Stable Carbon and oxygen isotope source tracing of marble sculptures in the Museum of Fine Arts, Boston and The Sackler Museum, Harvard

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Abstract

We have attempted to determine the quarry sources of eighty-three Greek and Roman sculptures in the Museum of Fine Arts, Boston (sixty-seven) and the Arthur M. Sackler Museum, Harvard University (sixteen) by means of stable carbon and oxygen isotope ratio measurements, coupled with mineralogical examination and X-ray diffractometry. When only two variables are used (carbon and oxygen isotopes), only two of the sculptures can be unequivocally assigned to one quarry source in the isotope database. Identification of the marble as dolomitic or fine-grained, white calcite raises the unequivocal source identifications to thirty-four (41%); contextual considerations bring the "probable single source" count to 72%. It is suggested that a quarry database of strontium isotope ratios (and/or other heavy isotopes), when combined with carbon and oxygen ratios, would make source identification reliable in the 80-90% range.

Introduction

This paper reports work in progress to determine the quarry sources of marble sculptures in the Museum of Fine Arts, Boston (MFA) and the Arthur M Sackler Museum, Harvard University. It results from a long-term collaboration, originally suggested by David Mitten of Harvard, of a group of researchers who include archaeology, art history, archaeological science, and geochemistry among their interests. We have measured the ratios of stable carbon and oxygen isotopes in eighty-three Greek and Roman sculptures in the Museum of Fine Arts (sixty-seven) and the Sackler Museum (sixteen) and compared the results with known isotopic data on marble sources of the Mediterranean. Additional analyses included mineralogical observation and X-ray diffractometry. These data allow us to evaluate the efficacy of using only two variables for source determination (carbon and oxygen isotope ratios), as opposed to using three or more. They also allow us to suggest an approach to the problem of source identification which minimizes cost and sample size, while raising the potential success rate of unequivocal attribution to at least 80-90%.

Methods and results

Sampling

Powder samples were removed from fresh breaks on the marble statues by scraping with the blade of a Swiss army knife, catching the grains on smooth weighing paper and transferring them to a small pill vial. When no fresh surface was exposed, the weathered crust was first removed and discarded; for fourteen of the eighty-three sculptures, samples of the weathered crust were also taken for isotopic analysis.

The sample requirement for X-ray diffractometry is one to two milligrams and for isotopic analysis (both carbon and oxygen) it is the same. A sample of ten milligrams is, therefore, ample to allow for repeated analyses, if they should prove necessary. This is such a small amount that the sampling site on the sculpture can be located only if one knows exactly where to look. The procedure is essentially non-destructive.

Isotopic analysis

Carbon and oxygen stable isotope ratios ($\delta^{13}C/^{12}C$ and $\delta^{18}O/^{16}O$) were measured in a VG PRISM II mass spectrometer located in the Department of Earth & Planetary Sciences at Harvard. This instrument is equipped with a carbonate autosampler, using a common acid bath. No sample preparation is necessary: one to two milligrams of marble powder is weighed out and placed in an appropriate sample cup, which is fitted into a numbered hole in a fifty-cup carousel. The carousel is pumped to vacuum, whereupon the samples are dropped one by one into a stirred bath of phosphoric acid at 90°C. The marble decomposes in the acid, releasing CO_2 gas and a small amount of water vapour. The CO_2 gas is cryogenically dried and purified before injection into the ion source of the mass spectrometer. Its carbon and oxygen isotope ratios are measured simultaneously by comparison with a standard CO_2 gas of known isotopic composition; the results are provided in delta notation ($\delta^{13}C$ and $\delta^{18}O$) relative to the PDB marine limestone standard.

Calcitic marble is essentially pure calcium carbonate, which dissolves very rapidly in phosphoric acid. Allowing five minutes for decomposition, before the next sample is dropped into the common acid bath, proved to be ample. Total analysis time for calcitic marble is about ten minutes. Dolomitic marble, however, which is a calcium magnesium carbonate, dissolves more slowly and can produce memory effects in the common acid bath. Ten minutes were allowed for the decomposition of dolomitic samples. To control for systematic errors and machine drift, samples of a limestone house standard of known isotopic composition were interspersed with the marble samples in the carousel, at intervals of one or more standard samples for every ten unknowns. The isotopic composition of the limestone standard has been repeatedly measured in our laboratory. For further quality control, it has also been measured in the Archaeometry Laboratory of the University of Cape Town, where CO_2 is prepared by hand and measurement is done in a VG602E mass spectrometer.

The precision of our measurement on the limestone standard is 0.1‰ for carbon isotopes and 0.2‰ for oxygen isotopes. This precision is taken into account when comparing the isotopic ratios of sculptures with those of quarries.

Given a functioning laboratory of the type described here, maintained by skilled technical personnel, the measurement of carbon and oxygen isotope ratios in marble is actually very simple. Most of the measurements reported here were done by Harvard freshmen as class exercises in a seminar on archaeological science. This helped to keep down costs to about US$10 per pair of mass spectrometer measurements (carbon and oxygen) at 1993 student prices. For other users the cost is US$20 per pair of measurements.

Results

The results of our isotopic measurements on the marble sculptures are provided in Table 1. For purposes of attribution, they were compared with quarry data produced by Herz (1992, with new additions), Matthews *et al* (1992), and Moens *et al* (1992). When the isotopic data for all known quarries are considered, unequivocal quarry attributions can be made for only two of the eighty- three sculptures analysed.

Using only two variables (carbon and oxygen isotopes) is obviously insufficient for source determination, because the data fields of many quarries overlap. Successful attributions may be made for many of the isotopically ambiguous cases, however, when additional information such as grain size, mineralogy (i.e. dolomite or calcite), colour, and archaeological/historical data are taken into account.

Source identification

When the carbon and oxygen isotope data for all relevant marble quarries are plotted, a bewildering amount of overlap of the isotopic quarry fields result. As demonstrated in Table 1, more than one source identification occurs for all but two of the eighty-three sculptures we analysed, the possibilities being more than a dozen in several cases. Adding a third item of information to the procedure raises the accuracy substantially. This is the case here for sculptures identified as being of dolomitic marble or of fine-grained white marble.

