Iron-Coated Quartz as a Provenance and Paleoclimatic Marker in the Rhône Delta, France

Jean-Daniel Stanley and Thomas F. Jorstad

Deltas-GLOBAL CHANGE PROGRAM
Paleobiology, E-206 NMNH
Smithsonian Institution
Washington, D.C. 20560 U.S.A.

ABSTRACT


Analyses of 430 surficial and core sediment samples in the Rhône delta and adjacent region in southern France record similar average proportions of iron-stained quartz (ISQ) on the delta plain and in the Rhône and Durance rivers (~30%). These two rivers, largely responsible for the formation of the Holocene delta, carried most of their sediment load from distal source terrains. The much higher ISQ values in sand on the delta's modern coast (to ~40%) and on older sand strand deposits (56%) subaerially exposed in the central and southern delta plain are primarily the result of post-depositional in situ formation of iron coating on quartz particles. However, some subsurface delta plain core samples with high stained-grain content (~40%) have a different origin. These layers were derived from Pleistocene and older sediment in proximal terrains, characterized by very high ISQ values (~67%), that were transported to the delta plain. Evidence indicating displacement of highly-stained material eroded from these more local source areas is quartz with high ISQ values (~50%) in the Vidourle and Gardon rivers with headwaters in adjacent uplands.

Strata with large ISQ values (~40%) in Holocene core sections are attributed primarily to periodically increased hydrological fluxes that resulted from marked paleoclimatic changes and, possibly, effects of human activity as seen in the archaeological record in southern France. Wetter climatic conditions increased the erosion and reworking of proximal ISQ-enriched terrigenous sediment to the delta. This study indicates that the stained-grain method can be used to help identify changes in sediment provenance and dispersal patterns related to Holocene paleoclimatic fluctuations and/or human activity in the Rhône delta region.

ADDITIONAL INDEX WORDS: Archaeology, climatic cycles, coastal processes, Costières, Crau, distal sediment, floods, Gulf of Lion, Holocene, iron oxides, Pleistocene, proximal sediment, quartz coating, sediment dispersal, size-sorting, source terrains, weathering.

INTRODUCTION

Deltas on marine coasts form primarily by the accretion of river sediment transported to the sea, and the erosion and removal of some of this material by wave-driven currents and other nearshore processes. The resulting deposits, accumulating by interaction of fluvial, coastal and nearshore processes, are composed of terrigenous sediment derived from both distal and proximal terrains in the river drainage basin. Most petrologic studies of deltas have emphasized the importance of fluvial transport from distal sources, a process of seaward displacement of material over time by episodic 'stop-and-go' events. These, triggered by repeated floods, typically involve episodes of erosion → downvalley and overbank transport → sediment deposition and temporary storage along the fluvial dispersal path → renewed sediment erosion and transport by subsequent flooding (MEADE, 1988; STANLEY and HAiEt, 2000). In contrast, there is much less documentation on the more direct sediment dispersal to a delta plain from proximal source terrains, such as those located in the lower river valley close to a delta's apex, on adjacent delta margins, and seaward of a delta's coast.

The present study evaluates the use of iron-coated quartz grains of sand size as potential petrologic markers to identify Rhône delta sediment derived from proximal terrains in southern France and seaward of the coast in the Mediterranean (Figure 1). As in most deltas, quartz sand particles are present in all samples of the study area. The terms iron-stained and iron-coated quartz grains are used here interchangeably, and abbreviated ISQ. This approach is based on findings in previous investigations made in different world coastal settings. Holocene delta sediments generally comprise lower ISQ proportions than older (pre-Holocene) alluvial deposits buried beneath Holocene deltaic sections (COLEMAN and ROBERTS, 1988; STANLEY et al., 2000). Also containing relatively high proportions of stained grains are Quaternary and older formations that are subaerially exposed near delta margins (MORCAN et al., 1959; STANLEY et al., 2001), and seafloor sediment on continental shelves seaward of deltas that were once subaerially exposed (SHEPARD et al., 1960; BERRYHILL, 1976; STANLEY and CHEN, 1991). Although commonly present in delta sequences, the ISQ grain component has only been used in a few instances to identify sediment provenance and dispersal from proximal terrains (STANLEY et al., 2001).
Focus herein is on ISQ distribution patterns in 13 specific surficial environmental settings in and near the Rhône delta so as to better interpret the sedimentary origin of stained grains in the subsurface Holocene record. This petrologic parameter is examined in delta cores to determine whether ISQ values remained generally constant in subsurface Holocene sections, or fluctuated markedly through time. From such an analysis of stained-grains in delta cores, it may be possible to test the following hypotheses: (1) older geologic formations in the lower Rhône River valley and other proximal regions served as sources of sediment dispersed to the Holocene delta; and (2) enriched stained-grain zones in subsurface sections, especially those that are laterally extensive, provide information on paleoclimatic fluctuations that affected Holocene sedimentation patterns in the Rhône depocenter.

BACKGROUND FOR A STAINED- GRAIN STUDY IN THE RHÔNE SYSTEM

The Rhône’s fluvial, deltaic and coastal margin systems have been extensively studied. On the basis of what is already known of its Holocene history, the Rhône delta is ideally suited for a petrologic investigation to distinguish proximal from more distal terrigenous quartz-rich sediment supply, and to identify paleoclimatic fluctuations that likely affected depositional patterns. Compared to other world deltas, the Rhône is of intermediate size. The distance between the delta apex just north of Arles and the mouth of its major distributary, the Grand Rhône, is ~42 km, and the delta shoreline is ~90 km in length (Figure 1). Margins on both the eastern and western delta plain comprise well-defined upland terrains, some with elevations exceeding 100 m.


