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^{-3} for this period from air samples and the observed averaged activity of 1060 pCi m^{-3} from the diode measurement. The observed peak value for the ^{133}Xe activity at ground level was 3900 pCi m^{-3} for the air sample taken late on 30 March. Peak concentrations of ^{133}Xe for the period before 30 March could have been higher, depending on the actual arrival time of the air containing ^{133}Xe. The average value from the diode measurement for the 24-hour interval preceding 30 March was 1390 pCi m^{-3}.

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 ^{133}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 (10^2 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 southeasterly. 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 γ-rays and x-rays from the passing ^{133}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 ^{131}I showed no measurable activity, even though the air was analyzed by a highly sensitive β/γ coincidence counting method (*5*). Albany air processed through a charcoal cartridge impregnated with triethylenediamine during the 24-hour period of highest ^{133}Xe activity did not contain ^{131}I at or above 8×10^{-4} pCi m^{-3}.

Nor did we observe a measurable increase of ^{85}Kr in air. This is not surprising, considering the long half-life of ^{85}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 ^{85}Kr measured in the sample from 30 March (1500 EST) amounted to 12.6 pCi m^{-3}, which is within the range of 10.9 to 18.4 pCi m^{-3} encountered in samples of Albany air for the period 1975 to 1979.

In conclusion, the elevated ^{133}Xe concentrations observed in Albany on 29 and 30 March 1979 could be attributed to releases from the Three Mile Island reactor accident. The ^{133}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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References and Notes

1. C. O. Kunz, in *Noble Gases* (CONF-730915, ERDA, 1973), pp. 209–217; C. J. Paperiello, in *ibid.*, pp. 239–248; C. O. Kunz and C. J. Paperiello, *Science* **192**, 1235 (1974).
2. L. Barret, Nuclear Regulatory Commission, Washington, D.C., personal communication.
3. L. Machta, K. Telegadas, and J. Heffter from the Air Resources Laboratory of the National Oceanic and Atmospheric Administration, Silver Spring, Md., provided us with forward and backward trajectory calculations for the entire period of interest.
4. U.S. Atomic Energy Commission, *Regulatory Guide 1.4: Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Pressurized Water Reactors* (revision 2, June 1974).
5. C. J. Paperiello and J. M. Matuszek, *IEEE Trans. Nucl. Sci.* **22**, 642 (1975).

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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 had some substantial significance for the Indian cultures of the time. Like many of the archeological objects found at burial and habitation sites (for example, shells, stones, and native copper implements), galena specimens were probably traded by hand-to-hand transactions and may ultimately have reached sites considerably distant from the mineral sources. We present data which suggest that some galenas were traded over long distances but that others were obtained much more locally. The observations are based on precise isotopic analyses of lead in galenas from four burial sites and one occupation site in southern Ontario.

Lead isotope "fingerprinting" of the sources of lead in glass and metallic artifacts has been applied with success to the identification of sources of objects manufactured by Old World cultures (*1*) but has not, to our knowledge, been applied in a North American context. The variations in the isotopic composition of lead in nature are the result of geological processes which operated before and during the formation of the minerals containing the lead; only by mixing with lead of different isotopic constitution can the composition of a lead sample be altered. The ability to use lead isotope measurements to identify possible sources of galena depends on two factors: (i) accurate analyses combined with an extensive base of isotopic data for potential natural galena mineralizations and (ii) significantly different sets of isotopic compositions among those mineralizations.

For the mineralized areas which might have been accessible to the North American Indians who collected galena, there is an extensive lead isotopic data base (*2-4*), within which the data fields occupied by isotopic ratio values from different areas are reasonably well defined and overlap only slightly (Fig. 1). In Fig. 1, we follow the custom of earlier investigators in plotting $^{207}Pb/^{206}Pb$ against $^{208}Pb/^{206}Pb$. Other ratios such as $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ can be plotted (and can be useful in eliminating ambiguities), but the former pair appears to be adequately selective for the present work (*5*).

