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Bronze Age Copper Sources in the Mediterranean: A New Approach

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Summary. Efforts by scientists to locate the sources of copper used in ancient Mediterranean and Near Eastern cultures through comparative chemical analyses of copper ores and archeological artifacts have largely failed for various mineralogical and metallurgical reasons. The isotopic composition of lead, an element present in a minor amount in many copper ores and bronze objects, is unchanged through metallurgical processes and may in principle be used to determine the sources of the copper used in Bronze Age artifacts. Results suggest that for Late Bronze Age Crete the Laurion region in Attica, Greece, may have been a more important copper source than Cyprus.

In attempting to understand the prehistory of the Bronze Age, the importance of a reliable scientific method for determining the geographic sources of the metals used to make ancient copper and bronze artifacts can hardly be overemphasized. Objects made of copper-based alloys take second place in archeological excavations of this period only to ceramic artifacts, and the reliable determination of the sources of copper used by the Minoan, Mycenaean, Cypriot, Egyptian, and Near Eastern cultures would add to the knowledge of cultural contacts and trade. Even textual studies and problems in ancient geography may be advanced by successful work on ancient sources of copper, a case in point being the location of Alashiya. Though often mentioned in Egyptian, Hittite, and Ugaritic documents, and most often equated by scholars with part or all of Cyprus, the location of Alashiya is still a matter of dispute (*1*). Textual evidence does not allow a certain identification (*2*), but it is certain that copper was a major export from Alashiya to Egypt, Syria, and elsewhere.

Typological studies (*3*) can provide information about cultural contacts, but such studies cannot help in the certain classification of some types of artifacts or the metal fragments and amorphous lumps found at many excavations. A method of establishing metal provenance based on metallurgical examination or scientific analysis has been sought since at least the 1920's (*4*), especially in Stuttgart (*5*) and Vienna (*6*). Numerous chemical analyses of artifacts and ores for their major and some minor elements have been described (*5–11*) and provide a record of the gradual evolution of the use first of fairly pure copper and then of arsenical and tin bronzes. These studies have not provided evidence of the sources of the copper, although a recent study in which many trace elements were analyzed and cluster analysis was used to link ores with artifacts has had more success (*12*). Nevertheless the judgment made by Catling (*3*, p. 12) in 1964 about chemical analyses may still be generally justified: "There seems little doubt that at the moment a solution to the problem of the sources of supply for ancient copper and bronze objects in the Mediterranean lands cannot be hoped for through the medium of the laboratory."

Chemical Analysis and Provenance Studies

Reasons for the general failure of chemical analysis for provenance studies of copper-based alloys are not hard to find. From a mineralogical point of view, it is necesary to assume that all copper ores in a particular locality have a similar chemical composition; not only is this rarely true (*13, 14*), but chemical heterogeneities can occur even over short distances in the same ore lode (*15*), usually because of varying admixtures of other metallic minerals, especially iron, lead, zinc, and arsenic. Nonetheless, it has been suggested that the occurrence in bronze artifacts of a particular element at an unusually high concentration may be characteristic of a particular geographic or mineralogical origin. For instance, some investigators (*16, 17*) hold that zinc is diagnostic of copper (or slag) from Cyprus, whereas others (*18*) suggest that zinc may be an indication that sulfide ores were used. In fact, the zinc content of Cypriot copper ores varies widely even in the same ore occurrence (*19*); furthermore, the presence of zinc is not specific to Cypriot copper ores, as demonstrated by the high zinc content of copper ores from Rio Tinto, Spain (*20*); Aibunar, Bulgaria (*21*); and the Laurion region of Greece. The zinc content of the ores from the Laurion and from Aibunar (Table 1), together with measurements by Tylecote *et al.* (*20*) of the partitioning during smelting of trace elements between copper and slag, show that the presence of zinc in an ancient copper object cannot be used to differentiate between its derivation from an oxidized ore or a sulfide ore. Neither is cobalt necessarily diagnostic of origin from an Anatolian source (*22*), since cobalt also occurs in high concentrations in some Cypriot ores (*23*) and no doubt elsewhere in the Aegean.

From a metallurgical point of view, the smelting of ores under primitive conditions also introduces poorly controlled changes in the pattern of minor and trace

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Table 1. Lead and zinc concentrations in copper ores from ancient mining regions. Abbreviation: N.D., not determined.

Mining region	Ore type	Reference	Copper (%)	Lead (%)	Zinc (%)
Aibunar, Bulgaria	Oxidized, malachite, some azurite	(*21*)	?(>10)	0.1 to 10	0.1 to 10
Timna, Sinai	Oxidized, malachite	(*20*)	23.7	0.05	0.05
Rio Tinto, Spain	Oxidized, chrysocolla, and malachite	(*20*)	18	0.02	0.05
Rio Tinto, Spain	Sulfide	(*20*)	17.4	0.05	4.0
Avoca, Ireland	Sulfide	(*20*)	15	0.45	3.1
Lakxia tou Mavrou, Cyprus	Oxidized	(*35*)	1.7	5	N.D.
Kalavasos, Cyprus	Sulfide	(*69*)	1.0	0.02	0.3
Laurion, Greece*	Oxidized, azurite		47.0	0.4	0.5
Laurion, Greece*	Oxidized, malachite		45.8	0.1	1.3

*Analyses made by the authors.

elements in the copper metal, when compared with the pattern in the copper ore, especially when one considers the contribution of elements contained in the added flux. The direct copper smelting experiments of Tylecote *et al.* (*20*) show, for instance, that some elements such as bismuth are so volatile as to be almost completely lost as fume, but some such as arsenic and antimony may be largely retained in the metal; however, higher smelting temperatures cause a greater reduction of zinc, antimony, lead, and bismuth in the copper. Sulfide ores lose large amounts of arsenic, antimony, and lead if dead-roasted before smelting (though zinc is not appreciably lost in this process); the alternative process of smelting sulfide ores to a matte that is then roasted before final smelting eliminates almost all of the bismuth, arsenic, lead, antimony, and zinc. The mineralogical and metallurgical difficulties standing in the way of correlating impurities in ores with those in the metals produced from them were described qualitatively over 20 years ago by Thompson (*15*) and again by Craddock (*9*) and by Muhly (*22*), but they have often been ignored.