In addition to natural phenomena, this region has been increasingly modified by human activity from the Neolithic to the present. Among recent studies discussing specific anthropogenic modifications of the delta are those of Jordà and Provansal (1990, 1996), Corre (1992), Stouff (1993), Pasqualini and Landure (1995), Morhange et al. (1998), Hensel et al. (1999), and Stanley (2000).
Table 1. Proportions of partially or fully coated iron-stained grains (ISQ values) in 184 surficial samples of modern, Holocene, and older age collected in 13 depositional environments of the Rhone delta and adjacent region. Sample localities are shown in Figure 1; distribution of ISQ values in the different environments is depicted in Figure 4.

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<th></th>
<th>Grand Rhône Margin (n = 13)</th>
<th>Petit Rhône Branch (n = 13)</th>
<th>Old Rhône Margin (n = 13)</th>
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Mineralogical analyses indicate that most sediments deposited in this delta were released by the Rhône river with headwaters in the high Alps of Switzerland. The river flows to France, first to the southwest, and then south, to the Mediterranean (Figure 1). Much of the material displaced toward the coast is derived from its major tributaries in France, including the Saône, Ain, Isère, Drôme and Durance, that drain geologically diverse terrains north and east of the delta. In addition to major distal sediment source areas in the Swiss Alps are the intermediate terrains in the Massif Central west of the Rhône river (drained by the Ardèche and other tributaries), the Jura and external Alpine chain in southeastern France (van Andel, 1955; Vivián, 1989; Arnaud-Fassetta, 2000). Much of the material transported from these regions is deposited in the delta plain as gray sand and olive gray to dark gray and dark olive gray silty mud (Kruyt, 1955; L’Homer, 1975, 1987). The near-horizontal deposits forming this low-lying, fan-shaped, wave-dominated delta (Duboul-Razavet, 1956; Galloway, 1975; L’Homer, 1992) are positioned between the apex just north of Arles and the Gulf of Lion shelf south of the coast.

Distinguishing potential sources of ISQ grains transported from proximal areas to the Rhône drainage basin is of special interest in this study. These could include highlands that comprise lithologically diverse formations near, and bordering, the delta proper (Figure 1). Proximal terrains include Pleistocene and older terrigenous strata (Costières and Crau terraces) and carbonate (Alpilles) units (Colomb and Roux, 1978; L’Homer, 1987; Jorda et al., 1990). Also recognized are the Holocene terrigenous units in the drainage basin of the southern Rhône that include reddish brown, orange and yellowish brown strata. Such deposits are identified in lithostratigraphic studies by general terms such as ‘terre rouge à cailloux siliceux’, ‘sol rouge’, ‘marnes jaunes’, ‘vases jaunes’, ‘sable gris-jaunatre’, and ‘cailloutis à matrice sableuse jaune’ (Colliot, 1904; Bonifay, 1957; Alosi and Duboul-Razavet, 1974; L’Homer, 1975; Colomb and Roux, 1978).

Petrologic analyses of these strata identify oxidized lithologies that include coated quartz and limestone particles of sand to pebble size and sandy mud matrices. Colors of individual particles include orange-brown, yellow-brown and tan. Iron oxides in paleosols, intergranular matrices and particle coatings (Alosi and Duboul-Razavet, 1974; Jorda et al., 1991b) in these older formations previously have been attributed to several phenomena, including weathering of limestone carbonate terrains and pebbles, soil development, and
staining by ground water flow in this region. In addition, it has been suggested that, after formation, some of these oxidized older materials in Provence and adjacent regions of southern France were extensively eroded, reworked and laterally displaced (Bonifay, 1957). Other potentially important proximal sediment sources of ISQ grains transported to the delta include Quaternary and older formations eroded from exposures along the Mediterranean coast and from those now submerged in the Gulf of Lion shelf south of the delta. Such Quaternary and older deposits in coastal and offshore settings are commonly ochre and tan (Aloisi and Duboule-Razavet, 1974; Labeyrie et al., 1976).

**METHODS**

**Sample Sites**

Numerous surficial samples (Figure 1) and subsurface sediment in cores were collected in the Rhone delta proper and in adjacent regions in order to (1) distinguish material of proximal origin from more distal derivation, and (2) to identify evidence of changes in provenance and dispersal through time. Stained quartz grains were examined in a total of 430 sediment samples from surficial sites and core sections. Of these, 184 samples were collected in 1999 (numbers 200–324) and in 2000 (numbers 325 to 383) from the modern (most recent to past several centuries), Holocene (past 10,000 years) and older geographic settings numbered 1 to 5 below. ISQ data from surficial samples are listed in Table 1. Averaged ISQ values and range of proportions of the surficial samples are given in Table 2. Another 246 samples, recovered in sediment cores, are of Holocene to late Pleistocene age (settings 6 and 7 below); ISQ data for these subsurface samples are listed in Table 3.

