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Accumulation of As, Pb, and Cu Associated with the Recent Sedimentary Processes in the Colorado Delta, South of the United States-Mexico Boundary

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Abstract Sediment cores from the Colorado River (CR) remnant delta were used to assess the changing sedimentation and pollutant deposition processes in response to extensive human manipulation of the river. The cores are formed of alternating layers of clays and silts, with isolated sandy horizons. The clayey units are interpreted as periods of flood flows into this low gradient and meandering estuary after dam construction in the United States. The geochemistry of these sediments is particular because of the association of MnO with CaO rather than with the Fe_2O_3-rich clays. Past pollution of the CR delta by As, and probably also Pb and Cu, is recorded in some cores. Enrichment factors (EFs) >1 for these elements and their statistical association suggest anthropogenic inputs. The most likely sources for these element enrichments (especially As) are the arsenate-based pesticides used intensively in the area during the first half of the 20th century. The transport of these elements from the nearby agricultural lands into the present river reaches appears to have been driven in part by flooding events of the CR. Flushing by river and tide flows appear to be responsible of a lower pollutant deposition in the CR compared to the adjacent Hardy River (HR). Arsenic in the buried clay units of the HR has concentrations above the probable toxic effect level (PEL) for dwelling organisms, with maximum concentrations of 30 μg g^{-1}. Excess ^{210}Pb activities ($^{210}Pb_{xs}$) indicate that fluxes of this unsupported atmospheric isotope were not constant in this estuarine environment. However, the presence of $^{210}Pb_{xs}$ does indicate that these sediments accumulated during the last ~100 years. Aproximate sediment ages were estimated from the correlation of historic flooding events with the interpretation of the stratigraphic record. They are in fair agreement with the reported onset of DDT metabolites at the bottom of one core.

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Estuaries are natural sinks of dissolved fluvial ions in the mixing zone with seawater (Hunter 1983). A drastic modification in the magnitude of river flow may affect the rates and paths of several chemical, biological, and geochemical processes in an estuary. During the last hundred years human intervention has modified water and sediment flows to the Colorado River (CR) delta. Since 1932 the water crossing the Mexican-U.S. boundary was significantly reduced by the construction of the Hoover Dam, and during 1960–1979 practically no water flowed across the southern international boundary (SIB) due to filling of the Glenn Canyon Dam. Water surplus above the official quota for Mexico was only recorded until 1980, after Glenn Canyon

dam was filled and excess water from unusual rainfall was sporadically released into the delta region (Fig. 1). The inflow of agricultural, geothermal, and municipal sewage discharges in addition to the high evaporation rates make the Colorado delta estuary an area that is especially sensitive to the accumulation of inorganic and organic pollutants. This area receives the agricultural return waters from the Mexicali Valley, which contain high concentrations of agrochemicals, as suggested from the 70,000 tons of fertilizers and 400,000 L of insecticides used alone during the 1990–1991 cycles in the Mexican part of the river basin (DGE 1993). Unlike the CR, the adjacent Hardy River (HR) derives its headwaters not only from agricultural returns and sewage, but also from the wastewater of the Cerro Prieto geothermal wells (Fig. 2). This geothermal power station started to operate in 1973 to export energy to the United States. High Hg, As, Sb, and B levels can be found near geothermal installations (Bargagli et al. 1997). Air and water pollution by Hg from the Cerro Prieto geothermal field has been reported (Acosta y Asociados 2001, in Yarto-Ramirez et al. 2004). Nevertheless, mean Hg levels in clam and fish tissue throughout the wetlands were below the 1.0 mg kg^{-1} FDA safe limits (Gutiérrez-Galindo et al. 1988). In the nearby Salton Sea developmental defects in fish embryos have been suspected to be related with pollutants from municipal sewage and agricultural discharges (Matsui et al. 1992). An increased exposure to Se, B, and Zn in birds from the delta region was observed by Mora and Anderson (1991), who did not find evidence for Zn, Cd, Cu, or Cr in liver tissue above the known thresholds for biological effects in birds. According to García-Hernández et al. (2001), only Hg and Cd slightly exceeded the potential toxic threshold in few biota samples from the delta region. Shumilin et al. (2002) suggested that no pollution by metals occurs in the upper Gulf of California. However, their results on a single studied core indicate a twofold increase in As concentrations toward the older (bottom) part of their core, compared with its top half. Because of the strong influence of tidal currents and the absence of freshwater flowing into the ocean, it is thought that any pollution was most likely recorded in the freshwater-estuarine unit of the delta region, rather than in the marine areas.

The present research is based on the hypothesis that the major fluctuations in river flow during the 20th century were recorded in the sediments from the Colorado delta. The main flooding events registered after 1935 and 1980 (Fig. 1) carried a significant load of sediments via the flushing of previously deposited material from the upstream reaches and dams. The sedimentation of the transported fine-grained particles partially took place in the lower reaches of the delta, where hydrodynamic energy was dissipated by the low gradient and meandering streams. The aim of the present study is to assess the historic variation in sedimentation and sediment geochemistry in the lower delta, as well as the deposition of inorganic pollutants, including As, Pb, Cu, Cr, and Hg, which are known to be potentially toxic.

Materials and Methods

The Study Site

The study area is located near the junction of the CR and HR south of the Baja California State capital city of Mexicali, adjacent to the U.S.-Mexican international