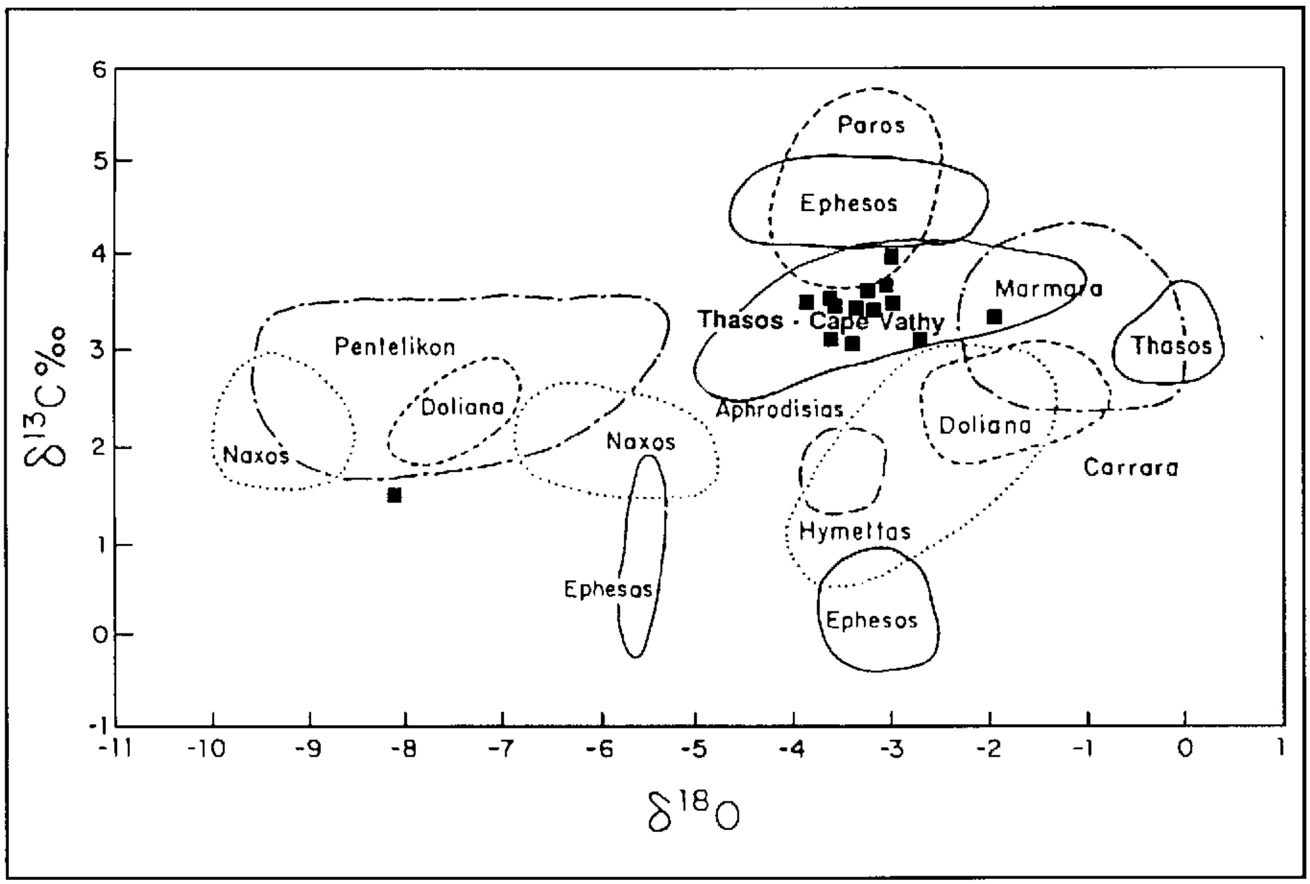

Fig.1. Carbon and oxygen isotope ratio field of the Cape Vathy dolomitic marble quarry on Thasos, superimposed on the database of Herz (1987). Thirteen of fourteen dolomitic sculptures (black squares) fall in this field.

Dolomitic marble (Cape Vathy, Thasos)

Herrmann (1992) reported briefly at the second ASMOSIA conference on some of our isotopic identifications of dolomitic marble from Cape Vathy on Thasos. Both Herrmann and Newman further explore the use of Cape Vathy marble in this volume and also provide more specific details on the Boston Three-sided Relief (MFA 08.205), which we found to be isotopically consistent with the Cape Vathy field.

Fourteen of the eighty-three sculptures were identified as dolomitic marble, both by visual inspection of fresh breaks (Herrmann), and XRD or Electron Microprobe (Newman). Thirteen of these fall in the isotopic field of Cape Vathy (Fig.1), suggesting that pure or nearly pure dolomitic specimens may be confidently assigned to this quarry source. The single outlier (MFA 76.729, Fragment of a Roman Sarcophagus) was determined by XRD to be a mixture of calcite and dolomite, and there is a strong possibility that the small powder sample we removed is not representative of the true quarry source. On re-examination, this sculpture clearly exhibits several different crystal morphologies, and additional samples have been removed for further isotopic tests.

The dolomite/calcite test, then, combined with isotopic data, has provided us with confident attribution of an additional thirteen sculptures, for a success rate of fifteen out of eighty-three (18%).

Fine-grained, white marble

The isotopic database assembled by Herz (1987, 1988, 1992, plus new additions) includes forty-two quarries, but unpublished analyses from an extensive database compiled by L. Moens, P. De Paepe and M. Waelkens have suggested that only three quarries were the principal suppliers of fine-grained white marble of sculptural grade (pers. comm.). These are Pentelikon in Greece, Dokimeion (or Afyon) in Turkey, and Carrara in Italy. The maximum grain size of marble from the three quarries is 1.8mm (Moens et al. 1990). For those sculptures that can be identified as fine-grained, a unique solution of high confidence is likely since these three sources overlap only slightly in their isotopic fields (Fig.2).

Twenty-three of the MFA and Sackler Museum sculptures have been identified so far as fine-grained, white marble. Ten fall in the isotopic field for Pentelikon (MFA 99.339; 04.12; 97.288; 01.8204; 99.341; 99.351; S1236; Sackler 1905.8; 1960.458; 1978.495.235); five are of marble from Carrara (MFA Res.08.34d; 95.68; 99.340;