1. Modern delta plain, a total of 9 environments (n = 148), include the Grand Rhone distributary and its channel margin (n = 13 samples), Petit Rhone distributary and its channel margin (n = 6), old (earlier relict) Holocene Rhone channels still preserved on the delta plain (n = 13), delta plain proper between distributary channels (n = 29), delta lagoons (n = 32), marshes (n = 17), beaches (n = 17), coastal dunes (n = 13), and relict Holocene sand strands exposed on the southern to central delta plain (n = 8);

2. Modern Rhone and Durance rivers, the two dominant fluvial systems that transported the bulk of sediment to the delta are the Rhone, flowing from the north, and the Durance

<p>| Table 1. Continued. |
|--------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| Rhone Delta Plain Setting | Relict Sand | Rhone and Durance Rivers | Massif Central Tributaries | Others |
| Dune (n = 13) | Strand (n = 8) | Above Delta (n = 5) | Tributaries (n = 7) | Delta Margin (n = 16) | Offshore Samples (n = 8) |
| 223 | 60.44 | 256 | 53.82 | 328 | 18.36 | 316 | 53.92 | 209 | 19.28 | 1 | 9.10 |
| 231 | 30.41 | 264 | 52.86 | 329 | 20.47 | 317 | 20.25 | 210 | 94.28 | 2 | 2.30 |
| 268 | 34.87 | 295 | 48.77 | 330 | 45.14 | 318 | 43.96 | 211 | 64.43 | 3 | 29.50 |
| 290 | 34.74 | 297 | 66.98 | 331 | 39.16 | 319 | 57.85 | 212 | 47.35 | 4 | 40.40 |
| 291 | 47.93 | 298 | 51.77 | 332 | 31.21 | 320 | 67.97 | 219 | 19.05 | 5 | 23.90 |
| 349 | 22.98 | 300 | 57.28 | 350 | 42.42 | 370 | 59.11 | 249 | 75.94 | 6 | 5.50 |
| 350 | 33.87 | 375 | 55.96 | 355 | 33.87 | 370 | 59.11 | 283 | 52.52 | 7 | 11.00 |
| 358 | 38.87 | 375 | 55.96 | 361 | 34.07 | 370 | 59.11 | 284 | 64.51 | 8 | 40.00 |
| 361 | 34.07 | 370 | 59.11 | 365 | 43.45 | 370 | 59.11 | 310 | 86.23 | |
| 372 | 38.87 | 375 | 55.96 | 377 | 42.44 | |
| 381 | 91.04 | | | 382 | 71.56 | |</p>
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<th>Clear Range of Values</th>
<th>Partially Stained Average</th>
<th>Partially Stained Range of Values</th>
<th>Fully Stained Average</th>
<th>Fully Stained Range of Values</th>
<th>Partial Plus Fully Average</th>
<th>Partial Plus Fully Range of Values</th>
<th>Sand Average</th>
<th>Sand Range of Values</th>
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<td>9.0-69.9</td>
<td>42.03</td>
<td>5.6-100</td>
<td>48.4</td>
<td>0.0-75.6</td>
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<td>0.0-25.9</td>
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<td>0.32-3.06</td>
<td>20.35</td>
<td>12.4-29.1</td>
<td>37.11</td>
<td>17.8-57.3</td>
<td>51.82</td>
<td>35.8-63.7</td>
<td>10.48</td>
<td>5.4-18.3</td>
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<td>0.0-2.1</td>
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<td>6.25-54.7</td>
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<td>4.4-73.9</td>
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Table 2. Stained grain and textural data (average and range of ISQ and grain size values) for 184 surficial sediment samples collected in 13 different settings of the Rhône delta study area.
(see Figure 1, inset), its major tributary flowing from the east and joining the Rhône at Avignon (n = 5; collected in 2000); (3) **Modern tributaries**, include those that transport substantial amounts of pre-Holocene material from proximal highlands: the Garden River (or Gard), a tributary with headlands in the SE Massif Central (Figure 1, inset) that joins the southern stretch of the Rhône north of Arles; and the Vidoule River that disperses sediment from the Costières southward to and across the western part of the modern delta (n = 7; collected in 2000; see Figure 1); (4) **Older formations close to the delta margin**, comprise largely Pleistocene, Pliocene and older units that are subaerially exposed in upland terrains close to the delta plain (n = 16; collected in 1999 and 2000); (5) **Gulf of Lion continental shelf south of the delta**, includes Holocene and Pleistocene formations and palimpsest-reworked relic sediment mixes thereof (n = 8); samples collected by the Cefrom Program, Université de Perpignan and provided courtesy of Dr. J.-C. Aloisi (ALOISI, 1973; ALOISI and CHARLET, 1975; ALOISI et al., 1975; GENSOUS and TESSON, 1996); (6) **Holocene delta subsurface strata**, recovered in 19 sediment cores (n = 238; Table 3), includes 18 borings (n = 212) that range in length from 18 to 71 m (described by OOMKENS, 1970; ter KERMS, 1971), and 1 boring (PI, Piton-1; 18.3 m in length; n = 34) collected in the city of Arles just west of the Rhône (courtesy of G. Arnaud-Fassetta, 2001, personal communication); and (7) **Lower Holocene (H) to late Pleistocene (P) alluvial sections**, recovered at the base of 7 of the 19 borings listed in setting.
Analyses

The quartz-grain method used here is described by Stanley and others (2000). Proportions of quartz particles of sand size that are clear (transparent and translucent grains); partially coated (grains whose surfaces are stained to 75% by pigment), and fully stained (grains that are >75% coated) were determined for all of the 430 surficial and core samples (Tables 1, 3). Percentages of the three quartz grain categories were determined by making stereoscopic microscope counts of at least 300 quartz grains in each sample, selected randomly from the sand-size fraction (63-2000 μm). The more than 139,000 particles in the samples were examined by one operator (the second author) so as to make consistent recordings and obtain reproducible data. Quartz grain counts of sand size were examined using a Leitz Wetzlar binocular microscope using a Dyonics fiber-optic light source. Grain color was determined by using dry grains under fluorescent lighting and using standards in color charts (Munsell Color, 1975).