The five burial and habitation sites from which galena specimens were obtained and the corresponding lead isotope ratios (*6*) are listed in Table 1. The isotope data have also been plotted in Fig. 1, and it is clear by inspection of this figure that correlation with specific source fields is defined adequately (*5*). The samples from the Finlan site have evidently come from the Wisconsin-Illinois-Iowa (Upper Mississippi Valley) lead-zinc mineralization area, whereas the galenas from the Constance Bay, Hind, Bruce Boyd, and Picton sites are from a mineralized zone (or zones) in the southeastern Ontario–northwestern New York area.

The data of Heyl *et al.* (*2*) suggest that the $^{207}Pb/^{206}Pb$ ratio varies in a regular fashion across the Upper Mississippi Valley lead-zinc mineralization district

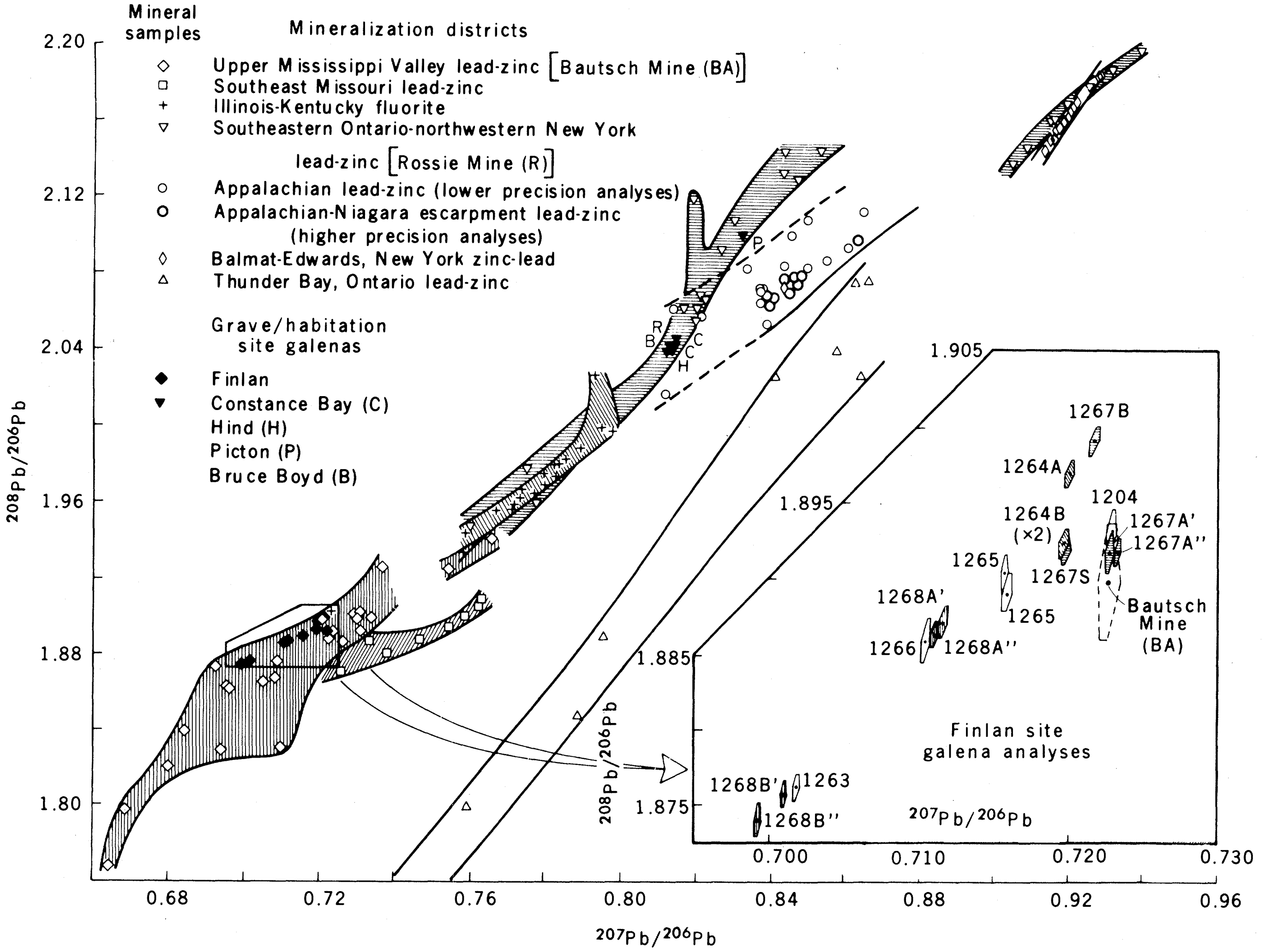

Fig. 1. Lead isotopic data for some North American lead-zinc mineral deposits which might have been accessible to prehistoric Indians. Data points for galena from grave and habitation sites in southern Ontario are also shown; ratios for the Finlan site are shown at the lower right on an enlarged scale.