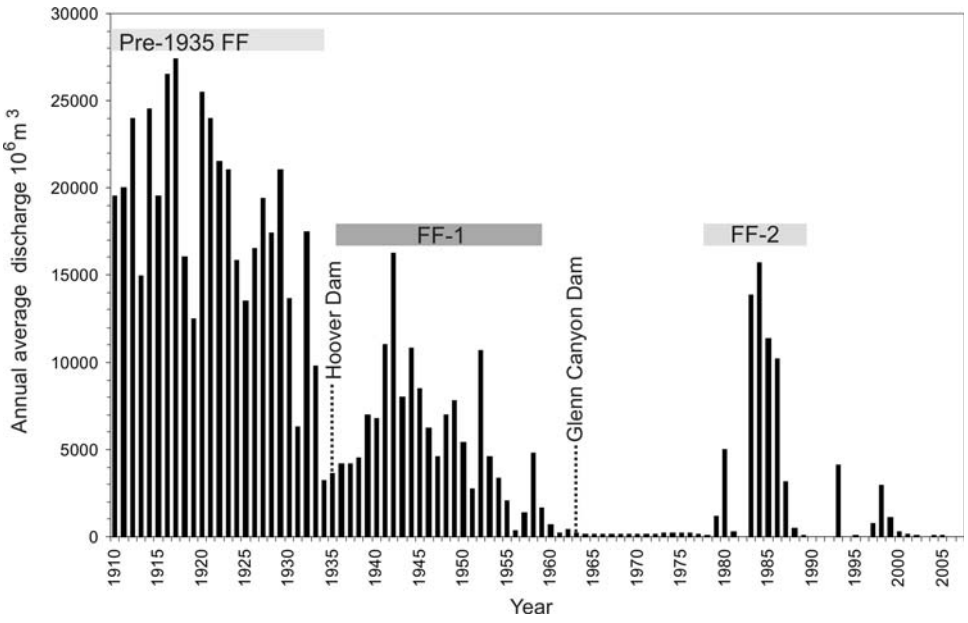

Fig. 1 Estimated yearly average discharge of the CR at the SIB, 1910–2007. Data for 1910–1949 from Cohen et al. (2001); measured discharge 1950–2007 from International Border Water Commission (2008)

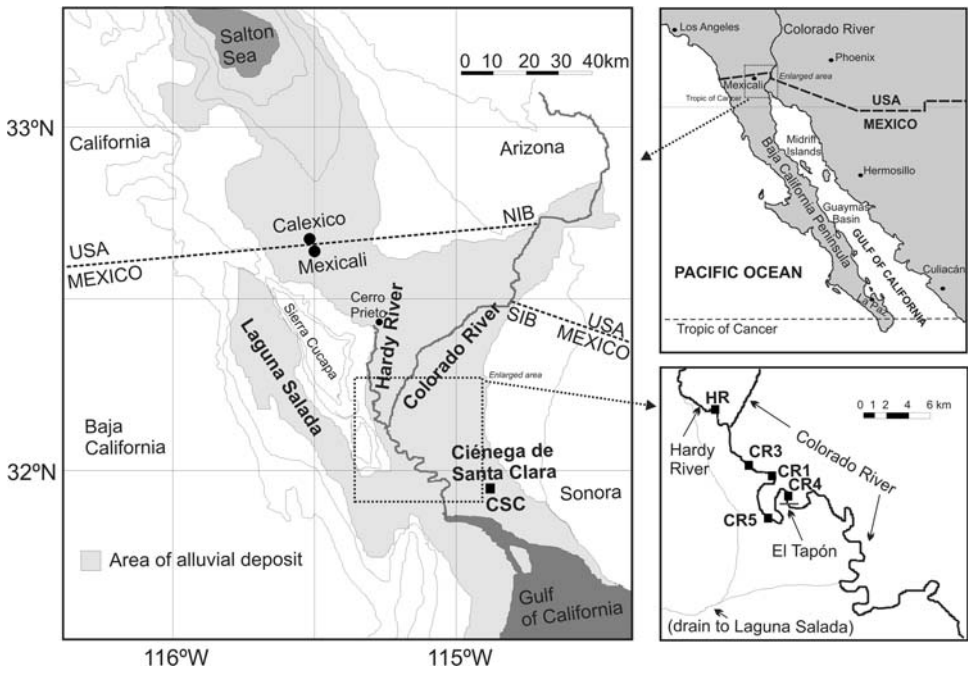

Fig. 2 Lower CR hydrological basin and location of cores HR, CR, and CSC in the CR delta

boundary (Fig. 2). The climate in this region is dry, with high evaporation rates and low annual rainfall (<100 mm [Palacios-Fest 1990]). In its delta, the CR has a low gradient, meandering streams with no firm channel with oxbows and backwaters, vestiges of former channels (Sykes 1937). The HR, which is a former channel of the CR, runs 24 km to its mouth at the CR, discharging primarily agricultural drainage from the Mexicali Valley and effluents from the Cerro Prieto geothermal power station (Sykes 1937; Valdés-Casillas et al. 1998). The CR was partially protected from the influence of tides and marine water by a small dam called "El Tapón," built in 2001 (Fig. 2). Salinity values in the studied reaches varied throughout the sampling year between 2 and 7 g L^{-1}. It has been discussed that during flood years, water flowed toward the ocean (Glenn et al. 2007) but probably also into nearby Laguna Salada (All 2006). In 1981 when Glenn Canyon Dam was filled, water from flood events was again released into the delta region, causing changes in the geomorphology of the delta channel and river system (Glenn et al. 1996). Since 1977, untreated return flows from the United States have been discharged into the eastern part of the delta, creating the Ciénega de Santa Clara (CSC) wetland (Fig. 2). Less significant agricultural returns from Mexico and the occasional flooding of the river have created or partially restored other small wetlands in the region.

Sampling

Six sediment push cores measuring 29–145 cm were sampled in 2006 and 2007 from the CR delta, including reaches of the HR and CR, as well as CSC (Fig. 2). Cores were retrieved from the river bottom with acrylic tubing by divers or directly from the boat at sites which local residents report to have been historically covered by water. These sites were located near the center of the watercourse. The cores retrieved along the riverbed from north to south are HR1, CR3, CR1, CR5, and CR4. The latter core was retrieved ~100 m north of "El Tapón." The cores were sliced and sectioned at 0.5- or 1.0-cm intervals and lyophilized.