Isotopic Analyses

Although patterns of minor and trace element composition are variable both in ores and the copper produced from them and are subject to change by varying additions of flux and varying conditions of smelting, roasting, and so forth, there is an elemental property that is unchanged through all these chemical processes. That property is the isotopic composition of each metallic element in the ore, and its conservation makes it particularly suitable as an indicator of provenance. Of course, it is necessary to find an element whose isotopic composition varies in nature, is more or less constant throughout a given ore body, but varies from one copper ore body to another (or at least from one geographic region to another).

The ideal choice would be copper itself, and it has been shown that small but measurable variations occur naturally in the isotopic composition of copper (*24*), covering a range of about 12 per mil in the ratio ^{63}Cu/^{65}Cu. But the largest deviations from the average terrestrial isotopic composition of copper occur in rare minerals such as volborthite and aurichalcite; in chalcopyrite, malachite, and azurite, the common ore minerals of copper, isotopic variations greater than the analytical error were not found.

An alternative choice is lead, which occurs in Bronze Age objects made of copper-based alloys in concentrations as low as 0.001 percent to as high (rarely) as 7 or 8 percent. The isotopic composition of lead varies in nature over a range of up to 5 percent (in, for example, the ratio ^{208}Pb/^{206}Pb) because three of the four natural isotopes of lead are partly derived from the radioactive decay of ^{238}U, ^{235}U, and ^{232}Th, respectively; such variations are the basis for lead isotope geochronology and lead isotope geochemistry (*25*). The suggestion that lead isotope studies might prove useful in establishing the geographic provenance of archeological objects made of lead or silver was investigated some 14 years ago independently by Brill and Wampler (*26*) and by Grögler *et al.* (*27*). Since then the method has been applied to the study of the sources of silver for Archaic Greek coinage (*28*) and to the sources of lead and silver in the Bronze Age Aegean (*29*–*31*).

Lead Isotopes and Copper Sources

The application of lead isotope analyses to the provenance of copper is not as straightforward as it is to provenance studies of lead and silver. We must be sure that in the ancient copper-based alloys studied the lead present is an accidental contamination coming from the copper ore and was not added by man. Lead occurs in many copper ores, both sulfidic and oxidized, as is demonstrated by analyses of ores from ancient mining regions (Table 1). Analyses of copper ores from the Laurion region and from the fourth millennium copper mines at Aibunar show that lead can occur in significant amounts in oxidized copper ores. It is to be expected that higher concentrations of lead will occur in copper ores from polymetallic ore deposits, of which the deposits in Aibunar and the Laurion are good examples.

When lead is present in a copper ore, the smelting experiments of Tylecote *et al.* (*20*) show that as a general rule the lead concentration in the copper produced is about half that in the copper ore, for both the direct smelting of oxide ores and for the two-step smelting of sulfide ores involving an intermediate dead-roasting process. Especially instructive is their smelting of an oxide ore containing 40 percent copper and 4.65 percent lead to produce a raw copper containing 2.02 percent lead (with 2.32 percent iron) and a crucible refined copper containing 1.5 percent lead (with 0.5 percent iron). It is possible for ancient objects made of copper-based alloys to contain up to 1 or 2 percent lead which has come from the copper ores themselves, though lower concentrations may be more common (Table 1).

Lead in Bronze Age Artifacts

The analyses of Bronze Age Greek artifacts made by Craddock (*9*), supplemented by those of Renfrew (*7*) and the Stuttgart group (*5*) for the Early Bronze Age, show that most copper, arsenical bronze, and tin bronze objects contain less than 1 percent lead. Indeed, out of a total of 185 analyses of Early and Late Bronze Age axes, chisels, swords, daggers, knives, spearheads, arrowheads, and figurines, 161 show lead contents of less than 1 percent, and only nine show more than 3 percent. Catling and Jones (*11*) found that of 141 Late Minoan II copper and bronze objects from the unexplored Mansion, Knossos, Crete, only three objects had as much as 1 percent lead. Only for Late Bronze Age statuettes is there any evidence of the use of deliberately leaded bronzes. Most Bronze Age Greek copper and bronze artifacts contain lead in amounts which are consistent with the lead being derived as a minor impurity from the copper ores (or fluxes) themselves.

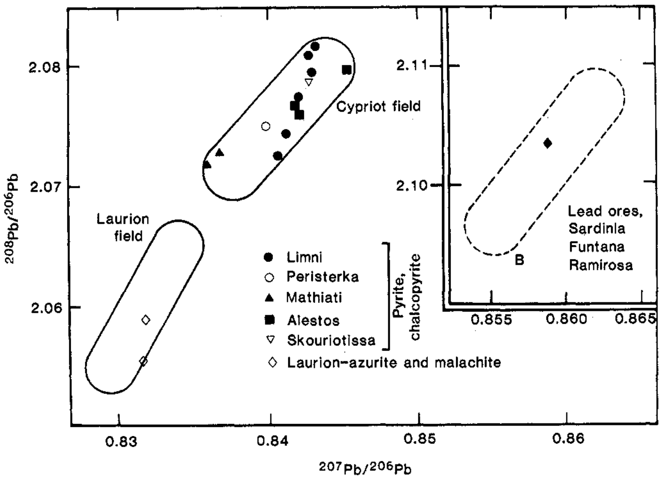

Fig. 1. Lead isotope analyses of copper ores from Cyprus and the Laurion, Attica. The dashed field B, which appears also in Figs. 3 and 4, is not yet defined by analyses of copper ores, but lead ores from Sardinia plot within it.

Indeed, Craddock (*32*) showed that it was not until Archaic times that the Greeks began to use deliberately leaded bronze. Leaded bronze for coins and large statues was introduced as late as the third century B.C., even though the ancient practice of adding lead to bronze (5 to 25 percent lead) was useful chiefly because it increases the fluidity of the molten bronze, which was especially advantageous in making thin-walled hollow castings or solid castings with great detail (*32*). The Bronze Age metalsmith was perhaps influenced by the fact that bronze with more than a few percent of lead is difficult to work (*33*) by hammering, a fact possibly reflected in the low lead content of Bronze Age Greek swords, daggers, knives, axes, chisels, spearheads, and arrowheads. Even in the Archaic and Classical periods lead was avoided for bronze vessels raised from sheet metal, since leaded bronze would have torn in the raising process; moreover, lead is absent from Hellenistic Greek decorative bronzes, which were extensively worked after casting, even though the contemporary statuettes are highly leaded (*32*).