(Fig. 2). Thus, one can define more closely the boundaries of the region from which the galenas were obtained. On the basis of the observed spread in ^{207}Pb/^{206}Pb in the Finlan galenas, the most likely source area has been outlined in Fig. 2 (approximately 15 percent of the total mineralized region). Mapping the values of ^{208}Pb/^{206}Pb does not define precisely the same area, but the boundaries common to both plots are close to those outlined in Fig. 2. As a check on the compatibility of our data with those of Heyl *et al.*, we have reanalyzed their sample from the Bautsch Mine in the southwest corner of the area and obtained good agreement with their results. The isotopic ratios for this sample are very similar to those of several of the Finlan galenas.

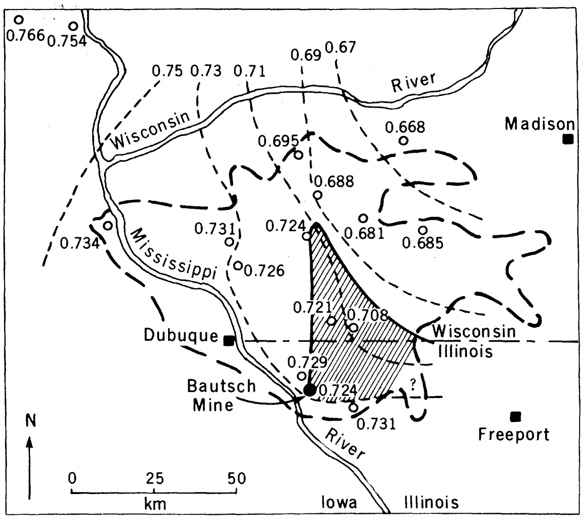

Fig. 2. The Upper Mississippi Valley (Wisconsin-Illinois-Iowa) lead-zinc mineralization district. Numbers adjoining open circles are ^{207}Pb/^{206}Pb ratios at sample sites quoted in Heyl *et al.* (*2*). Dashed lines represent ^{207}Pb/^{206}Pb contours and suggest an isotopic zoning across the area. The cross-hatched zone within the contours from 0.69 to 0.73 is the area from which the Finlan galenas are most likely to have come.

Table 1. Lead isotope data for the galena specimens.