The averaged proportions of clear, partially stained, fully stained, and partially + fully stained grains (ISQ), and range of values in the 184 surficial samples from 13 environments in the modern, Holocene and older geographic settings 1 to 5 cited above are summarized in Table 2. The fully stained particle values in most samples is low (<3% in the different settings) and thus, in this study, ISQ value refers specifically to the percentage of partially plus fully stained grains.

Two replicate grain-size analyses were performed on all 184 surficial samples collected in the 13 environments with a Coulter Counter laser particle analyzer (LS200). From these, the following data were averaged: relative percent of sand (63-2000 μm), silt (0.5-63 μm), and clay (<0.5 μm); and mean grain size (in μm). The textural information for the surficial samples in delta and adjacent settings are listed in Table 2.

The elemental composition of grain coatings was determined for 4 representative samples in the study area: modern delta beach (surficial sample 366; Figure 2A); subsurface delta sample from an ISQ-enriched layer in the Piton core (PI) at Arles (depth of 10.65 m from the core top; Figure 2B); highland exposure (Costières) adjacent to the delta (surficial sample 381; Figure 3A); and margin of the Vidourle River in the western delta (surficial sample 298; Figure 3B). The selected grains were prepared by preferentially mounting the specimens in epoxy on standard glass microprobe discs. Each disc was then polished to expose a transverse view of the samples, thus providing maximum exposure of the stained area coating the grains. Approximately 12 grains from each of the four localities were analyzed using backscattered (BSI) and secondary (SEI) imaging with a JEOL JMA-840A high resolution scanning microscope. Analyses and elemental distribution of micro-areas for the four samples were performed by Energy Dispersive Spectroscopy (EDS), with a working distance of 25 mm at 15 KeV and a beam current of 6 nA.

Analyses and interpretations of the mineralogical, faunal and floral components comprised in the sand-size fraction in each sample will be presented in a separate study (G. Randazzo and others, in preparation). A detailed petrologic study of core Piton collected at Arles is also presently in preparation by G. Arnaud-Fassetta, C. Beaudoin, H. Bruneton and others.

STAINED-GRAIN BASELINE DATA FOR THE STUDY AREA

The colors of the coated grains typically include dark red (2.5YR 3/6), dark reddish brown (2.5YR 3/4), dark yellowish brown (10YR 3/6), and orange (5YR 6/4). Interpretation of elemental component from the EDS analyses have identified the coatings on quartz particles as iron oxides (Dr. A. Logan, 2002, Personal Communication; Figures 2, 3). This study does not address any relationship or suggestion regarding the variability in iron-stained color.

The proportion of clear grains in the 9 delta environments averages ~70% and ranges from 44 to 80%. Four ISQ grain categories are identified on the basis of natural stained-grain groupings determined from surficial and subsurface sample sets in the study area (Figure 4 and Tables 1, 3): <30%; 30-39.9%; 40-49.9%; and >50%. Salient observations pertaining to ISQ values in the 13 environments of the 7 geographic sectors discussed in the previous sections are summarized below.

(1) Modern Rhône delta plain.—Most samples from the modern delta plain proper, from the Rhône valley just north of the apex to ~5 km landward (north) of the coast, but excluding relict sand strand deposits exposed on the delta surface, are characterized by relatively low and, in some cases, moderate, ISQ values (<30%). Calculated average values for Rhône distributary channel settings are as follows: Grand Rhône channel margin = 31%; Petit Rhône channel margin = 20%; and relict Holocene Rhône branches = 24%. Low average ISQ values are noted in three delta plain environments as follows: delta plain proper = 25%; lagoons on the delta plain = 23%; and marshes on the delta plain = 27.5%. Average values on beaches are 31% (example in Figure 2A), and in coastal dunes are 39%. In contrast, most samples collected on the southern and western delta plain surface within ~5 km of the coast record higher values, ranging from 30 to >50%. Sediment samples on the western delta plain in the vicinity of the Vidourle river also contain large ISQ values, commonly 40 to >50%. Samples from relict sand strand deposits exposed on the southern and central plain are characterized by the highest average ISQ proportion (56%) on the delta proper.

(2) Modern Rhône and Durance rivers.—The average ISQ value is 31% in channel samples at the juncture of these 2 major rivers, and in the 35 km-long Rhône valley stretch between Avignon and the delta apex just north of Arles. This value is the same as that of samples along the Grand Rhône margin on the delta plain.