The evidence that lead was rarely deliberately added to copper or bronze objects in the Greek Bronze Age seems compelling and indicates that the lead in ancient copper or bronze objects below about 1.5 percent (and certainly below 0.5 percent) is present as an accidental impurity from the copper ore (or flux) and that the isotopic composition of the lead will give the provenance of the copper ore (or flux).

Fluxes and Fuels

A flux is not necessary to smelt pure malachite or azurite ores such as those from the Laurion (Table 1); the small amounts of iron and silica present (less than 0.5 percent) do not require it. Such pure copper ores would not usually have been available; for instance, the oxidized ore analyzed by Ottaway (*34*) from the fourth millennium mines at Rudna Glava in Yugoslavia is essentially a malachite ore in a highly ferruginous gangue (goethite), and in order to remove the iron (typically as a fayalite slag) from the copper it would have been necessary to add the correct amount of sand. Sand does not contain lead, and where it is used as a flux any lead in the smelted copper will certainly have come from the ore. Sulfide chalcopyrite ores usually occur with a large excess of pyrite and after dead-roasting must be fluxed with sand, so that no contribution of lead from the flux is to be expected. Some oxidized ores, such as those from Timna in the Sinai (*20*), occur in a highly siliceous gangue and must be fluxed with an iron mineral such as hematite or limonite. Though often practically lead free, such iron oxides sometimes contain large amounts of lead, as is shown by the analyses of the gossan, or iron hat, of the copper ore deposits at Lakxia tou Mavrou in Cyprus (*35*). This lead is unlikely to invalidate isotope analyses since any iron minerals used as a flux are likely to have been taken from near the copper deposits, possibly from the gossan of those deposits; both will contain lead of the same isotopic composition.

A similar principle applies to a possible trace contribution of lead to copper from the charcoal used in the smelting (*20*); in the Bronze Age the trees used to make the charcoal for smelting are not likely to have come from very far away from the ore deposit, so that any lead contributed is most likely to have an isotopic composition identical with that of the copper ore. Trees grown away from a region containing ore deposits are unlikely then to have contained significant amounts of lead.

Copper-Based Alloys

For bronzes, lead may also have been introduced with the alloying metal arsenic or tin. It is improbable that arsenic was added to copper in the Greek Bronze Age. Earlier in the Bronze Age tin was perhaps added as cassiterite (*36*), though tin bronze may have been discovered accidentally by the smelting of oxidized copper ores containing an admixture of wood tin (*31*). Cassiterite deposits in Precambrian rocks are chiefly in granite pegmatites and contain no lead. Cassiterite deposits in rocks younger than Cambrian age display a wider range of genetic association, but of the five classes of deposit noted by Rapp (*37*) only the subvolcanic and the pneumotolytic-hydrothermal deposits rarely contain small amounts of lead. In the later Greek Bronze Age tin may have been added as the metal.

A consensus of opinion is forming that in the Early Bronze Age the discovery and early production of arsenical bronze was made by the accidental, and then roughly controlled, smelting of a mixture of oxidized copper ores together with green basic copper arsenates naturally occurring together with copper ores in parts of the same ore body (*31, 32, 38, 39*). If these arsenates contained lead, it would be of the same isotopic composition as that characteristic of the copper ore body as a whole. If arsenosulfide minerals such as enargite, tennantite, or arsenopyrite, which normally do not contain lead, were used in the smelting process (*36*), they would not disturb the isotope signature of the lead from the copper ore.

Lead Isotope Analyses

In principle the lead isotope method seems promising for determining the source of the copper used in Bronze Age objects from the Mediterranean world. The method was evaluated first by analyzing 13 Early Cycladic (2900 to 2100 B.C.) artifacts from Amorgos, Naxos, and Paros, 6 Late Minoan 1A (1550 to 1500 B.C.) objects from Thera, and 22 Middle and Late Minoan (1800 to 1360 B.C.) objects from Crete. These analyses must then be compared with lead isotope analyses of copper ores from

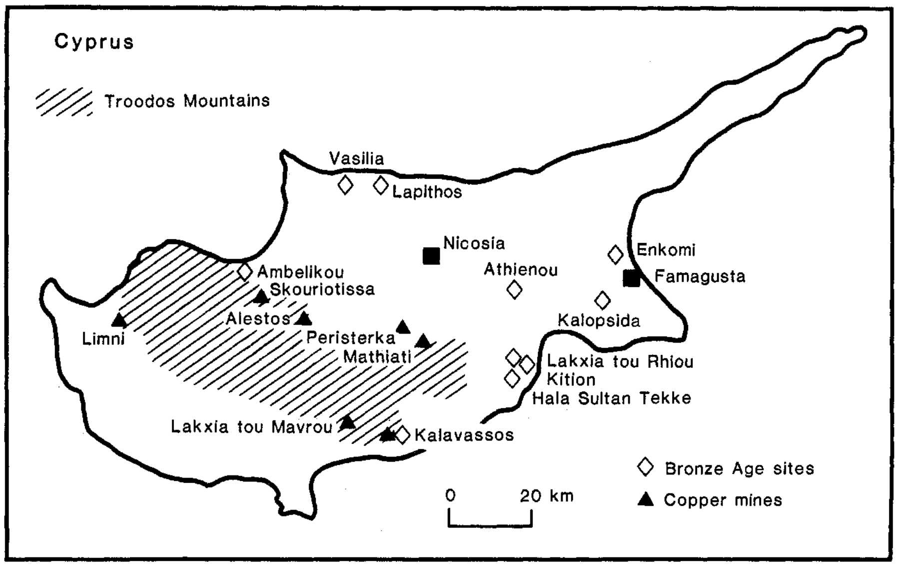

Fig. 2. Map of Cyprus showing the locations of copper mines from which ores have been analyzed and relevant Bronze Age archeological sites.

possible ancient mining regions. Because Cyprus is often assumed to have been an important source of copper for the Bronze Age Aegean, lead isotope compositions and lead concentrations (*40*) of pyritic copper ores from five mines on Cyprus were determined (Table 2); the lead concentrations are uniformly low and below 0.004 percent. We use the isotopic compositions of the Cypriot ores to define a field (Fig. 1). The locations of the mines are shown in Fig. 2; the Limni deposit was worked in Roman times and has large associated slag heaps (*41*); such undated slag heaps are also adjacent to the Mathiati (*41*) and Skouriotissa (*42*) mines.