Analytical Techniques and Quality Assurance

Grain size distribution was determined with a HORIBA LA910 laser particle size analyzer (Daesslé et al. 2002). Major and trace elements were analyzed with a Philips PW 2400 XRF. Copper was determined with a Spectroflame-D ICP-AES after total digestion of the samples in sealed PTFE vessels at 180°C using a combination of HNO_3, $HClO_4$, and HF. Recoveries for each element were determined by analyzing replicates of the USGS reference sediment MAG-1. Most elements had recoveries of 92–110%, except for Ce, Cu, Pb, and Zr (82–88%), and Cr, La, and Mo (121–125%). Recovery for As was 98%. Analytical precision was estimated through analyses of randomly selected replicate samples, and the error was $<\pm 1\%$ for the major oxides and $<\pm 10\%$ for the trace elements. Mercury was determined in eight samples from core HR1 and four samples from CR1 by cold vapor FIMS after aqua regia acid dissolution, using USGS GXR-4 reference material for quality control (recovery, 106%). Loss on ignition (LOI) was determined by combusting samples of known weight

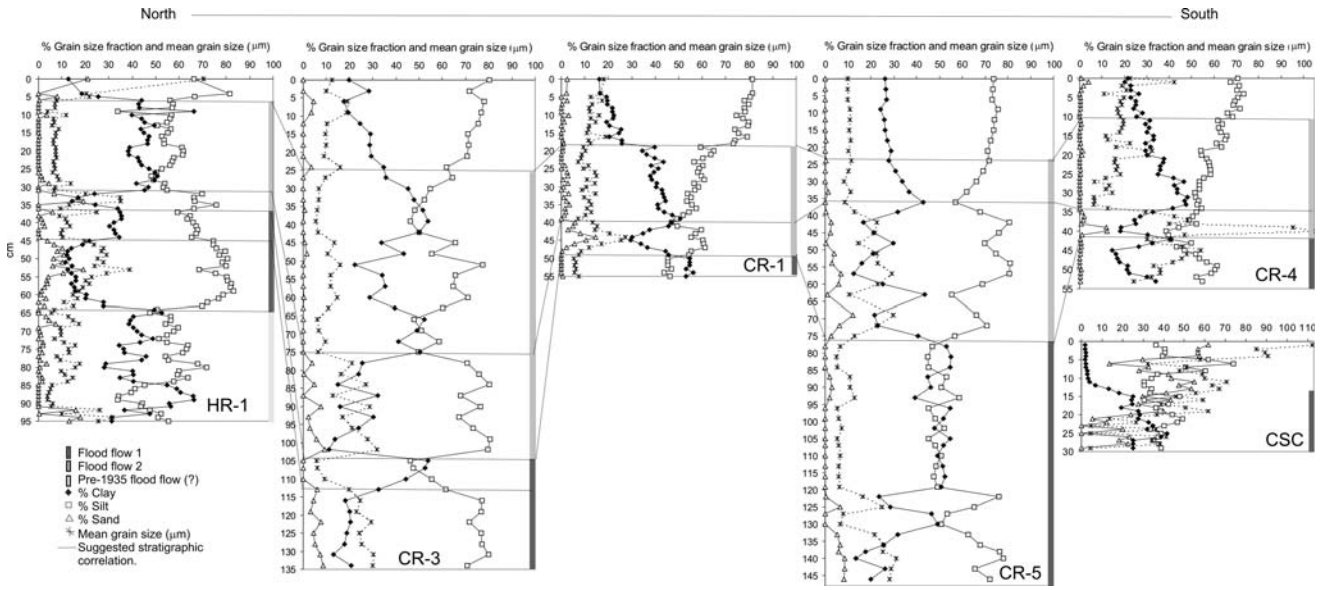

Fig. 3 Downcore distribution of percentage clay, silt, and sand and median grain size (μm) in cores from the CR delta. The suggested flood flows after dam construction are identified on the right margin of each core. The generalized stratigraphic correlation is based on grain size variability and Al_2O_3 downcore distribution

(±0.01%) at 1030°C. To test if sedimentation rates could be determined in these fluvial sediments using the ^{210}Pb method (Appleby and Oldfield 1978), eight selected samples from cores CR1 and HR1 were analyzed with a Canberra hyperpure germanium gamma detector. The upper 5 cm of core HR1 was saturated with water and thus not enough sediment was available for radiometric analyses of the surface. The detector was calibrated for energy and efficiency, and corrections for sample transmission and background activities were achieved using appropriate geometries of various densities and Isotope Products certified standards. Sediment dry density was calculated and the samples were stored for 21 days in gas-sealed containers to ensure equilibrium between ^{226}Ra and its daughter ^{222}Rn. Supported ^{210}Pb was determined from the activity of ^{214}Pb (351.9 keV), which is a short-lived daughter of ^{226}Ra (Joshi 1987). Subtraction of the supported from the total ^{210}Pb (46.5 keV) gives an estimate of the $^{210}Pb_{xs}$ (atmospheric) present at different depths in the sediments. The activity of the anthropogenic isotope ^{137}Cs (661.6 keV) was also counted in these samples.

Results

Grain Size Distribution and LOI

The relative abundance of clay- and silt-sized sediments throughout the cores is used as a tool to interpret changes in the sedimentation environment in the Colorado delta (Fig. 3). It is suggested that clay-sized material was the dominant sediment component carried by the river toward the Gulf of California before the free flow of the river was interrupted. Results for cores from the upper Gulf of California (Daesslé et al. 2004a) show a massive reddish-brown clay layer below an ~15-cm-thick horizon of sandy silts. Except for CSC, all the cores studied have alternating layers of clay-rich (max 66% clay) and silt-rich (max 81% silt) sediment, with some silt layers accompanied by the presence of sand (max 21% in cores HR1 and CRs). The increase in clay abundance is accompanied by a concomitant decrease in silt throughout the cores, indicating that these two sedimentary facies have different sources and/or were subject to different hydrodynamic conditions. Sand abundance, when present, covaries with that of silt. The varying thickness of the different layers and the variations in grain size in some of these horizons suggest that sedimentation rates and processes were not the same along the riverbed. The clay layers commonly have a brown to reddish-brown color. After combusting the samples at 1030°C, the intense red Fe-oxide color of the clays can clearly be distinguished from the gray and brown colors of the silt layers.