(3) Modern rivers flowing from adjacent highlands.—The Gardon and Vidourle, transporting sediment to the delta from proximal source terrains in the southern Massif Cen-
tributaries, comprise an average ISQ value of ~50% (example in Figure 3B). This is significantly higher than that recorded by samples in the Rhône and Durance (31%, see setting 2 above), the two much larger rivers responsible for the deposition of most Holocene delta strata.

(4) Older sequences close to the delta.—By far the largest average ISQ value (67%; range 19 to 94%) in the study area is calculated for the 16 samples collected in Pleistocene and Pliocene formations exposed in the Costières and Crau highlands adjacent to the Rhône delta (example in Figure 3A).

(5) Gulf of Lion shelf south of the delta.—Seafloor samples from the shelf south of the Rhône delta record an average ISQ value of 20%, with a range of sample values from 2 to 40%. Proportions of coated grains in the offshore are higher in sand than in mud-rich deposits.

(6) Holocene core sections.—Most of the 246 samples from the 19 borings recovered in the modern delta plain are of Holocene age and record a remarkably large range of ISQ values (from 0 to 94%). The following percentages of samples in the 4 coated grain categories are recorded: <30% ISQ occurs in 77% of all core samples; 30 to 39.9% ISQ in 15%; 40 to 49.9% ISQ in 5%; and >50% ISQ in 3% (Table 2). The distribution of subsurface strata that comprise high amounts (>40%) of stained grains (example in Figure 2B) is shown in Figures 9 and 10.

(7) Lower Holocene-Late Pleistocene core samples.—The ISQ values (range from 4 to 41%) are determined for strata recovered at the base of 6 delta cores which were previously attributed (cf. OOMKENS, 1970; TER KEURS, 1971) to early Holocene to late Pleistocene to age. The stained-grain values of samples collected at the base of 5 of these cores, identified here as H/P (Figure 9), are comparable to those of sediment carried in the modern Rhône and Durance rivers and to most Holocene deposits on the present delta plain; these proportions are much lower than those recorded for Pleistocene and older sequences exposed in the adjacent delta highlands (see...
The ISQ values range from 10 to 25% at the base of cores 110 and 114; based on overall lithology, these sections are likely late Pleistocene (P) strata. In marked contrast, analyses of basal samples in two cores, 118 (Figure 9) and PI (Figure 10), record ISQ values >40%, more firmly indicating that the base of these borings actually recovered late Pleistocene material beneath the Holocene section.

EFFECTS OF SIZE-SORTING AND IN SITU FORMATION

The average ISQ values in the Rhône delta region (~30%) are high when compared to average stained-grain proportions recorded in some other deltas, such as the Rio Grande (to ~20%; STANLEY et al., 2001) and the Ganges-Brahmaputra (8%) and Nile (~6%) (STANLEY et al., 2000). Several phenomena could account for the unusually high ISQ values of Rhône delta samples: grain size and associated size-sorting factors; in situ post-depositional development of staining on the delta plain, including biologically-generated staining; and reworking and dispersal of older iron-stained sediment from source terrains that include large proportions of particles that were iron-stained prior to their deposition in the delta.

To evaluate potential influences of grain-size and size-sorting effects on ISQ distributions, the percentages of sand, silt,
clay, and mean size were calculated for each sediment sample collected on the Rhône delta plain and adjacent surficial environments. With this information, determination was made of the relation between each of the four textural parameters and relative percentages of each of the four stained-grain parameters (clear, partially stained, fully stained, fully + partially stained quartz). A set of 192 graphs was generated with the available ISQ and textural data for each surficial sample in 12 environments (n = 176; this survey excludes the 8 offshore samples). Representative stained grain versus size graphs are shown in Figures 5 to 8.

As a first step, 16 graphs were compiled using the above-listed 4 stained-grain types versus the 4 grain-size attributes for all 176 surficial samples. The Coefficient of Determination (r^2 values) indicates a 'goodness of fit' correlation between variables, with r^2 values near 1.0 indicating a strong relationship between variables. The r^2 values determined for each of the 16 graphs range from 0.00987 to 0.220429. These low values and the random distribution patterns of data points on the graphs indicate an overall absent to weak statistical correlation between proportion of stained quartz and their grain-size attributes. Two representative graphs (Figures 5A,B) show that, when all sample data are evaluated, stained grains are independent of spec-

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<table>
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<td>Offshore Cézembre Samples</td>
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Figure 4. Distribution of ISQ (partially + fully stained grains) values for 184 surficial samples collected in 13 depositional environments of the Rhône delta study area. Triangle and associated percentage depict averaged ISQ value for each environment.
Figure 5. Graphs of surficial samples (n = 176) examined in this study (these data exclude offshore samples), showing plots of the proportion of partially + fully stained quartz grains versus (A) percentage of sand (r^2 = 0.193) and (B) mean grain size (r^2 = 0.169). r^2 = Coefficient of Determination.

Figure 6. Graphs showing ISQ values versus percentage of sand in surficial samples collected in (A) modern beach, (B) modern dunes and (C) older (relict Holocene) sand strand deposits on the Rhone delta plain. Note increasing ISQ values from A to C. r^2 = Coefficient of Determination.
specific grain size is preferentially stained and that size-sorting effects are minimal.

To more critically detect possible iron stain-grain size relationships, an additional set of 16 graphs was generated using the same 8 above-listed parameters for each of 12 specific modern environments (those shown in Figure 4, except the offshore samples). This resulted in a total of 176 graphs, in addition to the 16 cited above.

