nogami for their experimental assistance and invaluable comments. A.H.C. acknowledges support and guidance from Philips Research Laboratories Sunnyvale, Signetics Corporation, and in particular helpful discussions with W. T. Stacy. The HRTEM was purchased with funds from Stanford University, the Pew Foundation, and the NSF-MRL program of the National Science Foundation. The STM portion of this work was supported by the Defense Advanced Research Projects Agency.

6 April 1987; accepted 17 June 1987

Radioactive Cesium from the Chernobyl Accident in the Greenland Ice Sheet

C. I. DAVIDSON, J. R. HARRINGTON, M. J. STEPHENSON,
M. C. MONAGHAN, J. PUDYKIEWICZ, W. R. SCHELL

Measurements of cesium-134 and cesium-137 in Greenland snow together with models of long-range transport have been used to assess radionuclide deposition in the Arctic after the Chernobyl accident. The results suggest that a well-defined layer of radioactive cesium is now present in polar glaciers, providing a new reference for estimating snow accumulation rates and dating ice core samples.

NUCLEAR WEAPONS TESTS IN THE 1950s, 1960s, and 1970s emitted large amounts of man-made radioisotopes into the atmosphere. Some of this material eventually reached glaciers in the Arctic and Antarctic regions, providing a permanent record of the deposition of radioactivity associated with specific tests. These radioactive signatures have been used to determine snow accumulation rates, to date ice core samples analyzed for other contaminants, and to study long-range atmospheric transport (1). Emissions from the explosion and fire at the Chernobyl nuclear reactor in April 1986 have resulted in an additional radioactive layer in polar glaciers. This layer is of considerable interest: unlike weapons tests, which injected radioactive material into the stratosphere where residence times are more than a year, the Chernobyl emissions were confined to the troposphere where residence times are at most a few weeks (2). The resulting deposition thus occurred over a relatively short period, and this enables us to assign a narrow time interval to the radioactive layer. In the present study, we identify the Chernobyl signature in the Greenland Ice Sheet. We also attempt to relate characteristics of the deposited radioactivity to the atmospheric transport pathways and deposition processes involved.

Samples were collected from a snowpit 23 km southwest of Dye 3, Greenland, in late July 1986. This location is near the ice coring site established by Mayewski *et al.* (3). The snowpit walls were sampled in

continuous adjacent layers to a depth of 1.5 m under strict contamination control (3–5). The density was measured in each 5-cm layer, and the presence of ice strata and other distinguishing characteristics was recorded. Samples were collected each 5 cm for $\delta^{18}\text{O}$ analysis (6), and each 10 cm for radioactive cesium analysis (7).

Results of these analyses show that detectable levels of ^{134}Cs and ^{137}Cs occurred only in one layer, between 10 and 20 cm below the surface. The concentrations in three identical samples extracted from this layer are 2.0 ± 0.8 pCi/liter for ^{134}Cs and 6.2 ± 1.4 pCi/liter for ^{137}Cs (average \pm standard deviation). If we take into account the thickness of the layer and the density of the snow, these values correspond to total (wet plus dry) deposits of 0.072 ± 0.030 mCi/km² for ^{134}Cs and 0.22 ± 0.05 mCi/km² for ^{137}Cs . All of the data have been corrected to 1 May.

These deposition rates are much smaller than corresponding values measured in Europe shortly after the accident. For example, deposition onto soil and vegetation was highly variable throughout Scandinavia, Germany, the United Kingdom, and the Mediterranean region, with values for both radionuclides ranging from <1 to several thousand millicuries per square kilometer (2, 8). Deposition rates to bulk wet-dry collectors in North America were closer to, but still somewhat greater than, the Dye 3 snow values. Seven sites in the Canadian Arctic had total deposition rates averaging 0.3 mCi/km² for ^{134}Cs and 0.7 mCi/km² for

^{137}Cs during May (9). Total deposition rates of ^{134}Cs at sites in the western United States were typically 1 to 2 mCi/km², with Midwest and East Coast locations reporting 0.1 to 1 mCi/km² (10, 11). Values for ^{137}Cs in the United States were generally two to three times as large as those for ^{134}Cs , consistent with the $^{137}\text{Cs}/^{134}\text{Cs}$ activity ratio of 3.0 ± 0.24 observed in Greenland. A single sample of the 10- to 20-cm layer was also analyzed for ^{90}Sr , giving $^{137}\text{Cs}/^{90}\text{Sr}$ equal to 20.2 corrected to 1 May (12). In comparison, values of this ratio in accumulated deposition on soil and vegetation were 19.0 in Denmark (13) and 24.4 in Italy (14).