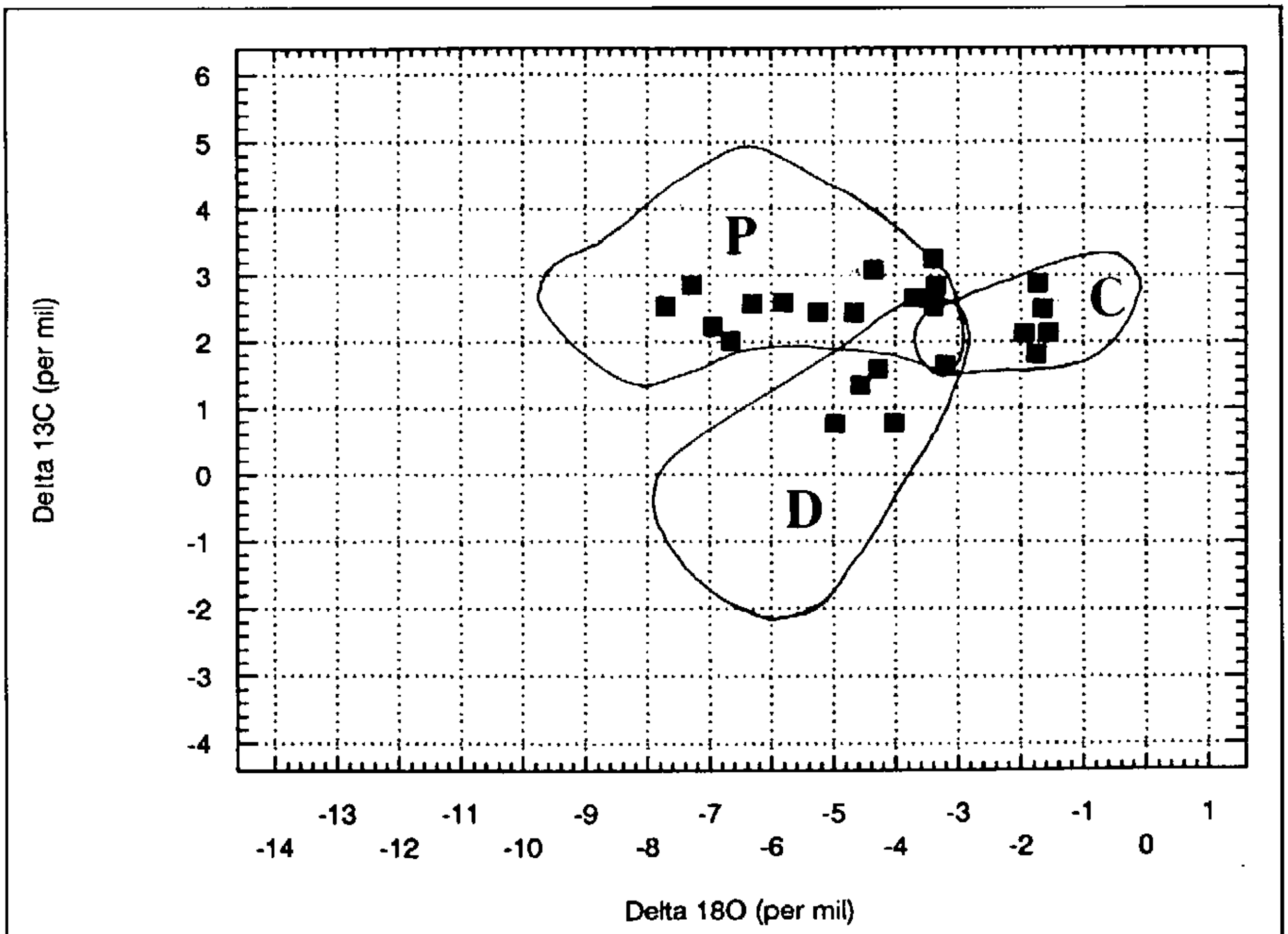

Figure 2. Three isotopic fields of fine-grained, white marble of sculptural grade. D = Dokimeion, Turkey; C = Carrara, Italy; P = Pentelikon, Greece. Twenty-three sculptures of fine grain fall in these fields.

24.150a; 24.150b); and four are of marble from Dokimeion (MFA 1971.746; 1984.167; 1977.712; 03.750). Only four (MFA 08.288; 65.1727; Sackler 1949.47.138; 1949.83) fall within the overlapping areas of these three quarry fields. We have thus identified the quarry sources of thirty-four out of eighty-three sculptures (41%) without considering their chronology or provenance.

These last two factors were considered preliminarily to identify the most likely quarry sources for the remaining forty-nine sculptures. We have given "probable" or "most likely" attributions to twenty-six of these sculptures; overall, 72% of the sculptures have thus been assigned to a single source. More detailed assessments of the individual works of art are still in preparation, however, and will be presented elsewhere.

Work in progress

Samples have been taken from an additional sixty Greek and Roman sculptures from the MFA, and their analysis is

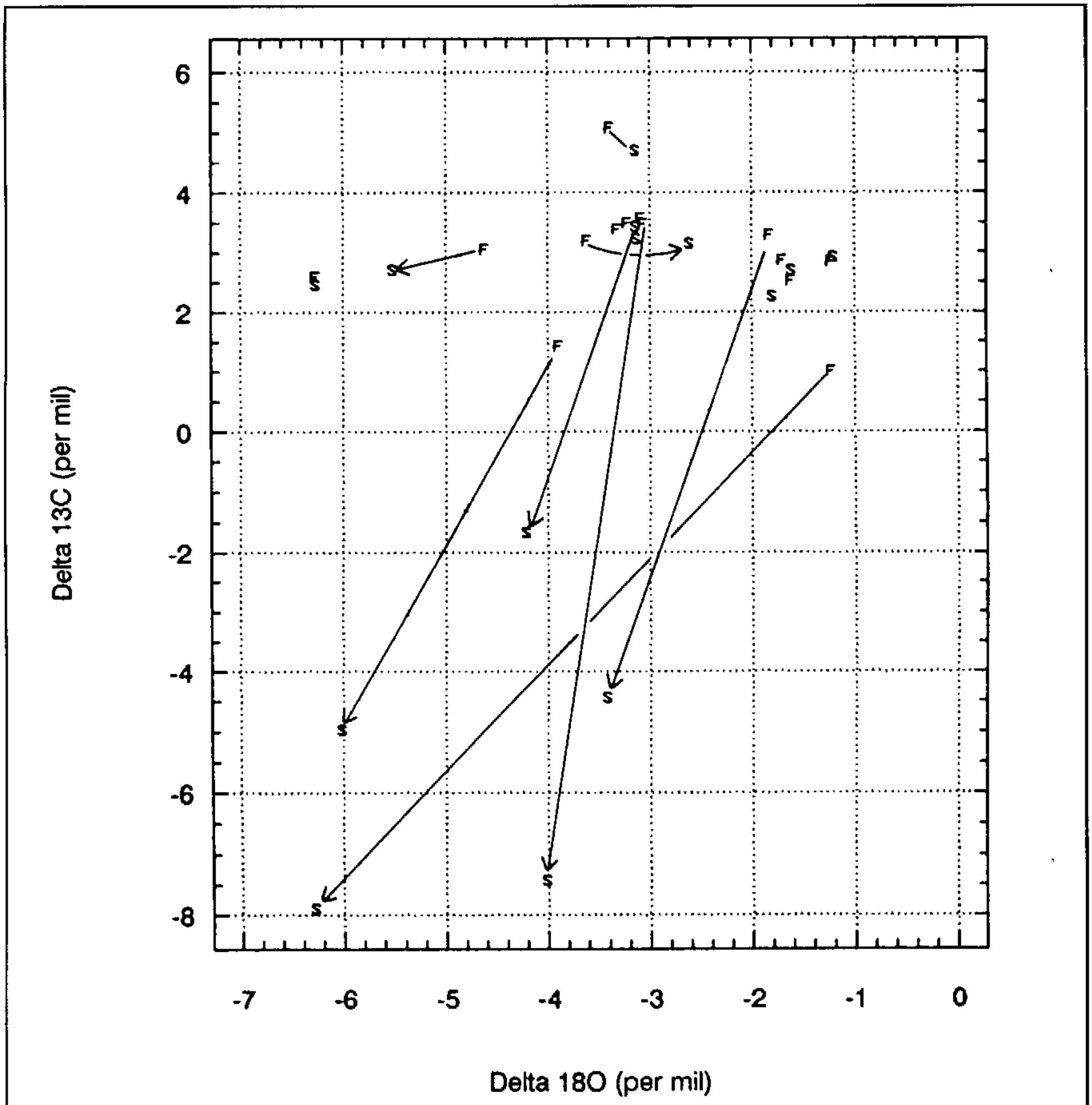

Figure 3. Comparison of the carbon and oxygen isotope ratios of weathered crusts (S) and fresh (F) marble of 14 sculptures. For seven sculptures, there is no significant isotopic difference; for six specimens, there is significant depletion of both isotope ratios.