Sample number*	^{206}Pb/^{204}Pb	^{207}Pb/^{206}Pb	^{208}Pb/^{206}Pb
Finland, 44°07′N, 77°37′W (late Archaic)			
1268A′	22.55 ± 0.03	0.7117 ± 0.0004	1.8872 ± 0.0011
1268A″	22.57 ± .02	.7111 ± .0002	1.8863 ± .0009
1268B′	22.94 ± .02	.7010 ± .0002	1.8757 ± .0009
1268B″	22.99 ± .02	.6993 ± .0002	1.8740 ± .0011
1263	22.90 ± .03	.7019 ± .0002	1.8761 ± .0009
1266	22.59 ± .01	.7106 ± .0003	1.8859 ± .0015
1265	22.38 ± .03	.7160 ± .0004	1.8891 ± .0013
1265†	22.39 ± .03	.7159 ± .0002	1.8905 ± .0011
1264A	22.28 ± .04	.7202 ± .0003	1.8969 ± .0009
1264B	22.25 ± .02	.7197 ± .0003	1.8924 ± .0009
1264B†	22.26 ± .02	.7198 ± .0003	1.8924 ± .0009
1204	22.14 ± .03	.7229 ± .0004	1.8927 ± .0019
1267A′	22.17 ± .02	.7229 ± .0003	1.8917 ± .0013
1267A″	22.15 ± .02	.7234 ± .0002	1.8918 ± .0009
1267B	22.20 ± .02	.7219 ± .0004	1.8891 ± .0010
1267S‡	22.26 ± .01	.7199 ± .0003	1.8919 ± .0009
Bautsch Mine, Wisconsin§			
1208	22.18 ± .05	.7228 ± .0007	1.8898 ± .0038
8 (*2*)	22.33	.7241	1.893
Constance Bay, 45°29′N, 76°04′W (490 ± 75 B.C.)			
1205A	19.21 ± .02	.8138 ± .0008	2.0411 ± .0020
1205B	19.20 ± .01	.8142 ± .0003	2.0414 ± .0010
1206A	19.18 ± .02	.8153 ± .0003	2.0459 ± .0010
1206B	19.18 ± .02	.8152 ± .0004	2.0456 ± .0014
Hind, 42°32′N, 81°35′W (900 to 1000 B.C.)			
1211	19.23 ± .01	.8131 ± .0003	2.0409 ± .0020
Picton, 43°59′N, 77°10′W (late Archaic)			
1210A	18.73 ± .01	.8333 ± .0003	2.0899 ± .0010
1210B	18.73 ± .01	.8323 ± .0003	2.0861 ± .0010
Bruce Boyd, 42°36′N, 80°28′W (520 ± 65 B.C.)			
1270A	19.24 ± .01	.8126 ± .0003	2.0405 ± .0010
1270B	19.24 ± .01	.8128 ± .0003	2.0412 ± .0010
Rossie Mine, New York§			
1242	19.23 ± .02	.8127 ± .0004	2.0389 ± .0012

*The four-figure number identifies a single galena specimen; letters designate distinct sample chips from opposite sides of the specimen; primed letters identify distinct sample chips located within 1 or 2 mm of each other. †Repeat mass spectrometric analysis of the same sample chip. ‡Lead carbonate (?) surface coating. §Mineral occurrence.

Heyl *et al.* mapped the isotopic zoning of the Upper Mississippi Valley region on the basis of 17 data points. They also analyzed the surface and the interior of a cluster of galena crystals and obtained a 0.07 percent difference in ^{207}Pb/^{206}Pb between the samples. Studies of lead-zinc minerals from other areas (*4, 7*) have revealed that there may be substantial differences in isotopic composition over distances as small as a few millimeters; as a check on this possibility, we selected three of the largest Finlan galenas (samples 1264, 1267, and 1268, each approximately 25 mm in length) and analyzed chips from the ends opposite those originally sampled and also analyzed the lead carbonate alteration on the surface of one of these samples (1267S) (see Table 1 and Fig. 1). In two of the samples (1264 and 1267) there are isotopic differences of the type observed by Heyl *et al.*, mainly in the ^{208}Pb content of the samples. The lead carbonate alteration in sample 1267S has a significantly lower ^{207}Pb/^{206}Pb ratio than PbS from either end of the galena and is similar in isotopic composition to PbS from one end of sample 1264. Chips from opposite ends of sample 1268, 2 cm apart, differ in ^{207}Pb/^{206}Pb by about 1.8 percent. Obviously, the distribution suggested in Fig. 2 can serve as only a rough guide to the location of samples. Although it is tempting to conclude that the Finlan galenas came from more than one mineral outcrop, a much more thorough investigation of lead isotope variations in the Upper Mississippi Valley will have to be made before this can be decided.