Figure 1 shows the calculated dispersion of ^{137}Cs from Chernobyl during late April and early May, based on an Eulerian long-range transport model. The simulation was developed by Pudykiewicz specifically for assessing atmospheric transport of emissions from the accident (15). This figure indicates that a portion of the radioactive cloud crossed Greenland near the end of April. The cloud continued moving south and west, reaching Canada and eventually the United States in early May. This scenario is consistent with available data: airborne measurements show that ^{134}Cs and ^{137}Cs from Chernobyl first reached the community of Alert in the northeastern Canadian Arctic on 1 May, the area north of Hudson Bay on 2 May, and sites in Alaska, southern Canada, and continental United States sometime between 6 and 10 May (9, 11). Furthermore, comparisons between results of the model and measured ground-level concentrations at several locations in Canada show good quantitative agreement (15). The dispersion patterns in Fig. 1 are similar to those calculated by Lawrence Livermore National Laboratory with a different simulation technique (16).

How did this atmospheric material reach the surface of the Greenland Ice Sheet? To explore this question, the $\delta^{18}\text{O}$ data from the snowpit have been compared with the meteorological records from Dye 3 to identify the most probable time period and the specific storms represented by the 10- to 20-cm layer. The method has been discussed elsewhere (5). The procedure indicated that this layer corresponds to a time interval containing the following storms (with accumulation rates in centimeters of snow): 9

C. I. Davidson, J. R. Harrington, M. J. Stephenson, Departments of Civil Engineering and Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA 15213.

M. C. Monaghan, Department of the Geophysical Sciences, University of Chicago, Chicago, IL 60637.

J. Pudykiewicz, Environment Canada, 2121 Trans-Canada Highway, Dorval, QU H9P 1J3 Canada.

W. R. Schell, Graduate School of Public Health, University of Pittsburgh, Pittsburgh, PA 15261.

April (1.9 cm), 18 April (0.3 cm), 21 May (0.6 cm), 5 June (3.5 cm), and 7 June (3.2 cm).

It is noteworthy that very little precipitation fell for nearly 6 weeks after the accident. By the first significant snowfall on 5 June, airborne concentrations of ^{134}Cs and ^{137}Cs had already peaked and were declining throughout most of North America. If we assume that airborne concentrations at Dye 3 during May were similar to those measured in the Canadian Arctic and eastern

provinces (9), dry deposition can account for roughly 25 to 50% of the observed ^{134}Cs and ^{137}Cs content of the snow. This estimate assumes a dry deposition velocity in the range 0.05 to 0.10 cm/sec, which is based on deposition data for submicrometer aerosol species such as sulfate and lead onto a snow surface (4, 5, 17). Cesium-134 and cesium-137 emitted from Chernobyl have activity median aerodynamic diameters of 0.2 to 0.7 μm (18), similar to those of sulfate and lead (19). It is therefore likely that a fraction of the measured radioactive cesium in Dye 3 snow is due to dry deposition, which has resulted in a narrow, concentrated layer. The small cesium deposition rates at Dye 3 relative to other North American sites reflect the very small precipitation rates on the ice sheet during May and June. The total amount of radioactive cesium reaching the earth's surface will generally be greatest in areas of high precipitation, since wet deposition is a far more efficient removal mechanism than dry deposition.

Wet deposition during the 21 May, 5 June, and 7 June storms also probably influenced concentrations in the snow. Joshi (20) reported that southern Ontario rainwater contained 0.6 to 3 pCi/liter of ^{134}Cs and 1.5 to 5 pCi/liter of ^{137}Cs during May and June. Other values similar to the Dye 3 data have been reported for precipitation in the continental United States (10, 11).

Overall, the measured airborne concentrations from several monitoring programs and results of the transport model indicate that the radioactive cloud from Chernobyl spread rather uniformly across North America in the weeks after the accident. Considered in light of the Greenland snow data, these results suggest that an identifiable signature is now present in glaciers throughout the Arctic.