Until a more extensive survey of the isotopic composition of lead in Cypriot copper ores is made, as well as surveys of the ancient slag heaps and especially of the stratified slags and copper metal from Bronze Age sites, we cannot be certain that we have completely defined the field characteristic of Cypriot ores. However, there is geochemical evidence suggesting that isotopic composition will not vary greatly from the field shown in Fig. 1. The lead isotopic compositions of metalliferous sediments (umbers) from deposits in the Cypriot pillow lavas form a tight grouping intermediate between the compositions characteristic of seawater and of mid-ocean ridge basalts, suggesting that the lead incorporated in the umbers was derived partly from seawater and has been partly leached out of the underlying basaltic oceanic crust during interactions of hot water and rock (*43*). The ore leads plot in the same field as the umbers, suggesting a broadly similar geochemical origin for the lead in the pyrite copper ores. Unless some ores in Cyprus have a different origin, and the work of Constantinou and Govett (*44*) suggests that this is unlikely, then one does not expect to find very different Cypriot compositions.

The field of lead isotope compositions for the Laurion mining region in Attica has been established by analyses of galena (*31*) (Fig. 1). Since the ore deposit in the Laurion is primarily a polymetallic (lead, zinc, iron) sulfide deposit, any sulfidic or oxidized ores of copper which do occur should contain small amounts of lead of the isotopic composition that characterizes the Laurion region. Partial analyses of malachite and azurite from the Laurion (Table 1) show the expected minor amounts of lead, and isotopic analyses (Fig. 1) show that the composition falls in the Laurion field.

The isotopic analyses of the Cycladic and Cretan artifacts are given in Tables 3 and 4; full chemical analyses are given either by Renfrew (*7*), Craddock (*9*), or Slater (*45*). From plots of the isotopic analyses of the artifacts (Figs. 3 and 4), it is clear that they tend to cluster in well-defined groups and that many Early Bronze Age Cycladic artifacts and Late Bronze Age Cretan artifacts derive their copper from the same two sources (the Laurion and field B).

These analyses allow us to assess the possibility that additions of arsenic or tin might carry with them enough lead to disturb the lead isotope pattern characteristic of the copper. For arsenic the analyses in Table 3 show that three typologically similar Early Cycladic daggers from Amorgos (1927.1359, AE240, and 1927.1360) have a virtually identical lead isotopic composition even though one (1927.1359) contains 8.7 percent arsenic and only 0.19 percent lead, and the other two (AE240 and 1927.1360) contain no arsenic and 0.54 and 0.38 percent lead, respectively; none of these daggers contain tin. Similar evidence comes from the Laurion isotope composition of the four tin-free Cretan objects AE605, AE600,

Table 2. Lead isotope and concentration (in parts per million) analyses of copper ores from five sites on Cyprus and the Laurion in Greece.

Sample	Mining region	Ore type	Lead (ppm)	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$
CY/75/12	Limni	Pyrite, chalcopyrite	12.9	2.07924	0.84295	18.525
CY/75/33	Limni	Pyrite, chalcopyrite	26.9	2.07420	0.84127	18.533
CY/75/33	Limni	Pyrite, chalcopyrite		2.07232	0.84075	18.553
CY/75/69	Limni	Pyrite concentrate	39.0	2.08170	0.84320	18.544
CY/75/69	Limni	Pyrite concentrate		2.08081	0.84279	18.548
CY/75/70	Limni	Chalcopyrite concentrate		2.07734	0.84217	18.524
CY/75/87	Mathiati	Pyrite, chalcopyrite	15.2	2.07256	0.83689	18.661
CY/75/89	Mathiati	Pyrite, chalcopyrite	30.4	2.07152	0.83603	18.658
CY/75/91	Peristerka	Pyrite, chalcopyrite	1.7	2.07498	0.84012	18.526
CY/75/98A	Alestos	Pyrite, chalcopyrite	1.6	2.07950	0.84554	18.404
CY/75/98D	Alestos	Pyrite, chalcopyrite	3.3	2.07584	0.84216	18.467
CY/75/98D	Alestos	Pyrite, chalcopyrite		2.07643	0.84214	18.493
	Skouriotissa	Pyrite, chalcopyrite		2.07864	0.84277	18.476
L1	Laurion	Azurite	4000	2.05850	0.83191	18.822
L2	Laurion	Malachite	1100	2.05499	0.83183	18.776

AE622, and 1910.181 (Table 4), whose As/Pb ratio varies from 0.17 to 4.8. Together these data establish an absence of disturbance for two different lead isotopic compositions over an As/Pb range from essentially 0 to 46. Evidence that additions of tin do not disturb the lead isotopic ratio comes from the uniformly Laurion composition of Cycladic objects AE236, 1927.1358, AE86, and AE87 (Table 3) and of the Cretan objects AE595, 1927.1386, 1924.13, and AE489 (Table 4), even though the Sn/Pb ratio varies from 0 to essentially 28.

The Cycladic objects from Amorgos, Naxos, and Paros (except 1927.1362) group into four fields (Fig. 3). Three objects from Amorgos and two from Naxos fall unambiguously in the Laurion field, one from Amorgos falls in the Cypriot field and another into the as yet unidentified field B, and four objects from Amorgos and one from Paros lie in field A (in the $^{208}Pb/^{206}Pb$ versus $^{207}Pb/^{206}Pb$ plot) on the edge of the Cypriot field. Until now, in the application of lead isotopes in archeology it has not been necessary to use the information contained in the third measured isotopic ratio, $^{206}Pb/^{204}Pb$, but it is necessary in the present situation. Figure 5, a plot of $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{206}Pb$ for the Cycladic objects and Cypriot ores, shows that of the objects on the edge of the Cypriot field in Fig. 3 only the flat ax 1910.618 from Paros (labeled P in both figures) is consistent with a derivation from Cypriot ores. The four objects from Amorgos in field A must come from another, as yet not identified, copper source; object S (compare Figs. 3 and 5) must come from yet another, non-Cypriot source. Of these 13 Early Bronze Age copper artifacts it seems that five derive from Laurion ores, two are consistent with a Cypriot origin, four come from unidentified source A, one from unidentified source B, and object S (1927.1362) either comes from yet a third unidentified source or represents a mixture of copper derived from Laurion and Cyprus.