The downcore trends in grain size and the variability of Al_2O_3 concentrations (Figs. 3 and 4) suggest a generalized stratigraphic correlation for the cores. HR1 is the core that most clearly shows the periods of regular clay deposition, with sudden interruptions, abruptly shifting to sandy-silt-dominated layers. Only the bottom half of CR5 shows a fairly constant period of clay deposition similar to those observed in HR1. Due to its location outside the current river reaches and its proximity to the Sonora Desert, the

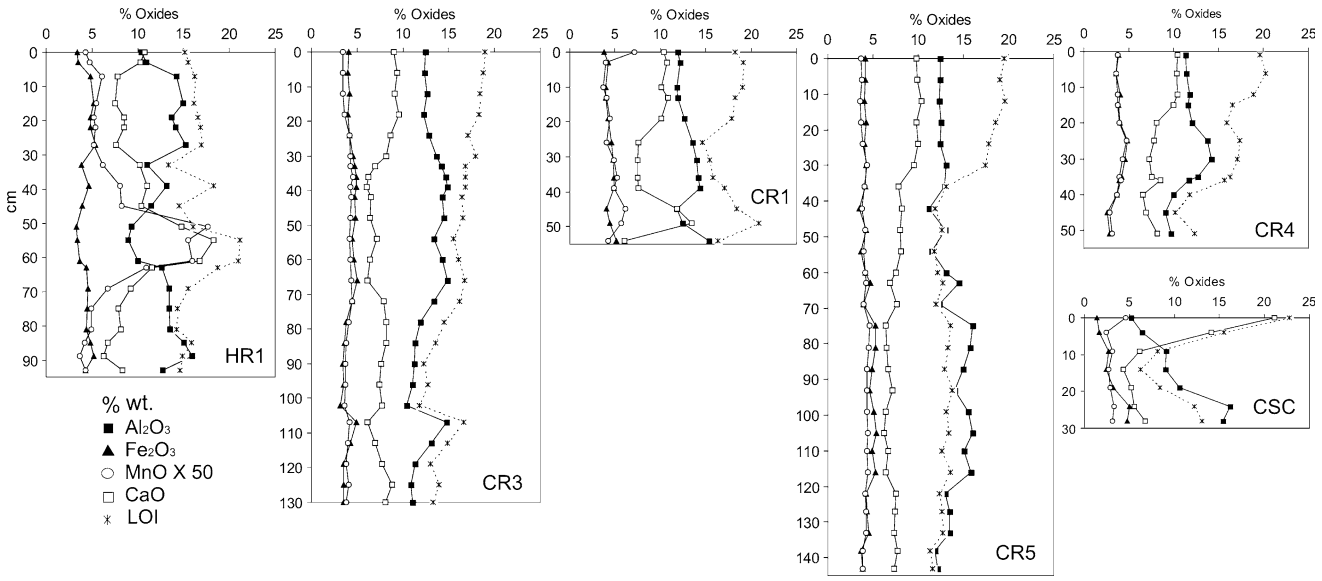

Fig. 4 Major oxide concentrations (Al_2O_3, Fe_2O_3, MnO, and CaO) and %LOI in cores from the CR delta

short core from CSC has an 11-cm-thick top layer where clay is almost absent, and sand and silt are dominant (Fig. 3). Below this sandy layer, however, an abrupt shift to clay-rich sediment is also seen, which probably represents the time when river sedimentation definitively stopped at CSC.

The percentage LOI (%LOI) gives an approximate measure of the abundance of carbonate minerals and organic matter in the sediments. Thus it correlates positively with the concentration of CaO. The %LOI does not vary much throughout the cores and is usually only 10–20% (Fig. 4). All cores, except HR1 show an increase in %LOI toward their tops. This surface enrichment starts at the same depth where the topmost silt layer begins. These enrichments in %LOI are indicative of a diagenetic front and/or an increase in biogenic deposition in most recent times. Additional peaks in %LOI are found in core HR1 within a sandy-silt layer at 55 cm, in core CR1 just below a silt layer at 50 cm, and at the bottom of core CSC.

Downcore Element Distributions and Enrichment Factors (EFs)

Downcore distributions of bulk Fe_2O_3, Al_2O_3, CaO, MnO, and LOI are given in Fig. 4, and those for As, Pb, Cu, and Cr in Fig. 5. The ranges of concentrations of potential contaminants in the Colorado Delta (As, Cr, Cu, Hg, Pb, and Zn) are given in Table 1. While the distributions of Fe_2O_3 and Al_2O_3 are similar to those for clay abundances (reflecting the composition of clay minerals), the downcore distributions of CaO and MnO mirror those for Fe_2O_3 and Al_2O_3, and are similar to the distribution of %LOI. A significant correlation between Ca and Mn in sediments from the upper Gulf of California (derived from the CR) was already observed by Daesslé et al. (2002). In this region Mn may be part of calcium carbonate minerals, rather than of the dominant detrital sediment. However, a diagenetic enrichment of MnO in the top sections of cores CR1 and CSC cannot be disregarded (Fig. 4). Most of the trace elements studied follow the downcore distribution of Fe_2O_3 and Al_2O_3 and, thus are enriched in the clay facies compared to the silt facies (Fig. 5). To assess if the relatively higher concentrations of As, Pb, and Cu, especially in the top half of core HR1, could have anthropogenic sources, EFs were calculated (Covelli and Fontolan 1997). This was done using the element/Al_2O_3 ratios in the samples, compared with the baseline ratio in the bottommost sample of core HR1, thought to be the oldest sediment available. EFs > 1 for As are seen in all the cores but are the highest in cores HR1, CR1, and CR4 (Fig. 6a); for As the EF was > 3 in core HR1. These cores also show EFs > 1 for Pb and Cu (Fig. 6b and c). To assess if MnO plays an important role in the enrichment of As in the sediments, the EFs for Mn were calculated, comparing with the baseline MnO/CaO ratio (Fig. 6d). This was done based on the correlation of MnO with CaO rather than with Al_2O_3. Only a partial overlap between the EF peaks of As and MnO was seen.