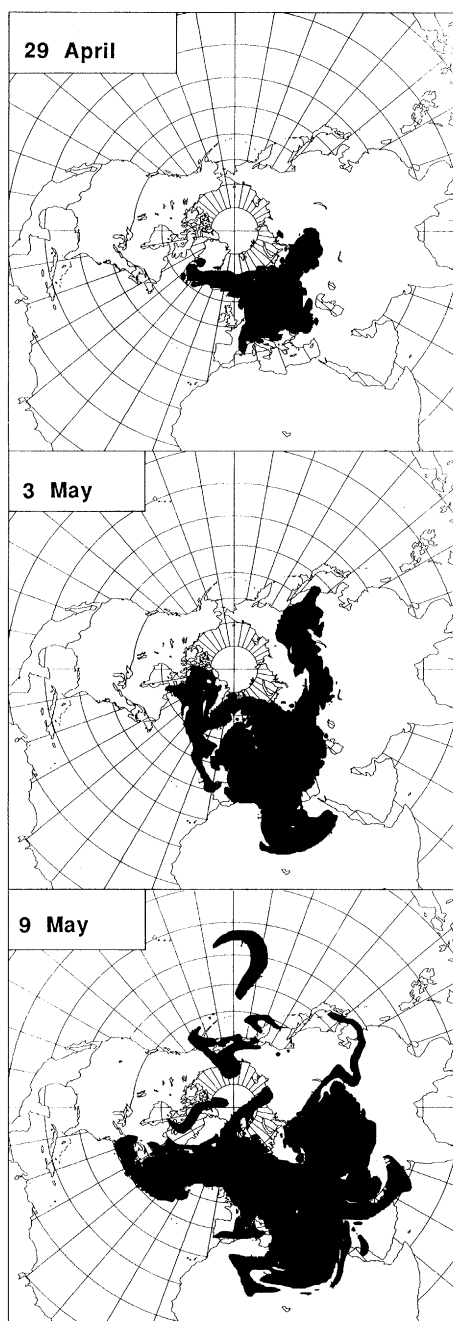


Fig. 1. Spread of ^{137}Cs emitted from Chernobyl as of the indicated date in 1986. Winds at 850 mbar have been used as inputs. The shaded areas indicate calculated airborne concentrations greater than 3 fCi/m³, based on estimates of the source strength.