Earlier work (*30, 31*) has shown that at this period in the Cyclades 7 out of a total of 20 lead or silver objects derived from an ore source in the Laurion and the rest chiefly from ores in Siphnos. It may not be without significance that the proportion (35 percent) of Early Cycladic lead and silver objects derived from a Laurion source is close to the proportion (38 percent) of Early Cycladic copper alloy objects derived from copper ore sources within the Laurion. It is interesting that of the four objects from Amorgos that fall within field A two (AE237 and 1927.1359) are in all three isotopic ratios within the Siphnian field (*32*) and two are close to that field; analyses of 11 ores from the Early Bronze Age mining region of Ayios Sostis on Siphnos (*46*) show that three contain respectively 37.5, 14.4, and 13.9 percent copper. Undoubtedly the majority of the ores mined at Ayios Sostis were silver-lead-antimony ores exploited for their silver, but a minor exploitation of copper ores cannot be excluded.

For the later Bronze Age in the Cyclades we have at present only the six analyses of Late Minoan IA artifacts from Akrotiri in Thera: three fall within the Laurion field and three within the Cypriot field in all three ratios (compare Figs. 3 and 5). At this time Thera imported nearly all its lead from the Laurion (*31*); further analyses are needed to determine the proportions of copper that came from the Laurion and Cyprus.

Of the 22 Cretan artifacts analyzed (Fig. 4) 11 fall definitely within the Laurion field (and that for Milatos may be Laurion), five fall within field B, two from Siteia are apparently just within or close to the Cypriot field, and three group below the Laurion field. A Cypriot origin for the copper of the two objects from Siteia is excluded by the $^{206}Pb/^{204}Pb$ ratio—definitely for 1966.542 and probably for 1966.547. These analyses provide no evidence that Crete obtained copper from Cyprus in the Late Bronze Age but show that at least four sources were being used; the dominant one was the Laurion followed by the unidentified source B. That Crete did not obtain much Cypriot copper in the Late Bronze Age is in accord with Catling's (*47*) interpretation of the archeological evidence that in his Industrial Phase I (up to 1380 B.C., or about the middle of Late Minoan IIIA in Crete and the end of Late Cypriot IIA in Cyprus) there was little contact between Cyprus and Crete and uncertain evidence of full industrial exploitation in Cyprus. Geographically, of course, Crete is closer to the Laurion than it is to Cyprus.

The Mixing Problem

Evidence from excavations suggests that at least during the Late Bronze Age in the Mediterranean it was common practice to melt down broken and worn bronze objects for reuse. This has suggested to many that analyses (whether chemical or isotopic) of copper alloy objects cannot give accurate information on the provenance of the copper. However, economic and technical factors such as supplies of wood and the difficulty and cost of smelting lower grade ores,

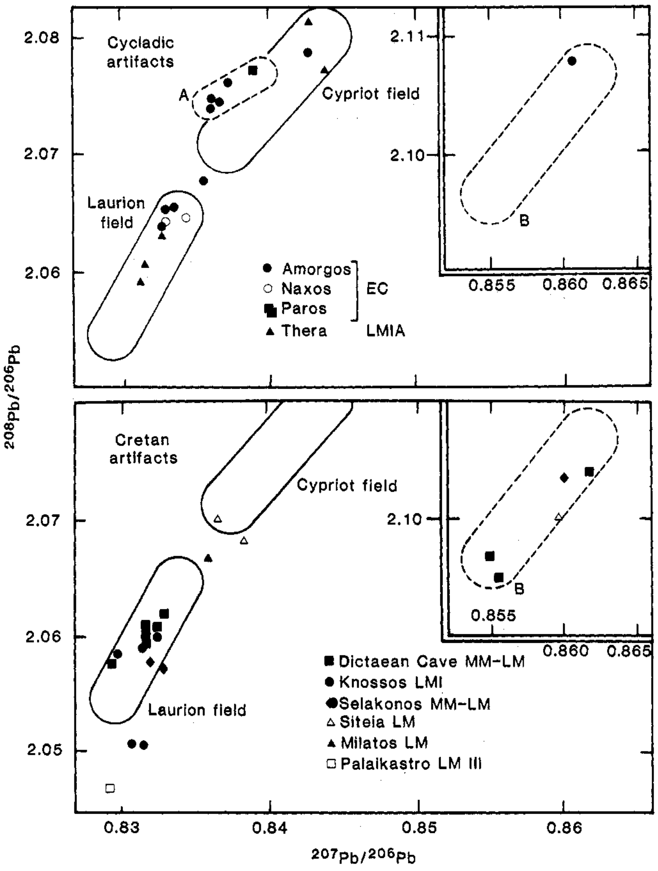

Fig. 3 (top). Lead isotope analyses of Bronze Age copper or bronze artifacts from the Cyclades. Fig. 4 (bottom). Lead isotope analyses of Bronze Age copper or bronze artifacts from Crete. Dashed fields A and B are groupings of artifact analyses only and are not based on associated ore analyses. *EC*, Early Cycladic; *LM*, Late Minoan; *MM*, Middle Minoan.

Table 3. Lead isotope analyses of copper-based alloy artifacts from the Cyclades. Abbreviations: EC, Early Cycladic; MC, Middle Cycladic; LC, Late Cycladic; LM, Late Minoan; N.D., not detected; n.d., not determined; N.A., not analyzed.