Factor Analysis (FA)

Varimax rotated FA was carried out using the bulk chemical composition (except Hg), %LOI, and % clay as variables. Multivariate statistics was used in order to further assess the distinct sediment geochemical components and to fingerprint possible events of enhanced pollutant

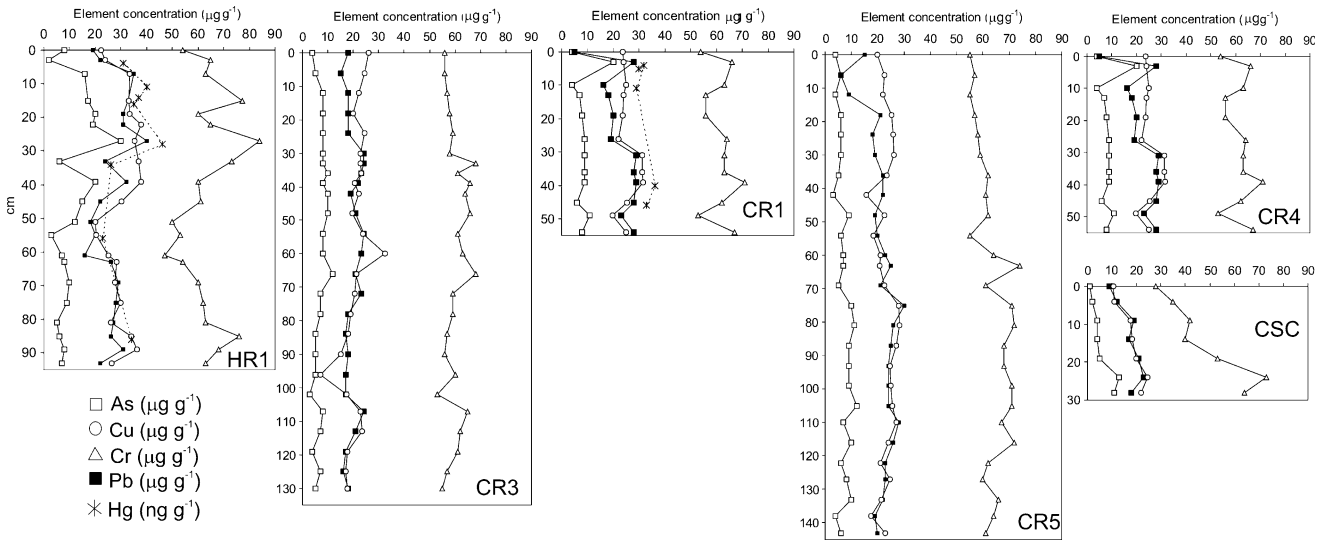

Fig. 5 Trace element concentrations (As, Pb, Cu, Cr, and Hg) in cores from the CR delta

Table 1 Comparison of element concentrations in sediment cores from the Colorado Delta with guideline toxicology levels and data published for CR sediments and suspended load at NIB in 2001, as well as from the upper Gulf of California

Site/effect level[a]	As (μg g^{-1})	Cr (μg g^{-1})	Cu (μg g^{-1})	Hg (μg g^{-1})[b]	Pb (μg g^{-1})	Zn (μg g^{-1})[c]
Lowest effect level[1]	6	26	16	0.2	31	120
Probable effect level[2]	17	90	197	0.486	91.3	315
Consensus-based TEC[3]	9.79	43.4	31.6	0.18	35.8	459
Consensus-based PEC[3]	33	111	149	1.06	128	121
Sediment from three cores in Lake Powell, Utha[4]	7–13	38–75	20–38	0.012–0.134	16–40	59–217
Sediment from Navajo Canyon, AZ[5]	3.7	20.1	10.4	<0.02	11.8	27.8
Suspended sediment NIB in 2001 (% error)[6]	6 (13%)	58 (5%)	15 (15%)	0.04 (22%)	16 (1%)	–
Marine section of the delta (surficial sediment)[7]	4.5 ± 1.9	28 ± 9	–	–	–	–
Marine section of the delta (core)[7]	0.38–15	–	–	–	–	–
HR1 core	3–30	50–84	20–38	0.023–0.046	16–40	74–122
CR cores	1–20	48–74	7–50	0.030–0.036	5–29	62–113
CSC core	1–13	28–73	11–25	–	9–23	34–94

Note: CR, Colorado River; TEC, threshold effect concentration; PEC, probable effect concentration; NIB, northern international boundary; HR, Hardy River; CSC, Ciénega de Santa Clara wetlands

[a] References (superscripts 1–7): (1) Presaud et al. (1993); (2) Smith et al. (1996); (3) MacDonald et al. (2000); (4) Hart et al. (2005); (5) Randle et al. (2006); (6) Horowitz et al. (2001); (7) Shumilin et al. (2002)

[b] In two outlayers (not included) in core 298 from Lake Powell, Hg = 0.72 and 1.7 μg g^{-1}. Hg in the CR is from core CR1 ($n = 4$) and for the HR, $n = 8$

[c] One outlayer in core 298 from Lake Powell has Zn concentrations of 989 μg g^{-1}

deposition within the cores. Three factors were determined, which together account for 74% of the total sample variance (Table 2). The first factor (F1) has significant positive loadings for all reported variables except SiO_2, MnO, CaO, NaO, SO_3, Ba, Sr, U, Zr, and LOI. Thus F1, which accounts for 51% of the total variance, groups the Al_2O_3- and Fe_2O_3 -metal-rich clayey sediments. Factor 2 (F2) accounts for 15% of the total variance. Significant positive loadings for LOI, CaO, and Sr suggest that this factor describes the biogenic $CaCO_3$ and organic matter component. It also groups MnO, reinforcing previous suggestions (Daesslé et al. 2002) that Mn is commonly present in this region as part of calcium carbonates and/or as a Mn carbonate. Significantly negative loadings in F2 are seen for SiO_2 and Na, signaling that these elements are probably present as quartz and Na-plagioclase. Finally, consideration is given to a third factor (F3), which groups MnO, Ba, As, Pb, and Cu and accounts for 8% of the total variance. This factor groups elements of environmental interest.