Table 1. Carbon and oxygen isotope ratios of 83 classical marble sculptures from the Museum of Fine Arts, Boston and the Arthur M. Sackler Museum, Harvard University. The results are compared with several isotope databases for Mediterranean marble quarries to achieve source attributions.

Harvard Lab. No.	Museum Number	Sculpture Name	Reference: Catalogue Description. Provenance	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)	Isotopic Quarry Matches	Attribution	Justification
HI-001a	Sackler 1905.8	Multi-Figured Grave Stele	V&B 25: fine-grained, Pentelic marble. From Rome	+2.60	-6.27	N,Pe,E-2?,Iz?,Sa?	Pe	fine-grained marble
HI-002a	Sackler 1961.86	Grave Stele: Melisto	V&B 24: fine-grained, Pentelic marble	+1.00	-1.23	M?,S?	S	from Attica
HI-055	Sackler 1902.10	Statue of a Young Athlete in Repose	V&B 19: Parian marble From Greece or Italy	+2.19	-3.33	A2,C,D,Pa-2,Pe,Pr, Th,U,H,He,M,My, N-M	probably Pa-2	coarse-grained marble (XRD: calcite)
HI-056	Sackler 1926.48	Statue of a Young God or Hero, Meleager?	V&B 30: Parian marble From Italy	+2.45	-2.81	A2,C,D,Pa-2,Pr,Th, U,De-1,De-2,H,He, M,My,N-M	Pa-2	white coarse-grained marble (XRD: calcite)
HI-057	Sackler 1900.17	Small Statue of a Divine Female; Aphrodite?	V&B 34: Thasian marble. Said to be from Asia Minor	+2.38	-3.02	A2,C,D,Pa-2,Pe?, Pr,Th,U,De-1?,H, He,M,My,N-M	A2,Pa-2,Pr,Th, U,De-1?He,My, N-M	coarse-grained marble (XRD: calcite)
HI-058	Sackler 1922.171	Head of a Young Divinity, Hero, or Athlete	V&B 20: Thasian marble. From Paris	+4.03	-2.95	Pa-1,Pr,Th-CV,E-1, Sa	Th-CV	XRD: dolomite
HI-059	Sackler 1969.175	Head of a Kouros in Relief	V&B 11: Thasian marble. Said to be from Asia Minor	+4.12	-2.32	Pa-1?,Pr,Th-CV, E-1	Th-CV	XRD: dolomite
HI-090	MFA 08.205	Three-Sided Relief (Boston Throne)	C&V 30: large-grained marble from Greek islands. From Rome	+3.48	-3.06	Pa-1,Pr,Th-CV	Th-CV	XRD: dolomite
HI-092	MFA 99.350	Body of Aphrodite, Capitoline Type	C&V 166: coarse-grained Greek marble. From Italy	+1.80	-3.66	A2,C,D,Pa-2,Pe,Pr, U,E-2,He,My,N-M?	probably Pa-2	white, coarse-grained marble (XRD: calcite)
HI-093	MFA 99.351	Head of Aphrodite, Capitoline Type	C&V 167: coarse-grained Greek marble. From Italy	+3.50	-3.73	Pa-1,Pr,Th-CV,D??, Sa?	Th-CV	EM: dolomite
HI-094	MFA 1974.127	Faun or Young Satyr	C&V 172: crystalline Greek marble, probably from Asia Minor	+2.49	-2.58	A2,C,D?,Pa-2,Pr, Th,U,Dol-2,H,He, Ia,M,My,N-M	A2,Pa-2,Pr,Th,C, He,Ia,My,N-M	coarse-grained marble
HI-095	MFA 1973.212	Lady in Quasi-Divine Guise	C&V 368: Greek or Asia Minor marble with strong crystals. Said to be from Asia Minor	+1.19	-3.51	A1,A2,D,Pa-2?,U, E-2,H,M,My	A1,A2,Pa-2?,U, E-2,My	white, coarse-grained marble
HI-096	MFA 1976.6	Herakles, after Lysippos	C&V 104A: Greek island marble	+1.82	-2.67	C,D,Pa-2,Pr,Th?,U, E-2,H,He,M,My, N-M	Pa-2,Pr,Th?,U, E-2,He,My,N-M	coarse-grained marble (XRD: calcite)
HI-097	MFA 39.552	Kouros (torso)	C&V 12: island marble, probably Parian. From Attica	+2.67	-2.70	A2,C,D?,Pa-2,Pr, Th,U,Dol-2,H,He, Ia,M,My,N-M	probably Pa-2	white, medium-grained marble (XRD: calcite)
HI-098	MFA 10.159	Left Hand Holding Alabastron	C&V 31: coarse-grained Greek marble. From Italy	+2.05	-1.38	C,Pa-2,Pr,Th,U, De-1,H,Ia,M,S	probably Pa-2	stylistically related to mainland Greece (XRD: calcite)
HI-099	MFA 36.218	Herm-Bust of a Youth	C&V 28: marble from the northern Aegean islands. From Thessaly	+2.16	-0.93	C,Pa-2,Pr,Th,De-1, Dol-2,Ia,M	probably Pa-2	coarse-grained marble (XRD: calcite)
HI-100	MFA 99.342	Herm-Bust of a Poet or Philosopher	C&V 123: coarse-grained Greek marble. From Naples	+4.87	-3.14	Pa-1,E-1	probably Pa-1	coarse-grained marble, from Italy (XRD: calcite)
HI-101	MFA 24.419	Man of the Antonine Period	C&V 359: translucent marble. From Egypt	+2.46	-3.37	A2,C,D,Pa-2,Pe,Pr, Th,U,He,M,My, N-M	probably Pa-2, but possibly N-M	coarse-grained marble (XRD: calcite)
HI-102	MFA 68.772	Polykleitan Athlete	C&V 147: white marble with large crystals	+1.16	-3.55	A1,A2,D,Pa-2?,U, E2,H,M,My	A1,A2,Pa-2?,U, E2,My	white, coarse-grained marble (XRD: calcite)
HI-104	MFA 84.25	Akroterion from Assos	Herrmann 1990:92	+2.84	-1.23	C,Pr,Th,U,De-1, Dol-2,Ia,M	Pr,Th,U,De-1, Dol-2,Ia,M	XRD: calcite
HI-106	MFA 84.37	Corner of a Funerary Altar from Assos	C&V 279: grayish local marble	+3.30	-1.42	C,Pr,Th-CV,De-1, Dol-2,Ia,M	probably Pr	medium-grained marble (XRD: calcite)
HI-107	JJH 1978.43	Ephesos, stray find: probably strip from revetment?	Unpublished	+2.69	-1.40	C,Pa-2,Pr,Th,U, De-1,Dol-2,Ia,H?, M	Pa-2	glittering white, coarse-grained marble
HI-109	JJH 1978.51	Chip found near Mausoleum of Belevi	Unpublished	+3.88	-3.83	Pa-1,Pe?,Pr,Th-CV, E-1,Sa	probably E-1	grayish coarse-grained marble, used locally
HI-111	MFA 63.2760	Titus (Caesar AD 69-79and Emperor 79-81)	C&V 344: island marble. Perhaps from Greece	+3.58	-3.09	Pa-1,Pr,Th-CV,Sa	Th-CV	XRD: dolomite