Object	Museum number*	Date†	Site	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$	Sn (%)	As (%)	Pb (%)	Assigned copper source
Flat ax	1910.618	ECII?	Paros	2.07730	0.83927	18.634	N.D.	3.4	0.25	Cypriot?
Flat ax	AE236	ECII?	Amorgos	2.06563	0.83394	18.794	0.85	1.1	0.13	Laurion
Dagger, class IIa	AE237	ECII to IIIA	Amorgos	2.07614	0.83756	18.740	N.D.	N.D.	0.5	A
Dagger, class VII	AE241	ECIIIA	Amorgos	2.06394	0.83299	18.859	N.D.	N.D.	1.2	Laurion
Dagger, class IId	AE231	ECIIIB	Amorgos, Arkesine	2.07883	0.84304	18.573	N.D.	3.5	0.1	Cypriot?
Dagger, class IVb	1927.1359	ECIIIB	Amorgos	2.07471	0.83708	18.748	N.D.	8.7	0.19	A
Dagger, class IVa	AE240	ECI-MC	Amorgos	2.07480	0.83656	18.762	N.D.	N.D.	0.54	A
Dagger, class IVa	1927.1360	ECI-MC	Amorgos	2.07397	0.83640	18.738	N.D.	N.D.	0.38	A
Dagger, class IVa	1927.1358	ECI-MC	Amorgos	2.06536	0.83365	18.823	0.27	N.D.	2.6	Laurion
Ax-adze	AE86	LC	Naxos	2.06470	0.83476	18.764	7.3	0.8	0.26	Laurion
Ax-adze	AE87	LC	Naxos	2.06421	0.83326	18.832	8.3	N.D.	2.1	Laurion
Sickle	1927.1362	LC	Amorgos	2.06770	0.83601	18.764	5.5	0.66	1.1	?
Shaft-hole ax	1927.2968	Uncertain	Amorgos	2.10840	0.86079	18.215	8.8	N.D.	0.59	B
Scale pan	Arvaniti 2	LMIA	Thera	2.08133	0.84320	18.476	12.9	0.14	n.d.	Cypriot
Sheet	4138	LMIA	Thera	2.06072	0.83186	18.846	9.2	0.15	n.d.	Laurion
Scrap	4135,Δ17	LMIA	Thera	2.07000	0.83958	18.609	5.2	0.14	n.d.	Cypriot
Scrap	25-8-71,Δ16	LMIA	Thera	2.07709	0.84405	18.495	7.5	0.13	n.d.	Cypriot
Tripod	?	LMIA	Thera	2.06312	0.83317	18.789	9.7	0.05	n.d.	Laurion
Jug	CBE17/1973/3582	LMIA	Thera	2.05897	0.83154	18.842	N.A.	N.A.	N.A.	Laurion

*Artifacts from Amorgos and Paros are from the Ashmolean Museum, Oxford, and have been described by Renfrew (*7*); artifacts from Akrotiri, Thera, are described by Slater (*45*). †The tentative dates for the objects from Amorgos, Naxos, and Paros were supplied by MacGillivray (*70*).

Table 4. Lead isotope analyses of copper-base alloy artifacts from Crete. The chemical analyses are from Craddock (*9*). Abbreviations: LM, Late Minoan; MM, Middle Minoan; and N.D., not detected.

Object	Ashmolean number*	Boardman number†	Date	Site	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$	Sn (%)	As (%)	Pb (%)	Assigned copper source
Male figurine	AE605	2	LM	Dictaean Cave	2.05989	0.83178	18.863	N.D.	0.1	0.6	Laurion
Male figurine	AE595	3	MM to LM	Dictaean Cave	2.05903	0.83166	18.853	0.4	0.1	0.29	Laurion
Male figurine	AE600	10	LM	Dictaean Cave	2.05749	0.82935	18.939	Trace	1.3	0.27	Laurion
Female figurine	AE622	31	LM	Dictaean Cave	2.06055	0.83179	18.897	N.D.	0.9	0.24	Laurion
Ram figurine	AE32	43	LM	Dictaean Cave	2.09495	0.85558	18.335	7.6	0.2	0.25	B
Ram figurine	AE598	44	LM	Dictaean Cave	2.10393	0.86185	18.143	5.35	0.2	0.39	B
Double ax	1927.1386	198	LM	Dictaean Cave	2.06156	0.83279	18.822	8.0	1.3	0.33	Laurion
Double ax	1927.1253	199	MM to LM	Dictaean Cave	2.09654	0.85508	18.273	4.2	0.1	0.47	B
Votary	AE14	372	LM	Patso Cave	2.06073	0.83230	18.833	0.6	0.7	0.25	Laurion
Double ax	1924.13		MM?	Knossos	2.05051	0.83077	18.826	6.4	1.4	1.45	?
Double adze	1910.181		LMI	Knossos	2.05810	0.82956	18.930	N.D.	0.2	0.07	Laurion
Sword	AE489	PM832	LMIB	Knossos	2.05052	0.83128	18.815	11.5	0.7	0.14	?
Spearhead	AE491	PM832	LMIB	Knossos	2.05867	0.83135	18.801	11.7	0.1	0.04	Laurion
Spearhead	AE492	PM832	LMIB	Knossos	2.05964	0.83226	18.804	3.6	0.25	0.16	Laurion
Sickle	1909.376		LMIII	Palaikastro	2.04703	0.82936	18.854				?
Ax-adze	AE88		LM?	Milatos	2.06628	0.83582	18.751	7.9	0.8	0.41	Laurion?
Double ax	AE85	CP6	MMIII	Selakonos	2.05756	0.83197	19.018	6.6	0.2	0.44	Laurion
Double ax	AE90		MM to LM	Selakonos	2.10344	0.85990	18.115	3.7	0.6	0.19	B
Chisel	AE92		LM	Selakonos	2.05697	0.83284	18.806	0.95	2.4	0.22	Laurion
Sword	1966.542		LMIII	Siteia	2.06984	0.83646	18.756	8.3	0.06	0.42	?
Sword	1966.543		LMIII	Siteia	2.10010	0.85960	18.200	8.2	0.2	0.48	B
Spearhead	1966.547		LMIII	Siteia	2.06800	0.83819	18.678	8.6	0.1	0.3	Cypriot?

*Registration number at the Ashmolean Museum, Oxford, England. †Number under which the object is described by Boardman (*71*).

would have focused the attention of Bronze Age miners and metalsmiths on regions yielding rich ores in reasonable quantity, and probably on mining regions fairly accessible to the sea for easy transport of the metal produced.

Even in the Late Bronze Age there is likely to have been a tendency to exploit to the limit the few known extensive and accessible sources of rich ores, and while they still yielded rich ore there would have been little incentive to invest time and labor in searching out other sources. The mixing problem may have been overemphasized. In reality it may reduce to the possibility of mixing of metal from only a few sources, of which but a couple may have been important to a particular culture at any one period. If mixing is an important problem then isotopic compositions of objects should be scattered over the whole range of compositions characteristic of the various metal sources. For lead artifacts in the Aegean cultures this is not the case (*29–31*); instead there appear to be two major groupings of isotopic compositions characteristic of the Laurion and Siphnian mines. For copper alloys the isotopic groupings suggest that but a few sources may also be involved and that mixing between them may not be much of a problem.