For the sites at which "local" galena sources are represented, the samples from the Hind site and the Bruce Boyd site are identical in isotopic composition with a galena from the Rossie Mine, one of a set of lead-zinc vein deposits in New York about 10 km southeast of the St. Lawrence River. The two specimens from the Constance Bay site are slightly different in composition from each other and from the Rossie Mine galena. Since we have made no detailed study of lead isotope variations in the Rossie district, we can only say that veins in this area would be prime candidates for the sources of the Constance Bay, Hind, and Bruce Boyd galenas. The Constance Bay site differs in age from the Hind site by about 500 years; perhaps the galena source was known and repeatedly exploited over this interval of time. The lead isotope ratios in the galena from the

Picton site have no near-equivalent in our data base, and no estimate of a more accurate location of the source of this galena can be made. The lead isotopic composition of the many small lead-zinc mineral prospects in northwestern New York–southeastern Ontario are variable and possibly distinctive. Further sampling of these minerals might reduce the uncertainty.

Our data demonstrate conclusively that lead isotope measurements can serve to identify the mineralization districts at which Indian burial and habitation site galenas have originated. For those regions for which a sufficient density of lead isotope data enables one to define isotopic zoning, the boundaries of possible source regions can be narrowed.

The data currently available for grave and habitation sites in southern Ontario indicate that at the time of the late Archaic culture some samples of galena were reaching the area from the Upper Mississippi Valley, 1100 km away. The lead isotopic differences that we observe among the Finlan site galenas suggest that more than one mineral deposit in that region was being exploited for galena. Indians of the region were aware of the existence of the lead-zinc mineralization there at the time when the area was first explored by Europeans (*8*), and it is possible that the locations of many mineral outcrops were known in much earlier times. Galena sources closer to the Ontario sites were also exploited. Because of the small number of sites for which we presently have isotopic data, it is not possible to discern any temporal change in galena sources, but both local and distant sources existed and they are clearly distinguishable on the basis of this technique. Further lead isotope measurements may therefore provide us with a means of investigating possible short- and long-term changes in trading patterns, if galenas can be obtained from a sufficiently large number of dated archeological sites.

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References and Notes

1. I. L. Barnes, W. R. Shields, T. J. Murphy, R. H. Brill, *Adv. Chem. Ser. 138* (1974), p. 1; R. H. Brill and W. R. Shields, *R. Numismatic Soc. London. Spec. Publ. No. 8* (1972), p. 279.
2. A. V. Heyl, M. H. Delevaux, R. E. Zartman, M. R. Brock, *Econ. Geol.* **61**, 933 (1966).
3. B. R. Doe, *Lead Isotope Data Bank, Lead Isotopes and Ore Deposits Project* (U.S. Geological Survey, Denver, 1975).
4. I. R. Fletcher, thesis, University of Toronto (1979).
5. Most of the data fields in Fig. 1 are adequately defined by relatively recent lead isotope ratios determined to better than ± 0.15 percent (2σ, where σ is the standard deviation). Only five data points are available for samples from Appalachian mineral occurrences (*3*). To obtain a geographically broader set of data, early isotopic analyses of lower precision (± 0.1 to 0.8 percent) have been included, and some expansion of the data field for this area has undoubtedly occurred. The high-precision data suggest that, when additional measurements for lead minerals from the area are available, the improved definition of the data field will enable us to unambiguously distinguish most Appalachian lead minerals from those in the southeastern Ontario–northwestern New York area.
6. We obtained the isotopic data by using direction-focusing mass spectrometer (15-cm radius of curvature) calibrated for lead mass discrimination by repeated analyses of National Bureau of Standards common lead standard SRM-981 or by double spiking. Uncertainties quoted for the isotopic ratios for each sample are approximately 95 percent confidence limits and include the uncertainties within analyses and in the mass fractionation correction. The lead contamination introduced in sample preparation is 0.2 ng per microgram of sample lead.
7. C. F. Austin and W. F. Slawson, *Am. Mineral.* **46**, 1132 (1961); R. S. Cannon, A. P. Pierce, J. C. Antweiler, K. L. Buck, *Econ. Geol.* **56**, 1 (1961).
8. R. G. Thwaites, *Wis. Hist. Collect.* **13**, 271 (1895).
9. We thank B. R. Doe for the Bautsch Mine sample, M. W. Spence for the Bruce Boyd galena, J. V. Wright for the remaining samples and for helpful discussion and guidance, and P. R. Kuybida for sample preparations. This research was supported by a grant from the National Research Council of Canada.