- All graphs compiled for 7 of these 12 environments (Rhône and Durance rivers sampled north of the delta apex, Grand Rhône channel margin on the delta plain, lagoon, marsh, dune, relict Holocene sand strand, delta margin settings) show no statistical correlation between ISQ values and sand-size (Figures 6A-C, 7A, and 8).
- Stain versus size graphs for the other 5 of the 12 environments (relict Rhône branches, interdistributary delta plain, some coastal facies) record a weak correlation (Figures 7B, C). Of these, only several show a moderate correlation: those depicting stained quartz versus relative percentage of clay in some near-coast environments and along the Petit Rhône channel; and those depicting stained grains versus relative percentages of clay and mean size of samples. These latter suggest size-sorting effects, probably the result of transport processes that preferentially affect the distribution of finer-size fractions (clay and silt) in the 2 delta environments.

The survey indicates that, for the most part, proportions of ISQ values are independent of, or only poorly related to, specific grain-size parameters of sediment transported to the Rhône delta. This implies that factors other than sediment transport, depositional mechanism, and size-sorting more directly influenced the ISQ values recorded in the study area.

We suggest two other more probable causes for the observed stained-grain variations and distributions: (1) post-depositional and in situ formation of iron-coated particles locally in the delta; and (2) deposition of older reworked quartz grains that were already iron-stained in their source terrains.

Figure 7. Graphs showing ISQ values versus percentage of sand in surficial samples collected on the delta plain, including the (A) Grand Rhône distributary channel, (B) old (relict) Rhône distributary branches, and (C) delta plain proper. $r^2$ = Coefficient of Determination.
prior to their transport to the delta. With respect to (1), conditions in some delta environments appear to have favored the formation of iron coatings on quartz particles after their deposition on the delta plain. Conditions leading to staining commonly involve the availability of iron in various forms, and chemical and/or biological alteration on grain surfaces that favor addition and/or removal of an iron film on particles (cf. Van Houten, 1968, 1973; Gadel and Pauc, 1973; Dorn, 1998; Newman, 2001; Yapp, 2001).

The processes that resulted in iron coating on quartz are not specified here. It appears that some post-depositional phenomenon may have resulted from the somewhat lower-than-average ISQ values of partially + fully stained grains recorded along the Petit Rhône margin (20%), in delta lagoons and wetlands (23%), and seafloor settings south of the delta margin (20%). This finding is in marked contrast to the much higher average values (39–56%) recorded in the sand-rich coastal (dune, relict sand strand) environments. The progressive increase of ISQ values (Figures 4, 6A–C), from modern beach (31%); to dune (39%) to older (relict) strand sand units on the southern to central delta plain (56%), indicates relatively recent Holocene to possibly ongoing formation of iron oxide films on particles in these predominantly coastal sand exposures. The absence of any apparent relation between grain size and staining as shown in Figure 6 is an additional, albeit indirect, indication for in situ formation of the higher than average ISQ values in these 3 environments, rather than the result of transport process.

### ISQ VARIATIONS AS A FUNCTION OF PROVENANCE AND DISPERAL

The ISQ distribution data provide evidence that a large proportion of particles transported by rivers to the Rhône delta was already partially or fully stained prior to reaching the modern delta plain environments. Sediment provenance of older stained material, rather than transport process per se, appears to be a major factor controlling the marked variation of ISQ values recorded in the study area. The large proportions of clear, non-stained grains and moderate ISQ values (average of ~70%) in most delta samples correlates well with other petrologic attributes of Rhône delta deposits, such as the composition of heavy and light mineral assemblages (Van Andel, 1955; Aloisi and Charlet, 1975). Considered together, these findings support the contention by most workers that the bulk of sediment forming the modern delta plain surface was derived from distal (Alpine, Jura) and intermediate (Massif Central) sources. Significant in this respect is the similarity of average ISQ proportions (~24–31%) and stained-grain colors in samples of the modern lower Rhône and Durance channels, lower Rhône valley between Avignon and the delta apex at Arles, Grand Rhône channel, relict Rhône distributaries on the delta plain and other environments on the delta plain proper (Table 2 and Figure 4).

In marked contrast with the above findings are the ISQ values (67%) in proximal subaerial exposures of older formations in upland terrains of the lower Rhône region (average 67%) and in the rivers that drain them (average 50%). These are much higher average values than on much of the modern delta plain and in Rhône-Durance rivers (~31%) (Figures 1, 4). This finding indicates that, at least in recent time, there has been a much smaller amount of sediment transported to the delta from proximal sources, relative to material derived from intermediate and distal terrains. To determine whether the present ISQ delta plain pattern remained constant, or fluctuated, during the Holocene requires a review of stained-grain data obtained from subsurface samples (Table 3). The core data show that ISQ content in the study area is highly variable in time and space (Figures 9, 10).