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HI-113	MFA 84.64	Head from a Funerary Statue	C&V 111: crystalline island? marble. From Assos	+3.50	-3.22	Pa-1,Pr,Th-CV,Sa	Th-CV	XRD: dolomite
HI-115a	MFA 41.909	Dionysos	C&V 160: coarse-grained Thasian? marble	+3.20	-3.62	Pe,Pr,Th-CV,U?, My	Th-CV	XRD: dolomite
HI-117a	MFA 69.2	Fragment of a Sarcophagus Relief	C&V 242: coarse-grained marble, from N. Aegean	+3.39	-3.32	Pa-1,Pe?,Pr,Th-CV, My?	Th-CV	XRD: dolomite
HI-213	MFA 46.841	Head of a Boy	Herrmann 1992:97	+3.23	-2.71	Pa-1?,Pr,Th-CV, De-1,Dol-2,He,M, My	Th-CV	EM: dolomite
HI-214	MFA 76.722	Foot of a Youth	C&V 223: coarse-grained Greek marble. From Italy	+2.48	-3.30	A2,C,D,Pa-2,Pe,Pr, Th,U,De-1,H,He,M, My,N-M	A2,Pa-2,Pr,Th,U, De-1,He,My,N-M	coarse-grained marble (XRD: calcite)
HI-215	MFA 76.749	Sarcophagus Fragment: Dionysiac Figure	C&V 257: marble with considerable crystals, from northern Greek islands. From Italy	+3.35	-3.23	Pa-1,Pe?,Pr,Th-CV, My?	Th-CV	XRD: dolomite
HI-216	MFA 95.68	Alexander the Great: A Hellenistic Likeness	C&V 127: crystalline white marble, probably from Asia Minor or Greek mainland	+2.02	-1.65	C,Pa-2,Pr,Th,U, De-1,Dol-2,H,Ia,M, S	C,Dol-2	fine-grained marble
HI-217	MFA 99.341	Goddess, perhaps Athena	C&V 48: Parian marble. From Rhodes	+3.08	-4.42	Pe,Th-CV	Pe	not coarse enough for Thasian marble
HI-218	MFA 64.703	Christian Osteotheke	C&V 275: marble from NE Greece or NW Asia Minor. From Istanbul	+2.35	-1.78	C,Pa-2,Pr,Th,U, De-1,Dol-2,H,Ia,M, My	Pa-2,Pr,Th,U, De-1,Ia,My	coarse-grained marble (XRD: calcite)
HI-219	MFA 99.351	Top of Head of Aphrodite, Capitoline Type	C&V 167: From Italy	+2.07	-6.64	N,Pe,U,De-2, Dol-1?,Iz	probably Pe	from Italy (EM: Calcite)
HI-220	MFA 03.750	Section of Draped Female	C&V 93: fine-grained Greek mainland marble. From Italy	+0.81	-4.00	A1,D,E-2,M,My	D	fine-grained marble
HI-221	MFA 1986.20	Statuette: Goddess, Water Nymph	V&C 12: Aegean island marble	+3.31	-2.81	Pa-1?,Pr,Th-CV, De-1,Dol-2,He,M, My?	probably Pa-1	gray shadow, medium-grained marble
HI-222	MFA 01.8204	Youthful Hero or Athlete	C&V 168: Pentelic marble. From Rome	+2.27	-6.90	N,Pe,U?,DI,Dol-1, Iz,Sa?	Pe	fine-grained marble; from Rome
HI-223	MFA 1972.356	Funerary Urn of Cassius	C&V 243: white marble from Greek islands or Asia Minor	+3.40	-3.88	D??,Pa-1,Pe,Pr?, Th-CV,Sa?	Th-CV	XRD: dolomite
HI-224	MFA 1982.286	Statuette of Aphrodite Anadyomene	V&C 24: marble from western Asia Minor	-3.34	-5.99	N,Pe,Sa	N,Sa	medium to coarse-grained marble
HI-225	MFA 1981.783	Upper Part of Weary Herakles of Lysippos	V&C 22: marble from Greek islands or western Asia Minor. From Germany	-1.36	-3.41	A2,C?,D,Pa-2,U, E-2,H,He,M,My, N-M?	A2,Pa-2,U,E-2, He,My,N-M?	coarse-grained marble
HI-226	MFA 97.288	Herm-Bust of Menander?	C&V 121: fine-grained Italian? marble. From Italy	+2.48	-4.64	A2?,D?,N,Pe, Th-CV,U,Ch-1,E-2, My	Pe	fine-grained marble
HI-227	MFA 1975.292	Hadrian, Emperor AD 117 to 138	C&V 356A: Greek marble, probably Naxian. From Egypt	+3.66	-2.64	Pa-1,Pr,Th-CV, De-1?	probably Pa-1	medium-grained marble
HI-228	MFA 84.34	Ionic Capital from the Gymnasium, Assos	Unpublished	+2.87	-0.81	C,Pr,Th,De-1, Dol-2,Ia,M	Pr,De-1,Ia	faintly grayish, medium to coarse-grained marble
HI-229	MFA 76.732	Bearded Male Head	C&V 267: coarse-grained marble, probably Thasian. From Italy	+3.12	-3.54	Pa-1?,Pe,Pr,Th-CV, My	Th-CV	XRD: dolomite
HI-230	MFA 97.286	Torso of a Goddess	C&V 91: coarse-grained Greek island marble. From Italy	+1.79	-3.65	A2,C,D,Pa-2,Pe, Pr?,U,E-2,H,He,M, My,N-M	probably Pa-2	grayish, coarse-grained marble
HI-231	MFA 1971.746	Tyche-Fortuna	C&V 190: marble from western Asia Minor. From Pisidia	+0.80	-4.96	A1,D,Ch-1?,E-2,M, My?	D	fine-grained marble
HI-232	MFA 99.340	Seated Cybele	C&V 92: Pentelic marble. From Italy	+2.11	-1.83	C,Pa-2,Pr,Th,U, De-1,Dol-2?,H,Ia, M,My,N-M?,S	C	fine-grained marble
HI-233	MFA 99.339	Stele of a Mounted Warrior	C&V 18: Pentelic marble. From near Thebes	+2.54	-7.68	N,Pe,Dol-1,Iz,Sa	Pe	fine-grained marble
HI-234	MFA 89.152	Head of a Major Divinity: A Goddess?	C&V 90: crystalline gray-streaked marble, from western Asia Minor?	+2.45	-0.54	C,Pa-2,Pr,Th,De-1, Dol-2,Ia,M	Pa-2,Pr,De-1,Ia	medium to coarse-grained marble
HI-235	MFA 96.695	Dionysos	C&V 46: Greek island marble. From Athens	+5.24	-3.09	Pa-1	Pa-1	EM: calcite