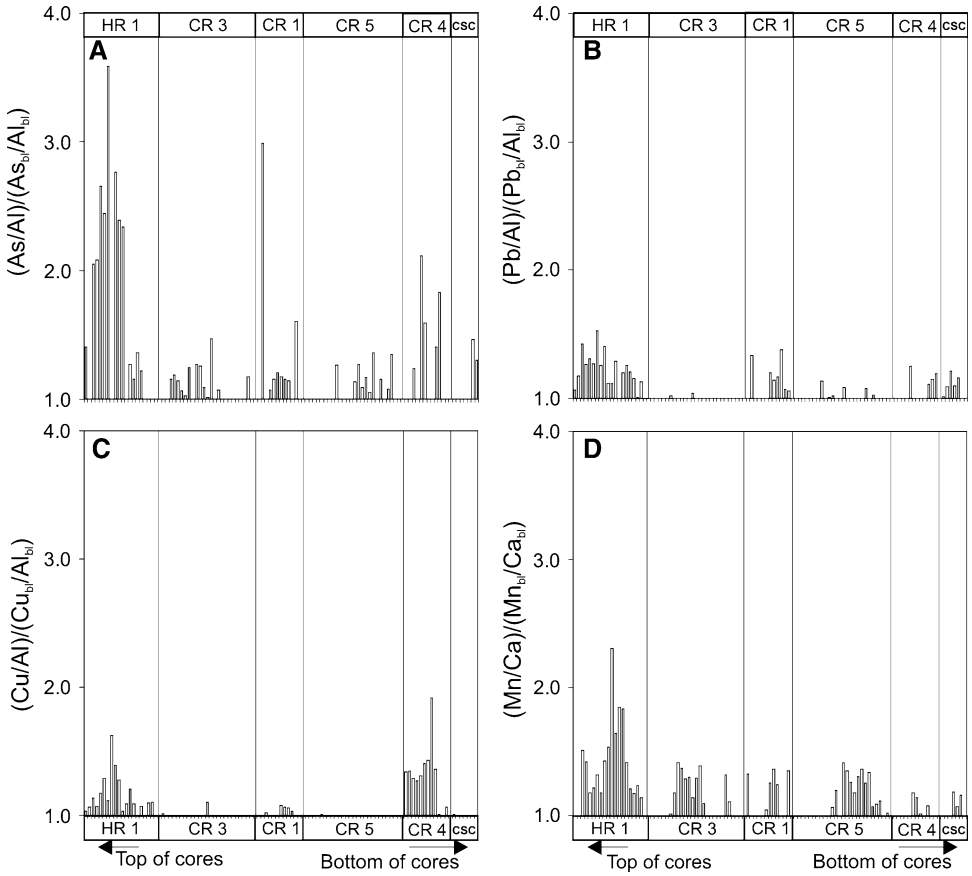

Fig. 6 Enrichment factors (EFs) in sediment cores from the Colorado River delta for (**a**) As, (**b**) Pb, and (**c**) Cu relative to the baseline element$_{bl}$ /$Al_2O_{3\ bl}$ ratios at the bottom of core HR1. The As EF peaks overlap only partially with those for (**d**) MnO, which were calculated relative to CaO

$^{210}Pb_{xs}$ and ^{137}Cs Activities

The activities of $^{210}Pb_{xs}$ were very low in most of the samples, averaging 18 Bq kg^{-1} (Table 3). Core CR1 had much lower $^{210}Pb_{xs}$ activities (0–14 Bq kg^{-1}) than core HR1 (12–66 Bq kg^{-1}). Maximum $^{210}Pb_{xs}$ was found at a 40- to 41-cm depth in both cores, and not at their near-surface as was expected from the exponential isotopic decay pattern of $^{210}Pb_{xs}$ in modern sediments. This indicates that the sedimentation, as well as the atmospheric and fluvial fluxes of $^{210}Pb_{xs}$, was not constant and varied significantly at least in the upper ~50 cm of the cores. The presence of sediments with significantly lower $^{210}Pb_{xs}$ above and below 40–41 cm suggests that the flux of $^{210}Pb_{xs}$ increased significantly during the time of deposition of this layer and/or that the sediments above 41 cm have mixed-age-source heritages from the erosion and redeposition of geologically older sediments at the site of these cores. The 40–41-cm horizon in both cores is located near the limit between facies FF-1 and FF-2. When applying a constant rate of supply (CRS) model for core HR1, a sedimentation rate of 0.9 cm a^{-1} at 28–41 cm is suggested. However, no reliable sedimentation model can be applied for $^{210}Pb_{xs}$ with the available data. The simple presence of $^{210}Pb_{xs}$ down to 51 cm in core HR1 would suggest average sedimentation rates of at least 0.3–0.5 cm a^{-1}, based on a detection limit of five to seven times the half-life (22 years) of ^{210}Pb.

Discussion

Sedimentation Processes in the Colorado Delta

Unlike the main Colorado riverbed, the HR is not connected to any upstream source that may significantly contribute to its sediment load. It does, however, derive its water from the geothermal wells at the nearby Cerro Prieto power station, agricultural return flows, and other effluents. The low water flow in HR would suggest that shifts in sedimentation patterns are driven mainly by hydrodynamic changes induced during flooding periods of the CR main watercourse. Luecke et al. (1999) observed that when flood flows were inexistent, most of the CR south of the northern international boundary (NIB) was a dry ecosystem all the way to the junction with the HR (Fig. 2). Below this junction the channel was perennial due to discharge of agriculture drains and inflow of tidewater. For these reasons, the sediments in HR are expected to contain a more constant and undisturbed sediment record than those in CR.