A likely explanation for this is a fluctuating sediment input...
to the delta from proximal sources (southern France, Provence, offshore). This possibility has been suggested recently by others (Arnaud-Fassetta and Provansal, 1999; Arnaud-Fassetta, 2000). Delta sediments with enriched ISQ content (>40%) are attributed here to two distinct proximal sources, one terrestrial, the other seaward of the Rhône mouth. The large ISQ values of samples at the delta's modern coastal margin (Figures 1, 2A) result, at least in part, to sediment derived from presently submerged sources on the continental shelf south of the delta. Yellowish to ochre sands and silts have been recovered between the coast and outer shelf in the Gulf of Lion (cf. Aloisi and Dubouy-Razavet, 1974). However, our analyses here indicate that ISQ values in surficial offshore samples are low (average 20%), and that samples in 5 cores (112, 114, 116, 117, 118) recovered along the coast also record only low to modest ISQ values in subsurface sections (Figure 9). We propose two origins for delta coastal margin sediment that comprise high ISQ values: post-depositional iron-stain formation in Holocene to modern dune settings as indicated earlier, and dispersal of sediment from eroded older coastal exposures in the southern France region, as yet undetermined, that already comprise a high ISQ content and are moved along the coast. Coast-parallel transport is a response to the high-energy wave regime in this region (cf. Corbe, 1992; L'Homer, 1992; Bruzzi and Provansal, 1996; Durrieu de Madron and Provansal, 1996; Suárez and Provansal, 1996; Estoufrel et al., 2001). An important portion of this reworked iron-stained material is driven landward onto the delta's nearshore margin primarily by strong winter storms (Bruzzi and Provansal, 1996).

An origin other than the above, however, is needed to ex-
plain the large ISQ variations recorded in subsurface sections of Holocene non-marine sediment, especially those in borings collected in the northern and central sectors of the Rhone delta. Most ISQ sample values in subsurface sections are <30%. Available core data indicate that subsurface strata with large (>40%) ISQ values in this part of the delta constitute only a relatively small part of the total Holocene section, and it appears that these layers formed only periodically on the delta plain (Figures 9, 10). Of note in this respect are the relatively thin (<30 cm), enriched ISQ layers (values >40%) identified in 7 of the 19 cores examined; these high ISQ samples account for a total of 22 (or 9%) of the 246 samples examined. Although few in number, the stain-rich strata are widely distributed in the delta’s subsurface sections (Figure 9): they occur in delta plain cores located close to adjacent uplands (core 129), in the central delta plain (for example, cores 108 and 110) and, locally, along the coast (cores 105 and 106).

STAINED-GRANm VARIATIONS AND CLIMATIC FLUCTUATIONS

Distinct stained-grain changes occurred in the Rhone delta through Holocene time, as recorded in cores of the north and central plain. We attribute these variations to two factors: paleoclimatic fluctuations, especially those involving changes in precipitation, and human activity, including increased deforestation and intensified, or modified, agriculture practices that affected Provence and adjacent regions of southern France (JORDA and PROVANSAL, 1990, 1996; JORDA et al., 1991a,b; PROVANSAL, 1995). Since they are often closely associated, it is difficult to distinguish between the relative effects of climate change and human activity (cf. BUTZER, 1982; PROVANSAL, 1995). Nevertheless, previous studies have shown that climatic conditions in this region fluctuated markedly between the early Holocene and the present, and that such changes altered the flow and sediment discharge patterns of the Rhone and Durance, smaller rivers and the tributaries flowing from the southern Massif Central (VIVIAN, 1989; TRICART and BRAVARD, 1991; BRAVARI et al., 1992; FRUGET, 1992; RODITIS and PONT, 1993; ARNAUD-FASSETTA and PROVANSAL, 1999; PROVANSAL et al., 1999; ARNAUD-FASSETTA, 2000).

To help interpret the delta’s Holocene depositional evolution, it is useful to determine whether the large fluctuations of ISQ values recorded in Holocene delta subsurface sections are related in some manner to climatic change. Presently, transport of Rhone and Durance sediment from intermediate and distal source terrains prevails, with deposition in the delta region of relatively low (~30%) ISQ average proportions (Table 1). At times in the past when climate was similar to the present one, available stained-grain data for core sections show comparable ISQ values (<30%). It appears that major sediment input of intermediate and distal provenance masked the supply of material dispersed from more proximal sources, with its enriched ISQ values (>40%).

We envision a considerably different depositional regime in the past, however, when increased precipitation, or changed anthropogenic activity, or both, affected the southern France-Mediterranean region. During wetter periods, proportions of sediment input derived from sheet-flow scour and erosion of source terrains closer to the delta increased substantially (cf. PICHAud, 1995). At such times, a larger amount of sediment characterized by high ISQ values were dispersed from adjacent uplands (Costières, Crau and others), southern Rhone valley sectors, southern Massif Central and from intermediate (including the Massif Central, western Alps) source terrains. Thus, this periodically increased influx of more proximal material (ISQ values >40%) would have masked the fluvial sediment of more distal derivation with lower ISQ values (~30%) that was transported more frequently by the Rhone and Durance systems to the delta.

If the above scenario is correct, it should be possible to correlate strata with enriched ISQ values in the 18 delta borings numbered 105 to 129 (shown in black in Figure 9) with phases of periodically marked hydrological fluxes and increased deposition. However, some enriched ISQ subsurface intervals were probably not detected in this investigation because: (1) some of the original high ISQ layers may have been removed by natural factors (erosion by lateral migration of distributary channels) during development of the delta; and (2) our sampling intervals may have missed such layers that can be quite thin (core depths listed in Table 3). Thus, strata that include high ISQ values cannot be correlated regionally in all cases. Moreover, the relatively small number of radiocarbon-dates presently available for these and other borings in the region (cf. OOMKENS, 1970; TER KEURS, 1971; LABEYRIE et al., 1976; JORDA et al., 1991b; PROVANSAL, 1985; GENOUS and TESSON, 1996; MORHANGE et al., 1998; ARNAUD-FASSETTA, 2000) precludes establishing a firm, well-defined stratigraphy based only on enriched ISQ core layers.