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HI-236	MFA 96.694	Aphrodite	C&V 158: coarse-grained, Greek island marble	+1.93	-1.05	C,Pa-2,Pr?,Th, Dol-2,H,Ia?,M,S	probably Pa-2	coarse-grained marble (EM: calcite)
HI-238	MFA 67.948	Commemorative or Funerary Tondo	C&V 278: marble from SW Asia Minor. From Asia Minor	+1.43	-3.90	A2,D,Pa-2,U,E-2, He,M,My,N-M?	A2,Pa-2,U,E-2, He,My,N-M?	coarse-grained marble (EM: calcite)
HI-240	MFA 1988.327	Trajanic Female Portrait	Herrmann 1991	+5.08	-3.39	Pa-1,E1	Pa-1	white, coarse-grained marble
HI-242	MFA 1980.212	Funerary Relief	V&C 46: Thasian marble. Said to be from Greece	+3.29	-1.83	C?,Pr,Th-CV,U?, De-1Dol-2,Ia,M	Th-CV	XRD: dolomite
HI-244	MFA 1979.477	Statue of Eros	V&C 20: marble from Greek islands or western Asia Minor	+2.51	-3.08	A2,C,D,Pa-2,Pe,Pr, Th,U,H,M,My,N-M	A2,Pa-2,Pr,Th,U, My,N-M	grayish, coarse-grained marble
HI-245	MFA 1977.712	Fragment of Statue: Helmeted Head of Ares	V&C 21: white crystalline marble, probably from SW Asia Minor	+1.58	-4.29	A2,D,Pa-2,U,E-2, He,M,My,N-M?	D	fine-grained marble
HI-246	MFA 72.337	Zeus or Dionysos	C&V 212: crystalline island or western Asiatic marble. From Cyprus?	+2.19	-3.51	A2,C,D,Pr,Pa-2,Pe, Th,U,H,He,M,My, N-M	A2,Pr,Pa-2,Th,U, He,My,N-M	medium to coarse-grained marble (XRD: calcite)
HI-247	MFA Res.08.34d	Herakles and a Nymph	C&V 116: crystalline marble, probably from Greek mainland. From Rome	+1.91	-1.70	C,Pa-2,Pr,Th,U,H, Ia,M,S	C	fine-grained marble
HI-248	MFA 84.63	Table Support (Trapezophorus)	C&V 317: marble from NW Asia Minor. From Assos	+2.45	-1.38	C,Pa-2,Pr,Th,U, De-1,Dol-2,H,Ia,M	Pa-2,Pr,Th,U, De-1,Ia	coarse-grained marble
HI-249	MFA 84.70	Man of the Flavian Period	C&V 348: coarse marble, probably from western Asia Minor. From Assos	+4.29	-2.84	Pa-1,Pr,Th-CV,E-1, Sa	Pa-1,Pr,Th-CV, E-1,Sa	
HI-250	MFA 76.729	Fragment of a Muse Sarcophagus	C&V 253: Pentelic marble. From Italy	+1.42	-8.05	N,Pe,De-2	N,Pe,De-2	
HI-251	MFA 96.698	Girl from Corinth	C&V 353. Greek island marble. Said to be from Corinth	+2.98	-4.75	N,Pe,Th-CV,S?	N,Pe,Th-CV,S?	
HI-252	MFA 1988.463	Statuette of a Good Shepherd	Unpublished	+2.89	-2.02	C,Pr,Th,U,De-1, Dol-2,H,He,Ia,M, My	probably Pr	coarse-grained marble
HI-253	MFA 1988.463	Base	Unpublished	+2.12	-2.67	A2,C,D?,Pa-2,Pr, Th,U,De-1,Dol-2,H, He,M,My,N-M	probably Pr	coarse-grained marble
HI-254	MFA S1236	Table Support (Trapezophorus)	C&V 316: probably Pentelic. From Italy	+3.20	-3.38	Pa-1?,Pe,Pr,Th-CV, My	Pe	fine-grained marble
HI-255	MFA 04.12	Carian Zeus	C&V 44: fine-grained Greek marble. From Asia Minor	+2.63	-5.81	N,Pe,Th,E-2,S?	Pe	fine-grained marble
HI-256	MFA 1924.150 B	Tomb of Mino da Fiesole	Young et al 1937: Carrara marble. From Florence	+2.87	-1.70	C,Pr,Th,U,De-1, Dol-2,Ia,M	C	fine-grained marble
HI-258	MFA 1924.150A	Tomb of Mino da Fiesole	Young et al 1937: Carrara marble. From Florence	+2.53	-1.62	C,Pa-2,Pr,Th,U, De-1,Dol-2,H,Ia,M, My	C	fine-grained marble
HI-260	MFA 28.893	Table Support (Trapezophorus)	C&V 318: Greek, probably Pentelic marble	+1.92	-2.02	C,Pa-2,Pr,Th,U, De-1,He,Ia,M,My,S	perhaps Pa-2	medium-grained, translucent marble (XRD: calcite)
HI-261	MFA 15.856	Goddess	C&V 47: Parian marble. Allegedly excavated in Athens	+4.48	-3.37	Pa-2,Pr,E-1,Sa	probably Pa-2	grayish, coarse-grained marble
HI-262	MFA 01.8207	Arsinoe III	C&V 130: Greek island marble. From Egypt	+5.22	-3.08	Pa-1	Pa-1	
HI-263	MFA 65.1727	Saturn (Kronos) with Features of Commodus	C&V 367: Greek island or western Asia Minor marble. From Algeria	+2.85	-3.37	D?,Pe,Pr,Th-CV,U, De-1,He,M,My	probably Pe	white, with some gray spots, fine-grained marble
HI-264	MFA 08.288	Grave Stele of a Youth	C&V 16: Pentelic marble. From Thebes	+1.67	-3.19	A2,C,D,Pa-2,Pe?, Pr,U,E-2,H,He,M, My,N-M	probably Pe	fine-grained marble
HI-265	MFA 1984.167	Statuette of Hunter with Dog	V&C 27: crystalline marble, probably from western Asia Minor	+1.34	-4.51	A1,A2,D,N?,Pa-2?, U,E-2,He,M,My	D	fine-grained marble
HI-348	Sackler 1991.640	Dionysus	Unpublished	+3.06	-4.62	N?,Pe,Th-CV	Th-CV	looks like Thasian marble
HI-349	Sackler 1949.47.138	Head of a Bearded Man	V&B 142: Luna (Carrara) marble	+2.67	-3.61	A2,D?,Pe,Pr, Th-CV,U,He?,M, My,N-M?	C,D,Pe	fine-grained marble