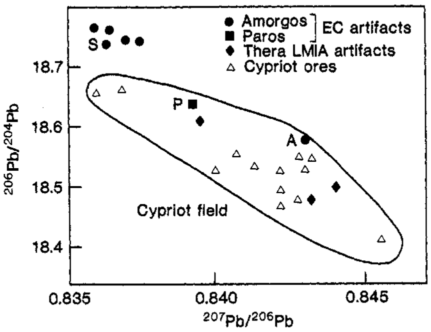

Fig. 5. Alternative plot of the lead isotope data for Cycladic artifacts and Cypriot copper ores. Of the four artifacts from Amorgos and one from Paros which lie on the edge of the Cypriot field (Fig. 3), only the Parian object truly belongs to the Cypriot field. *A*, *P*, and *S* correspond, respectively, to artifacts AE231, 1910.618, and 1927.1362 in Table 3. *EC*, Early Cycladic; *LM*, Late Minoan.

Copper Ores in Cyprus and the Laurion

That the Laurion may have been a more important source of copper than Cyprus to Late Bronze Age Crete will come as a surprise to many. Cyprus was renowned as a source of copper in late antiquity; its copper mines were mentioned by Theophrastus in about 310 B.C. (*48*) and by Pliny (*49*, Book 34, 2–4) in the first century A.D., though it should be noted that Pliny also mentions copper mines in Italy, Germany, Gaul, and other places. Scholars, especially philologists, have tended to extrapolate back into the Bronze Age the undoubted importance of Cyprus as a copper source in late antiquity and to use this extrapolation in a partly circular argument to equate Cyprus with Alashiya, but there is no direct evidence that Cyprus was a major exporter of copper so early. The extreme position of Davies (*50*), however, that there is practically no evidence for the production of copper on Cyprus in the Bronze Age can no longer be maintained in the face of the evidence from stratified archeological excavations, accumulated since he wrote, of Early Bronze Age copper production at Ambelikou (*51*), dated Early Cypriot III to Middle Cypriot I (2025 to 1800 B.C.), and Late Bronze Age copper production at Apliki (*52*), Enkomi (*53*), Athienou (*54*), and Kition (*55*), dated variously between 1650 and 1200 B.C. but chiefly after 1400 B.C. The scale of this Bronze Age copper industry is not known, and there is still no direct evidence that the Cypriot mines were exploited in the Bronze Age. The many old slag heaps and mining galleries on Cyprus have so far not been associated with other than Roman material (*56*), though some must have been worked before or at the time when Theophrastus wrote of them.

Copper ores on Cyprus, which (except for the Troulli outlier) are located in the pillow lavas in the foothills surrounding the Troodos massif, are overwhelmingly pyritic ores containing small amounts of chalcopyrite and average about 1 to 3 percent copper, though uncommon minor amounts of oxidized ores such as malachite, azurite, and cuprite have been reported (*57*). A copper content of 3 percent is regarded as the mark of a rich Cypriot ore; it is quite exceptional for enrichment to be as much as 5 percent. Though rich by modern standards the Cypriot ores are poor in comparison with ores from other ancient mining areas (Table 1). Smelting such ores in the Bronze Age would have been difficult; it seems more likely that on Cyprus, as elsewhere, richer oxidized copper ores were worked in the Bronze Age. Malachite was recorded by two early authors to exist in the Cypriot mines. All commentators agree that the green "smaragdi" described by Pliny (*49*, Book 37, 65–75) as coming from the Cypriot copper mines were massive forms of malachite, as were the "false smaragdi" described by Theophrastus as occurring in these mines (*58*). Nevertheless, without more direct evidence it seems well to heed Wheeler's warning (*59*) that it is conceivable, though perhaps unlikely, that the copper being worked in the second millennium on Cyprus was imported from sources outside the island.

If oxidized copper ores exist in the Laurion it need come as no surprise to find that they were worked in the Bronze Age. There is now overwhelming evidence that the silver-lead ores of the Laurion were worked in the Bronze Age and that these two metals traveled from Attica to Perati, Athens, Mycenae, and Vapheio in one direction and to the Cyclades and Crete in another direction (*30*). The techniques and furnace temperatures required to smelt efficiently both impure oxidized copper ores and impure silver-lead ores are similar; both ores require a minimum temperature of about 1200°C in order to slag the gangue effectively away from the metal, usually as a fayalite slag. Once the technical problems in smelting lead have been overcome there would have been little difficulty in applying essentially similar techniques to smelting copper.

Mainland Greece has largely been ignored by archeologists as a possible source of copper on any scale in the Bronze Age, yet copper appears with lead, silver, zinc, and manganese in a list of the metals produced by Greece in a 1944 naval intelligence handbook (*60*) and the Laurion is one of four regions specifically listed as important for copper production. The guide (*61*) accompanying the metallogenetic map of Greece lists over 70 copper ore deposits on mainland Greece and, although in the 1950's Greece produced only about 10 percent as much copper ore as Cyprus, nevertheless in 1955 the mainland Greek copper production was not negligible at about 2000 tons and in 1938 was nearly 5000 tons (*60*). For the Bronze Age it is probably more important to concentrate on the availability in various regions of rich copper ores rather than to compare such figures for the modern exploitation of ores containing no more than 2 percent copper. Because the lead isotope data indicate that Laurion was an important Bronze Age copper source we confine our attention to evidence that rich copper ores do indeed occur there.