For these reasons, it is useful to investigate results of the ISQ study of the subsurface section in core PI collected in Arles: there are several radiocarbon dates for this boring, and numerous samples were collected at close interval. The core includes five zones characterized by high (>40%) ISQ values (consisting of 8 of the 34 examined samples). From core top (elevation of 5 m above m.s.l.) to base, these horizons (<20 cm thick) occur at depths of ~0.4–0.7 m, 2.96 m, ~3.5 m, 10.30 m and ~18.10–18.3 m. These strata can be used for an ISQ-paleoclimate correlation (Figure 10). Climate-related depositional phases have been defined independently in earlier studies by using diverse criteria, including lithology (JORDA et al., 1991b), fossils (BAZILE et al., 1986; CAMBON et al., 1997), geomorphology (PROVANSAL, 1995), and archaeology (JORDA and PROVANSAL, 1996). Five periods, some involving markedly altered sedimentation patterns, occurred during the Neolithic, Bronze Age, Iron Age, post-Roman to end of Middle Age, and Little Ice Age (Figure 10).

Preliminary findings that associate some high stained-grain signals for the Rhone delta with dated paleoclimatic phases are encouraging. For example, three of the four upper enriched ISQ samples of Holocene age in core PI (Figure 10, layers a,c,d) appear to match periods of altered terrigenous sedimentation identified between ~5000 years ago and the present in Provence and the southern Alps (JORDA and PROVANSAL, 1996). Layer (b) remains less clearly defined stratigraphically. Additional radiocarbon-dating information for
the lower part of the boring is needed to help confirm that the two enriched ISQ samples (>40%) at the core base layer e) are of late Pleistocene age.

This ISQ-based stratigraphy in the Rhône delta warrants further testing with more tightly sampled and well-dated cores.

CONCLUSIONS

Most surficial samples in Holocene core sections of the modern Rhône delta plain are characterized by high proportions of clear quartz grains (~70%) and moderate proportions (~30%) of partially plus fully iron-coated quartz particles. The ISQ values recorded on the modern delta plain are similar to the material presently carried by the Rhône and Durance, the major rivers that transported most material to the delta. These findings support the conclusion of earlier workers that most surficial and subsurface sediments forming the delta plain during the Holocene were derived from intermediate and distal source terrains in the Massif Central, Alpine chain in SE France, Jura and high Swiss Alps. Of special interest in this investigation are the higher ISQ values (>40%) recorded in some modern fluvial systems and deltaic environments, and in upland settings located close to the delta proper.

Large proportions of stained quartz (to >40%), for example, are recorded on the delta's modern coast, especially in sand-rich dunes. Some of these enriched ISQ sands were derived from the landward reworking of older iron-coated quartz-rich sediment eroded from coastal exposures and perhaps also from continental shelf terrains in the Gulf of Lion south of the delta. However, even higher average ISQ values (56%) are obtained for the relict sand strand deposits of mid to late Holocene age exposed on the central to southern delta plain. This marked increase of ISQ value in these progressively older coastal sand-rich sediment on the delta plain surface suggests post-depositional in situ formation of additional iron coatings of quartz during the Holocene.

A different origin is proposed for the high stained-grain content (>40%) recorded in some subaqueous sections of the modern delta proper: periodic reworking and preferential transfer from proximal sources of sediment that was already characterized by high ISQ values. This displacement includes material eroded from Pleistocene and older geologic terrains exposed in the lower Rhône uplands close to the delta. Such reworked sediment is generally characterized by much higher average ISQ values (67%), and is readily distinguished from most modern and Holocene deposits forming the Rhône delta plain (~30%). Evidence favoring the displacement of eroded, highly stained material from proximal highlands to the delta is the high (50%) average ISQ value that characterizes the present sediment load of the Vidourle and Gardon rivers. Flowing from the southern Massif Central, these shorter rivers carry large proportions of reworked sediments with high ISQ values, releasing these as overbank deposits in their lower valleys and on the Rhône delta plain.

We postulate that the Gardon, Vidourle and other tributaries that flow from the proximal upland terrains transported increased proportions of ISQ-enriched sediment to the delta primarily during periods of higher precipitation. It appears that some subsurface strata with large ISQ values (>40%) in Holocene core sections of the central and northern delta can be correlated with increased hydrological fluxes that were periodically induced by paleoclimatic phases affecting southern France. Such wetter climatic conditions and, in some instances, associated human activity, resulted in increased erosion and displacement of terrigenous sediment with high ISQ values to the Rhône delta. It was during these episodes that material from local (including Costières, Crau) and intermediate (Massif Central) terrains masked the fluvial input from more distal Rhône and Durance sediment sources.

The stained-grain method holds promise as a practical petrologic and stratigraphic tool to help identify effects of paleoclimatic fluctuations and periodically altered sediment provenance and depositional patterns. This new information will enhance our understanding of Rhône delta development during the Holocene.

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LITERATURE CITED


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