Harvard Lab. No.	Museum Number	Sculpture Name	Reference: Catalogue Description. Provenance	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)	Isotopic Quarry Matches	Attribution	Justification
HI-350	Sackler 1949.83	Head of a Bearded Man	V&B 140: Greek mainland marble	+2.56	-3.36	A2,C,D,Pa-2?,Pe, Pr,Th,U,H?,He,M, My,N-M?	C,D,Pe	fine-grained marble
HI-351	Sackler 1960.458	Funerary Relief	V&B 105: Pentelic? marble	+2.45	-5.18	D?,N,Pe,Th-CV,U, Ch-1,E-2	probably Pe	fine-grained marble
HI-352	Sackler 1963.54	Head and Shoulder of Tiberius	V&B 135: Greek, probably Pentelic marble. From Italy	+5.02	-3.81	Pa-1,E-1	Pa-1	white marble
HI-353	Sackler 1977.216.2185	Stele of a Lady and Her Servant	V&B 103: Greek island marble	+2.13	-1.74	C,Pa-2,Pr,Th,U, De-1,H,Ia,M,N-M?, S	probably Pa-2	coarse-grained marble
HI-354	Sackler 1977.216.2186	Funerary Stele	V&B 106: Greek island marble	+2.44	-4.20	A2,D,Pe,Th-CV,U, My	A,My,U	coarse-grained, probably local Asiatic marble
HI-355	Sackler 1978.495.235	Fragment of a Dionysiac Relief	V&B 97: Thasian? marble	+2.86	-7.25	N,Pe,Dol-1,Iz,Sa	probably Pe	fine-grained marble
HI-356	Sackler 1988.459	Aphrodite	V&B 35: low-grade marble, perhaps from western Asia Minor	+1.91	-3.50	A2,C,D,Pa-2,Pe,Pr, U,E-2,H?,He,M, My,N-M	A2,Pa-2,Pr,U, E-2,He,My,N-M	medium to coarse-grained marble

References:
C&V = Comstock & Vermeule 1976; V&B = Vermeule & Brauer 1990; and V&C = Vermeule & Comstock 1988.

Key to Marble Quarries:
A1 (Aphrodisias 1); A2 (Aphrodisias 2); C (Carrara); Ch-1 (Chemtou 1); Ch-2 (Chemtou 2); D (Dokimeion=Afyon); De-1 (Denizli 1); De-2 (Denizli 2); DI (Djebel Ichkeul); Dol-1 (Doliana 1); Dol-2 (Doliana 2); E-1 (Ephesos 1); E-2 (Ephesos 2); H (Hymettos); He (Heracleia); Ia (Iasos); Iz (Iznik); M (Mani); My (Mylasa); N (Naxos); N-M (Naxos-Melanes); Pa-1 (Paros 1); Pa-2 (Paros 2); Pr (Prokonnesos=Marmara); S (Sounion); Sa (Sardis); Th (Thasos-Cape Phaneri & Thasos-Aliki); Th-CV (Thasos-Cape Vathy); U (U ak)

in progress. The results will be reported elsewhere, in the first instance at the annual meeting of the Archaeological Institute of America, to be held in Washington, DC in December 1993.

Alteration of the isotope signature

We analysed fourteen samples of weathered crust and compared their isotopic ratios with those of the unweathered interiors of the same sculptures; the results are listed in Table 2 and illustrated in Figure 3. Six of the weathered crusts had isotopic shifts greater than 1‰ away from the original fresh marble. Five of these show a depletion of the oxygen isotope ratio of 1‰ or more, accompanied by a depletion of the carbon isotope ratio of 5-10‰. For the sixth sculpture, the oxygen isotope ratio is enriched by 1‰, but the carbon isotope ratio is unchanged.

A number of researchers (Doehne *et al*, 1992; Margolis & Showers, 1988, 1990; Newman, 1990) have previously described isotopic shifts in weathered surfaces of dolomitic marble. Dedolomitization (or calcitization) is the process by which dolomite is transformed into micritic calcite. Calcite is usually precipitated from groundwater, but it may also be produced by recrystallization of the pre-existing dolomite. Both carbon and oxygen isotope values of groundwater tend to be more negative than those of marble. Doehne *et al* (1992), for example, found a shift of -0.81‰ in $\delta^{18}O$ for a Zeus head from the J. Paul Getty Museum and concluded that this shift was the result of inorganic dedolomitization; the significant shift in $\delta^{13}C$ that is typical of further alteration by soil micro-organisms was not evidenced in the Zeus head. Their result was consistent with the analysis by Herz (1987:38) of a sarcophagus in the British Museum, which showed oxygen isotope depletion of 0.6‰ and little change in the carbon isotope ratio. Our results show similar depletions of 1-2‰ in oxygen isotope ratios, but also show significant changes in the carbon isotope values, suggesting the biological mediation of microorganisms and/or lichens. Margolis and Showers (1988:table 3; 1990: table 4) have reported negative shifts in carbon isotope ratios of up to 13‰ and in oxygen isotope ratios of up to 3.2‰ in marble sculptures. The experiment of Heller and Herz (this volume), of course, shows that the action of organically produced oxalic acid on dolomitic marble can produce negative shifts of this order through the production of calcium oxalate, and in a matter of months.

A number of uncertainties remain about the phenomenon of isotopic shifts in weathered crusts on marble. Seven of the fourteen crusts we measured showed no alteration in their original isotopic composition: it is not clear if these sculptures had been chemically cleaned in the recent past. The production of calcium oxalate from dolomite by microorganisms (Heller and Herz, this volume) can result in substantial shifts in both oxygen and carbon isotope ratios in a very short time. Inorganic calcitization of dolomite proceeds at a slower rate, and oxygen ions are likely to be exchanged between marble and the environment during this process. It is not clear what mechanisms other than biological ones are likely to cause alteration of both carbon and oxygen isotope ratios. Alteration of both isotope ratios in calcitic marble is particularly problematic.

When isotopic alterations of marble crusts are invoked as evidence for the antiquity of sculptures (see Margolis

Table 2. Comparison of carbon and oxygen isotope ratios of weathered crusts and fresh marble of fourteen marble sculptures.