REFERENCES AND NOTES

1. M. Koide and E. D. Goldberg, in *Greenland Ice Core: Geophysics, Geochemistry, and the Environment*, C. C. Langway, Jr., H. Oeschger, W. Dansgaard, Eds. (American Geophysical Union, Washington, DC, 1985), pp. 95–100.
2. C. Hohenemser *et al.*, *Environment* **28**, 6 (1986).
3. P. A. Mayewski *et al.*, *Science* **232**, 975 (1986); *Atmos. Environ.* **21**, 863 (1987).
4. C. I. Davidson, S. Santhanam, R. C. Fortmann, M. P. Olson, *Atmos. Environ.* **19**, 2065 (1985).
5. C. I. Davidson *et al.*, *ibid.* **21**, 871 (1987).
6. The $\delta^{18}\text{O}$ analyses by gas-source mass spectrometry were conducted at the Geophysical Isotope Laboratory, Copenhagen, Denmark [W. Dansgaard, S. J. Johnsen, H. B. Clausen, N. Gundestrup, *Med. Greenland* **187**, 1 (1973)].
7. Analyses for ^{134}Cs and ^{137}Cs by intrinsic germanium gamma-ray detection were conducted at the University of Chicago. Samples from the first four 10-cm layers were analyzed individually. Samples below 40 cm were pooled and analyzed in aggregate.
8. P. W. Krey *et al.*, in *Report EML-460* (Environmental Measurements Laboratory, U.S. Department of Energy, New York, 1986), pp. 155–213; "Updated summaries of data received in Europe re: Chernobyl reactor accident" (World Health Organization, Regional Office for Europe, 13 June 1986).
9. Monitoring data were provided by the Bureau of Radiation and Medical Devices, Health and Welfare Canada. Data for the Arctic regions include airborne concentrations for Alert, Fort Churchill, Hay River, Resolute, and Whitehorse, and bulk wet-dry deposition rates for Coral Harbor, Goose Bay, Hay River, Inuvik, Resolute, Whitehorse, and Yellowknife. Airborne concentration data were also obtained for sites further south in Newfoundland, Nova Scotia, New Brunswick, Quebec, and Ontario.
10. Z. R. Juzdan *et al.*, in *Report EML-460* (Environmental Measurements Laboratory, U.S. Department of Energy, New York, 1986), pp. 105–154.
11. R. J. Larsen, C. G. Sanderson, W. Rivera, M. Zamicheli, in *Report EML-460* (Environmental Measurements Laboratory, U.S. Department of Energy, New York, 1986), pp. 1–104.
12. Analysis for ^{90}Sr was conducted at the University of Pittsburgh using Environmental Protection Agency method 905.0 ["Prescribed procedures for measurement of radioactivity in drinking water," *Report EPA-600/4-80-032* (U.S. Environmental Protection Agency, Washington, DC, 1980)].
13. A. Aarkrog, "Preliminary conclusions to be drawn from the studies of Chernobyl debris in Denmark," paper presented at the Seminar on the Cycling of Long-Lived Radionuclides in the Biosphere: Observations and Models, Commission of the European Communities and Junta de Energia Nuclear, Madrid, Spain, 15 to 19 September 1986.
14. A. Ceroni, *Not. Enea Energia Innovazione* **32**, 65 (1986).
15. J. Pudykiewicz, "Numerical simulation of the transport of radioactive cloud from the Chernobyl nuclear accident" (Environment Canada, Dorval, Quebec, 1987). The model was run with wind data at pressure levels of 1000, 850, 700, 500, and 300 mbar from the Canadian Meteorological Centre. On the basis of these results, a more comprehensive model was developed and run at 850 and 700 mbar; the 850-mbar simulation showed better agreement with measured concentrations. Note that the model is used only as a rough indication of transport over Greenland, since atmospheric flows are complicated by the elevation of the ice sheet (790 mbar at Dye 3).
16. M. H. Dickerson and T. J. Sullivan, "ARAC response to the Chernobyl reactor accident," *Report UCID-20834* (Lawrence Livermore National Laboratory, Livermore, CA, 1986); P. H. Gudiksen and R. Lange, "Atmospheric dispersion modeling of radioactivity releases from the Chernobyl event," *Report UCRL-95363* (Lawrence Livermore National Laboratory, Livermore, CA, 1986).
17. H. Dovland and A. Eliassen, *Atmos. Environ.* **10**, 783 (1976); M. Ibrahim, L. A. Barrie, F. Fanaki, *ibid.* **17**, 781 (1983).
18. E. A. Bondietti and J. N. Brantley, *Nature (London)* **322**, 313 (1986); R. H. Knuth and C. G. Sanderson, in *Report EML-460* (Environmental Measurements Laboratory, U.S. Department of Energy, New York, 1986), pp. 291–300.
19. J. B. Milford, C. I. Davidson, *J. Air Pollut. Control Assoc.* **35**, 1249 (1985); *ibid.* **37**, 125 (1987).
20. S. R. Joshi, "Chernobyl radioactivity in Canada: Characteristics and transport," paper presented at the Seminar on the Cycling of Long-Lived Radionuclides in the Biosphere: Observations and Models, Commission of the European Communities and Junta de Energia Nuclear, Madrid, Spain, 15 to 19 September 1986.
21. We thank W. Dansgaard, and N. Gundestrup for conducting the $\delta^{18}\text{O}$ analyses; K. Kuivinen, S. Watson, K. Swanson, R. Tillson, and J. Klinck for assistance in the field; J. Cragin for providing sampling equipment; L. A. Barrie, B. L. Tracy, D. Meyerhof, P. E. Cobbold, S. R. Joshi, and Z. R. Juzdan for providing radionuclide data from other monitoring programs; and R. S. Tsay and M. J. Small for help in data interpretation. The manuscript was prepared by S. A. Knapp. This work was supported by National Science Foundation grants DPP-8315452 and DPP-8618223, a scholarship from the Richard K. Mellon Foundation, and a grant from the Louis Block Fund of the University of Chicago.

24 February 1987; accepted 8 June 1987