Table 2 Varimax-rotated factor analysis for 102 samples in cores HR, CR, and CSC from the remnant CR delta

Variable	F-1	F-2	F-3
Clay	0.81	0.08	−0.15
SiO_2	−0.33	−0.92	0.08
TiO_2	0.85	−0.12	−0.02
Al_2O_3	0.96	−0.08	−0.18
Fe_2O_3	0.98	0.06	−0.07
MnO	−0.04	0.52	0.40
MgO	0.83	−0.07	−0.47
CaO	−0.52	0.77	0.16
Na_2O	−0.43	−0.52	−0.01
K_2O	0.89	−0.39	−0.01
P_2O_5	0.79	0.19	0.10
LOI	0.07	0.95	−0.10
As	0.61	0.12	0.50
Ba	−0.12	−0.05	0.93
Ce	0.54	−0.09	0.06
Co	0.78	0.08	0.21
Cr	0.90	−0.18	0.07
Cu	0.51	0.24	0.44
Ga	0.94	0.05	−0.08
La	0.67	0.06	0.17
Nb	0.81	−0.22	0.04
Pb	0.66	−0.12	0.42
Rb	0.89	0.13	0.34
Sr	−0.53	0.79	0.15
V	0.95	0.06	−0.01
Y	0.69	−0.35	−0.23
Zn	0.91	0.16	0.16
Zr	−0.59	−0.32	0.36
% of total variance	51%	15%	8%

Note: HR, Hardy River; CR, Colorado River;

CSC, Ciénega de Santa Clara wetlands; LOI, loss on ignition

Underlined loadings are considered significant and represent the discussed geochemical components

After the Hoover dam was built in 1935 two main flooding periods occurred during the 20th century that must have altered the natural sedimentation regime in the Colorado delta. The first such flood flow (FF-1) lasted ~21 years, from 1939 to 1960. Later, after a period of nonflow between ~1961 and 1978, a second significant flood flow (FF-2) of ~9 years occurred between 1979 and 1988 due to heavy rainfall during El Niño conditions, followed by significant flooding shortly after in 1993. Since then, only minor and short-lived flood flows have occurred. The yearly water volume passing the SIB during 1910–2007 describes the general conditions and abrupt changes in the flooding regime affecting the delta (Fig. 1).

During nonflow periods, an interruption in sediment transport and thus of sedimentation must have occurred. On the contrary, during flood flows the sediment load carried by the river may have settled as it reached the deltaic plain, where the hydrodynamic energy dropped significantly. The beginning of the main flooding events described here was probably accompanied by a water-wave of higher energy which carried suspended sediment and also bed load, including coarser grain size fractions that reached the sand-limited portions of the stream. Sand supply and sand supply limitation prior to and after the construction of Glenn Canyon Dam were described in the Grand Canyon by Topping et al. (2000). Sandy-silt horizons (~20% sand) are present in all the cores studied. In core HR, these are located at depths of 31–34 and 41–54 cm. Other isolated peaks at the core top and bottom are present. A significant enrichment in sand accompanied by a concomitant decrease in clay at 34–31 cm in core HR1 has a geometry strikingly similar to the enrichment in CR1 at 40–48 cm (Fig. 3), and thus they are considered time equivalent. Sand enrichments in the other cores accompanied by clay depletions are also seen. The sediment record of the two main flooding events during the 20th century are interpreted as follows: (a) a sudden drop in clay sedimentation which marks the time at which river flow stopped and sedimentation rates dropped significantly during nonflood periods, (b) the onset of sand and silt deposition signaling the water and sediment wave preceding the inundation of the river reaches during flooding periods, and (c) continuous flow of water and deposition of suspended load, mainly clay-sized material. These sedimentary facies are indicated in Fig. 3.

The presence of $^{210}Pb_{xs}$ in cores CR1 and HR1 (Table 3) suggests sediment ages of the order of tens to hundreds of years. Sediments carried by the CR during flood flows were most likely eroded from the reaches upstream, including release from the dams. Thus, although the sediments in the lower reaches were deposited recently, they must have integrated materials of different isotopic compositions. One possible explanation for the much higher $^{210}Pb_{xs}$ activities at 41 cm in these cores is that during the intense rainfalls caused by El Niño conditions after 1979, local atmospheric $^{210}Pb_{xs}$ fluxes increased significantly. Although no precise sedimentation rates could be estimated from $^{210}Pb_{xs}$ results, the onset of DDT metabolites, 2,4′- and 4,4′-DDE and –DDD, studied by Ramos-Delgado (2008) in core CR1 can be used as an additional time marker. The first and largest peak of 4,4′-DDE + 4,4′-DDD (62 ng g^{-1}) in this core was found at a 49-cm depth, and the second significant peak of 65 ng g^{-1} at a 4-cm depth. Concentrations were 2–30 ng g^{-1} between these two peaks. According to the estimated ages of maximum DDTs in sediments from San Francisco Bay, California (Venkatesan et al. 1999), the

Table 3 Radiometric data for cores CR1 and HR1

Sample	^{210}Pb (Bq kg^{-1})			^{137}Cs (Bq kg^{-1})
	Total	Supported	Unsupported	
CR1				
0–3 cm	28.2 ± 2.0	24.2 ± 1.2	4.0 ± 0.34	n.d.
7–8 cm	37.1 ± 2.2	28.2 ± 1.4	9.0 ± 0.70	n.d.
19–20 cm	24.3 ± 2.7	24.2 ± 1.5	0.1 ± 0.01	n.d.
40–41 cm	53.3 ± 3.2	39.2 ± 1.6	14.0 ± 1.01	n.d.
HR1				
6–7 cm	80.1 ± 5.6	56.5 ± 2.3	23.7 ± 1.91	n.d.
27–28 cm	76.8 ± 5.4	61.1 ± 2.4	15.7 ± 1.26	n.d.
40–41 cm	133.3 ± 8.0	67.6 ± 2.0	65.7 ± 4.40	n.d.
50–51 cm	63.1 ± 3.8	51.3 ± 2.6	11.8 ± 0.92	n.d.

Note: CR, Colorado river; HR, Hardy river

bottom DDT metabolite peak in core CR1 could represent an age of ~1965–1974 or younger, resulting in average sedimentation rates of at least 0.7–1.0 (above 49) cm in this core. The first peak of DDT metabolites in core CR1 lies directly underneath FF-2, which we suggest started during the flood flow that began after 1979. Considering the beginning of FF-2 in cores CR1 and HR1, a similar sedimentation rate (1.3–1.8 cm a^{-1}) can be estimated for the upper section in both cores. The grain size distribution patterns in the bottom of FF-2 layers in cores CR1 and HR1 are strikingly similar and were used as equivalent stratigraphic time markers (Fig. 3). These gross estimates for sedimentation rates are higher than the 0.3–0.5 cm a^{-1} (or more) previously suggested from the detection limit of five to seven times the half-life of ^{210}Pb.