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Benjamin Franklin and Timothy Folger's First Printed Chart of the Gulf Stream

Abstract. *A print of the Benjamin Franklin and Timothy Folger 1769–1770 chart of the Gulf Stream, all copies of which have been "lost" for nearly 200 years, was found in the Bibliothèque Nationale in Paris. This is the first chart of the Gulf Stream and continues today to be a good summary of its strength, course, and breadth.*

Benjamin Franklin had printed in about 1769–1770 the first chart of the Gulf Stream in order to help the captains of the British packets avoid this swift current and to speed their passage to New York (*1–3*). Partly because the British captains slighted it and partly because of Franklin's role in the confrontation between the American colonies and England, this chart became very rare. Despite considerable effort by many people to find a copy of the Franklin-Folger chart in England and America, not one has been found until now. The chart that most people associate with Franklin, the 1786 version (*4*), was published nearly two decades after the first chart and is really a copy of a copy of it. What may seem surprising is that until now the oldest existing chart of the Gulf Stream was not due to Franklin at all but was one published by William Gerard DeBrahm in 1772 (*5*).

The Franklin-Folger chart is important for several reasons. First it is the first good chart of the Gulf Stream. Before 1768, charts showed only the most rudimentary pictures of currents; only much later were chronometers used to determine ship drift velocities and maps of the surface currents. Second, the chart is a summary of the Nantucket whalers' knowledge of the Gulf Stream. These seamen frequently hunted whales along the edges of the Gulf Stream and learned a lot about its speed, course, and breadth. Third, the Franklin-Folger chart remains today a good summary of the mean path and width of the Gulf Stream and the speeds in its high-velocity core. The Gulf Stream is a large and complex current system that fluctuates energetically in space and time. Even today, the system is difficult to measure and interpret; the measurements that we have agree with the Franklin-Folger chart (*6*).

The creation of the first Gulf Stream chart has been described in Franklin's own hand (*2, 4*). While he was in London as Deputy Postmaster General for the American colonies, Franklin was consulted on the question of why the mail packets took a fortnight longer to sail to America than the merchant ships. In October 1768 Franklin discussed this problem with his cousin Timothy Folger, a Nantucket ship captain then visiting London. Folger told him the packet captains were ignorant of the Gulf Stream and frequently sailed in this current, stemming it. Folger sketched the Gulf Stream on a chart and added written notes on how to avoid the Gulf Stream, and Franklin had the chart printed in 1769 or 1770. However, the British captains slighted the chart, probably because they did not appreciate the implication that American fishermen knew more about ocean currents than the British did. During the early 1770's, the colonies began to revolt against England and Franklin may have suppressed the chart to keep it out of the hands of the British Navy.

In September 1978, I found two prints of the Franklin-Folger chart in the Bibliothèque Nationale in Paris (Fig. 1) (*7*). It

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