The analyses of Laurion copper ores (Table 1) were made on 0.5-kilogram hand specimens; massive malachite and azurite still occur in the Laurion, but we have not yet surveyed the extent of such remaining ores. Marinos and Petrascheck (*62*) mention that native copper and cuprite occur at Camaresa, Ilarion, and Sounion in the Laurion region. Phillips (*63*) records that at Laurion the lead ore is more or less cupriferous; at the Jean Baptiste shaft it contains sometimes 6 to 12 percent copper, and at Karysto the galena contains from 5 to 18 percent copper. Phillips (*63*) notes that elsewhere in the Laurion both sulfide and carbonate copper ores occur. In the 1830's Fiedler (*64*) describes occurrences of malachite, azurite, and cuprite at several places in the Laurion. Cordella (*65*) mentions that copper is sometimes found as chalcopyrite veins in the mica schists of Camaresa, that even the calamine (zinc carbonate and silicate) often contains some copper, and that the calamine at Loutsa is also nickeliferous. Cambresy (*66*), who was mining engineer at Ergastiria in the Laurion, mentions occurrences of small amounts of native copper, chalcopyrite, brochantite, and cuprite as well as occurrences of malachite and azurite. He also mentions that a 2000-kg mass of rich oxidized copper ore was found in a calamine deposit; even if it were as poor as 25 percent copper it corresponds with 500 kg of copper metal, enough to make 5000 Class IVa Early Cycladic daggers like one from Amorgos which is in the British Museum (*7*, plate 7). Cambresy (*66*) also mentions the rare occurrences of olivenite (copper arsenate) and arsenates of nickel and cobalt. Marinos and Petrascheck (*62*) record the presence of many complex copper arsenates and iron arsenates in the Laurion, especially in the Camaresa and Dhipseliza regions. It seems that in the Laurion there are not only rich copper ores but also arsenic in forms in which it may easily have been accidentally incorporated into copper metal, coming in either with the copper ore or with arsenical iron minerals used as a flux.

There is some textual evidence that copper was known in Roman times to exist in the Laurion. In writing of smaragdos, identified as malachite by modern authors, Pliny (*49*, Book 37, 65–75) says that it was found also in the silver mines at Thorikos in the Laurion, where finds of Middle Helladic lead and litharge (*67*), now shown to derive from the Laurion (*30*), demonstrate the exploitation of the Laurion for lead and silver in the Bronze Age.

For the source of metals to Crete in the Late Bronze Age we have the definite evidence pointing to the Laurion for lead (*30*), and evidence suggesting that Crete used Laurion copper sources at this time comes from Caskey's excavations at Ayia Irini on the island of Kea (*68*). On Kea, which lies just off the coast of the Laurion region, Caskey uncovered large amounts of Bronze Age lead and litharge together with crucibles used for casting copper, a number of copper ingots, and many bronze artifacts. Contact with Crete is demonstrated by the Middle Minoan IA to Late Minoan IB pottery found at Ayia Irini.

Several lines of evidence suggest that the Laurion was one of a number of important sources of copper to the Bronze Age Aegean, including our lead isotope analyses. Our finding that Laurion copper was important for Crete harmonizes with our earlier finding that the Laurion was an important source of lead for Crete in the Bronze Age. Perhaps these findings explain why in the ancient Greek myth Theseus went from Athens to Crete with the tribute sent to Minos. However, we do not wish to press the few existing data too far; they rest too much on artifacts of uncertain stratigraphy and on too few comparative analyses of appropriate ore bodies, ancient slags, and the like. For Crete in particular further analyses of well-stratified bronze objects through the Bronze Age may show that different copper sources were used earlier in the Bronze Age or may reveal different patterns of usage in central Crete and eastern or western Crete. Cyprus may prove to have been of more importance as a source of copper to the eastern Mediterranean than to the Minoan or Mycenaean worlds.

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Controlling Cotton's Insect Pests: A New System

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Summary. Cotton is more heavily treated with insecticides than any other crop in the United States. In southern Texas, this heavy treatment resulted in insecticide-resistant strains of major pests which almost destroyed the industry in the late 1960's and early 1970's. An integrated insect control program based on new short-season cotton varieties and traditional cultural practices has restored production in the area. The new system has been widely implemented because it produces greater net returns by reducing the use of insecticides, fertilizer, and irrigation.

Almost 50 percent of all insecticides applied to crops in the United States are applied to cotton. As a result, most major pest insects of cotton have developed resistance to one or more of these insecticides. Some pests, such as the tobacco budworm [*Heliothis virescens* (Fabricius)] and spider mites (*Tetranychus* species) are now resistant to most of the insecticides registered for use on cotton in the United States. This is rapidly depleting the arsenal of effective insecticides for use on cotton.

An Insecticide-Induced Disaster

The decline in cotton production in northeastern Mexico and southern Texas in the late 1960's and early 1970's is an excellent example of what can happen when insect pests become so resistant to insecticides that control fails. In northeastern Mexico the area of land planted in cotton declined from more than 700,000 acres during the 1960's to less than 1000 acres in 1970 (*1*). Little cotton is grown in the region today. In the Texas Gulf Coast and lower Rio Grande Valley the area planted in cotton declined from 166,000 and 320,000 acres, respectively, in 1968 to 55,000 and 103,000 acres in 1975 (*2*). These reductions occurred because the tobacco budworm developed resistance to all registered insecticides. Although growers treated fields many times, the budworm inflicted such damage that cotton was not profitable to grow.

To understand how this situation developed, it is necessary to review the evolution of insecticide use on the crop in southern Texas, beginning in the 1930's. During this period the boll weevil (*Anthonomus grandis* Boheman), the pink bollworm [*Pectinophora gossypiella* (Saunders)], and the cotton fleahopper [*Pseudatomoscelis seriatus* (Reuters)] were the key pests of the crop. (A key pest occurs annually in a crop and must be controlled to achieve a profitable yield.) The boll weevil was controlled by calcium arsenate dust and the fleahopper by sulfur dust. Although these insecticides permitted profitable production of the crop, substantial yield losses occurred. Early planting of and early destruction of crop residues controlled the pink bollworm.

The advantage of calcium arsenate and sulfur was that they did not kill a great percentage of the insect enemies (parasites and predators) of cotton pests. As a result, outbreaks of two major secondary pests, the tobacco budworm and the bollworm [*Heliothis zea* (Boddie)], occurred only sporadically. (A secondary pest is one that attains crop-damaging numbers only when its natural enemies are decimated.)

Shortly after World War II the new synthetic chlorinated hydrocarbon insecticides, such as toxaphene, DDT, benzene hexachloride, endrin, and dieldrin became available for use on cotton. These had a spectacular effect on cotton production, since they provided almost complete control of the pest insects at an economical cost. Now cotton could be protected throughout the growing season, and 10 to 20 insecticide applications per growing season were common. Varieties were developed whose fruiting periods were more indeterminate, and irrigation and fertilizer were increased so that fruiting could be maintained longer. These practices resulted in major increases in yield.

On the surface, pest insect control in

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