Harvard Lab. No	Museum Number	Marble Type	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)	Sample Location
HI-001a	Sackler 1905.8	prob. calcite	+2.60	-6.27	fresh marble
HI-001b	Sackler 1905.8	"	+2.47	-6.27	surface
HI-002a	Sackler 1961.86	calcite	+1.00	-1.23	fresh marble
HI-002b	Sackler 1961.86	"	-7.89	-6.28	surface
HI-090	MFA 08.205	dolomite	+3.48	-3.06	fresh marble
HI-091	MFA 08.205	"	-7.42	-4.01	surface
HI-104	MFA 84.25	calcite	+2.84	-1.23	fresh marble
HI-105	MFA 84.25	"	+2.92	-1.20	surface
HI-111	MFA 63.2760	dolomite	+3.58	-3.09	fresh marble
HI-112	MFA 63.2760	"	-1.67	-4.21	surface
HI-113	MFA 84.64	dolomite	+3.50	-3.22	fresh marble
HI-114	MFA 84.64	"	-3.44	-3.14	surface
HI-115a	MFA 41.909	dolomite	+3.20	-3.62	fresh marble
HI-116	MFA 41.909	"	+3.15	-2.61	surface
HI-117a	MFA 69.2	dolomite	+3.39	-3.32	fresh marble
HI-118	MFA 69.2	"	+3.23	-3.13	surface
HI-238	MFA 67.948	calcite	-1.43	-3.90	fresh marble
HI-239	MFA 67.948	"	-4.97	-6.01	surface
HI-240	MFA 1988.327	prob. calcite	+5.08	-3.39	fresh marble
HI-241	MFA 1988.327	"	+4.70	-3.14	surface
HI-242	MFA 1980.212	dolomite	+3.29	-1.83	fresh marble
HI-243	MFA 1980.212	"	-4.44	-3.42	surface
HI-256	MFA 24.150 B	prob. calcite	+2.87	-1.70	fresh marble
HI-257	MFA 24.150 B	"	+2.70	-1.61	surface
HI-258	MFA 24.150 A	prob calcite	+2.53	-1.62	fresh marble
HI-259	MFA 24.150 A	"	+2.27	-1.80	surface
HI-348	Sackler 1991.640	prob. dolomite	+3.06	-4.62	fresh marble
HI-347	Sackler 1991.640	"	+2.71	-5.52	surface

and Showers, 1988, 1990; Newman, 1990), it is clear that the composition of the fresh marble and its end product must be known and the mechanism by which it was produced must be understood. Petrographic and XRD techniques can be used to differentiate dolomite, metamorphic (type I) calcite, biologically produced (type II) calcite, and calcium oxalate, which is associated with lichens and microorganisms (Del Monte and Sabbioni, 1987; Heller and Herz, this volume). A high concentration of magnesium is often taken as evidence for dolomitic marble; lichens chelate magnesium, however, so high magnesium, as determined by various elemental techniques, does not necessarily prove dolomite.

It should also be noted that dolomitic marble from Thasos does have up to 13.9% natural metamorphic calcite present (Doehne *et al*, 1992; Herz, 1988), so that a small sample taken from a Thasian sculpture is likely to contain some calcite, and the isotopic analysis of such a sample will typically show a depletion of a few tenths per mil relative to pure dolomite. Furthermore, both calcitization and the organic production of oxalic acid may penetrate beyond the surface of a sculpture when cracks or fissures are present, potentially explaining the inverse relationship of the oxygen isotope ratios in sculpture MFA 1941.909, where the 'interior' sample may have included natural and/or weathering crust calcite.

The potential of heavy stable isotopes

It is clear that a single method of analysis, such as light stable isotope measurements, is not particularly effective as a means for tracing marble quarry sources. Combining carbon and oxygen isotope measurements with the identification of dolomitic marble (visual inspection and XRD) and the visual identification of fine-grained, white marble gave us a success rate of only 42% of unequivocal source tracing. Other techniques, such as cathodoluminescence (Barbin *et al*, 1992), electron spin resonance (Mandi *et al*, 1992; Lloyd *et al*, 1988), and trace element analysis (Mello *et al*, 1988; Moens *et al*, 1988) are also effective to some extent, but at least two methods must be used simultaneously to achieve a significant level of resolution (see, e.g., Moens *et al*, 1992). Which methods are used largely depend on the extent of sampling allowed by museum curators. Small powder samples (for isotopic analysis and/or XRD) can be frequently obtained, whereas the chips and core samples necessary for thin sections (cathodoluminescence, petrography/grain size measurements) or trace element analysis (large samples are required to overcome heterogeneity) can typically be removed only when sculptures are undergoing restoration or remounting.

We propose that heavy stable isotopes, such as those of strontium and/or neodymium, could be very strong discriminators of marble quarries, and along with light stable isotope ratios (carbon and oxygen) may resolve nearly all the potential sources. Herz (1992;191; Herz *et al*, 1982) has briefly examined the utility of strontium isotope analysis for marble source identification. We base our confident proposal on our experience with modern elephant ivory (van der Merwe *et al*, 1990) and rhinoceros horn (Hall-Martin *et al*, 1993), in which we can differentiate more than 20 game park sources in Africa. While the analysis of heavy stable isotopes is more costly than carbon and oxygen isotope analysis, it requires only one to ten milligrams of marble powder; the ability to rely entirely on minimally destructive techniques would undoubtedly increase the number of marble artifacts available for source determination. The combination of light stable isotope analysis (carbon and oxygen) and a heavy isotope ratio such as strontium, along with informed visual inspection (grain size, dolomitic marble), XRD (dolomitic marble), plus archaeological/historical information, should allow a confident attribution of quarry source for at least 80-90% of all classical marble sculptures, using essentially non-destructive methods.

To use strontium or other heavy isotope ratios for marble source identification requires the construction of an isotopic database. The database will require measurements of high precision (to, say, five decimal places for strontium) and could be quite costly. Commercial charges for heavy isotope ratios range upwards from US$200 per sample, depending on the isotope ratios involved. The announcement by Moens (this volume) that analyses of strontium and neodymium isotope ratios in a series of quarry samples have been started by Noël Gale and Zofia Stos-Gale of Oxford University is to be warmly welcomed.

If a strontium isotope database were constructed, it might also prove possible to make quarry attributions for unknown samples by less precise (and less costly) isotopic methods. ICP-MS, for example, yields semi-quantitative isotopic information at a per-sample cost comparable to light stable isotope measurements and provides, in addition, trace element information on very small powder samples. While the precision is not as good as stable heavy isotope analyses provided by thermal ionisation mass spectrometers (typically, five decimal places for strontium by thermal ionisation, two to three decimal places for strontium by ICP-MS), it may be sufficient to resolve the equivocal source identifications which remain after the determination of carbon and oxygen isotope ratios, mineralogy, and maximum grain size.

Conclusion

We have successfully identified the quarry sources of thirty-four out of eighty-three classical marble sculptures by measuring their carbon and oxygen isotope ratios and determining that they are of dolomitic or fine-grained, white marble. An additional twenty-six have been tentatively attributed to a single quarry source based on archaeological, art historical, and provenance considerations. Powder samples of less than ten milligrams were required for these analyses. We propose that unequivocal source identifications of at least 80-90% could be possible if a quarry database of strontium isotope ratios (and/or other heavy isotope ratios like neodymium) were to be constructed.

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