A low atmospheric flux of ^{210}Pb and ^{137}Cs appears to be a regional issue in the southern CR and Gulf of California basins. Rama et al. (1961) measured ^{210}Pb activities in water from the CR and observed a rapid depletion of this isotope from the area of its feed waters (0.245 Bq l^{-1}) to Lake Mead (0.0048 Bq l^{-1}), highlighting the significance of inorganic chemical and biochemical reactions affecting the composition of river waters. Sediments from the Culiacán River estuary (southern Gulf of California) also have conspicuously low ^{210}Pb$_{xs}$ (1.1–28 Bq kg^{-1}) and no detectable ^{137}Cs (Ruiz-Fernández et al. 2007). The higher ^{210}Pb$_{xs}$ in sediments from the central Gulf of California appears to be controlled by scavenging processes related to intense upwelling there. This is suggested by the low concentration of dissolved ^{210}Pb in water adjacent to the Midriff Islands (0.00025–0.006 Bq l^{-1} [Bruland et al. 1974]) and a high concentration of ^{210}Pb$_{xs}$ in sediments there compared to the northern Gulf of California (Daesslé et al. 2004a, b). Adjacent to the Midriff Islands ^{210}Pb$_{xs}$ in sediments is as high as 670 Bq kg^{-1} (Daesslé unpublished data), and in the Guaymas Basin it is ~500 Bq kg^{-1} (Baumgartner et al. 1985).

The depletion and nonconstant fluxes of atmospheric ^{210}Pb$_{xs}$ in the study area may be attributed to one or more of the following factors: (a) depletion of dissolved ^{210}Pb in CR water as it reaches the delta, (b) limited rainfall in this desert area, (c) low atmospheric flux drived from the influence of oceanic air water masses depleted in ^{210}Pb (Preiss et al. 1996), and (d) redeposition and mixing of sediments from geological old strata eroded along the CR basin. Further and more detailed studies regarding age determinations in this region are required.

Sediment Geochemistry

Different results have been published on the sediment geochemistry of the CR and the upper Gulf of California (Table 1). The results for the Colorado delta are compared to different sediments from the lower Colorado basin in the United States, the NIB, and the upper Gulf of California (Horowitz et al. 2001; Shumilin et al. 2002; Hart et al. 2005; Randle et al. 2006). Maximum concentrations of As, Cu, Cr, Pb, and Hg in the cores studied are above those reported for suspended sediments in 2001 at the NIB, especially As in core HR1, which reaches five times the concentration at NIB. It has to be kept in mind, however, that the composition of suspended load at NIB during FF-1 and FF-2 periods is unknown. Unlike Cr, Cu, Pb, and Hg, As in cores HR1 and CR is also significantly enriched compared to its concentrations in sediments from Lake Powell and Navajo Canyon. Concentrations of As in most CR and CSC cores are comparable to those in sediments from the upper Gulf of California. In HR1, however, As concentrations are twice the maximum given for the upper Gulf (Table 2). The EFs calculated for As, Pb, and Cu (Fig. 6) suggest that an additional source for these elements other than the suspended load passing the NIB is likely, especially for As in cores HR1, CR1, and CR4. As, Cu, and Pb are also associated statistically (Table 2). Factor 3

describes an association among As, Cu, and Pb, probably partly sorbed to MnO and/or partly mobilized during oxic diagenesis. Also, an association with Ba is seen. This sediment component is best represented in core HR1, but also at some depths in cores CSC, CR1, and CR4. The downcore distribution of the factor scores for factors 1 and 3 in core HR1 is shown in Figure 7. Positive factor 3 scores are found throughout the core, but increase significantly from 63 cm upward, the same depth at which FF-1 is thought to have started. Later, at ~36 cm, factor scores increase again at the same depth at which FF-2 starts. Thus, the association of As, Pb, Cu, Ba, and probably MnO appears to be related to flooding events. To assess the possibility that sorption onto Mn-oxyhydroxide minerals was responsible for the association of MnO with As, Pb, Cu, and Ba in factor 3, factor analyses were carried out excluding MnO. However, significantly higher loadings for As in factor 3 persisted along with variable (but significant) loadings for Cu, Pb, and Ba. Thus it appears that Mn and Ba are associated with As, Pb, and Cu in factor 3 mainly due to time-similar accumulation rather than significant

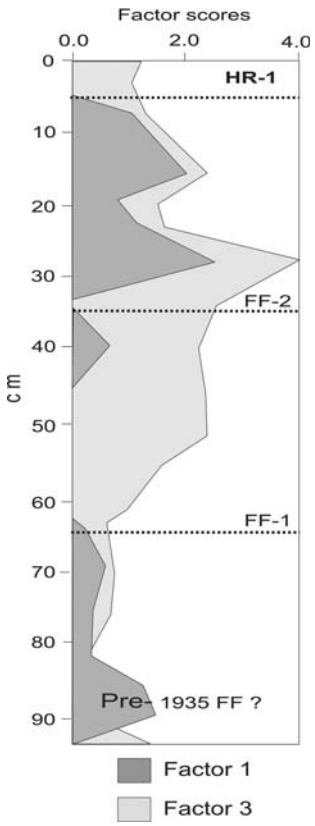

Fig. 7 Downcore distribution of factor scores F1 and F3 in core HR1 as determined from the factor analysis shown in Table 2. Positive scores for F1 represent the clay-metal-rich sediment geochemical component, and scores for F3, the superimposed geochemical signal derived from anthropogenic sources. Suggested flood flows FF-1 and FF-2 are identified

geochemical interactions. This is also suggested from the EF of MnO in Fig. 6d. The EF peaks for MnO (excess MnO relative to CaO) are not located at the same depths as the peaks in As, and overlap only partially in core HR1 Any geochemical association of As, Pb, and Cu with MnO in these sediments appears to be controlled by similar deposition processes, rather than by a dominant geochemical control of MnO, which is predominantly associated with CaO in these sediments.

The most likely sources for As, Pb, and Cu in this agricultural area are arsenate-based pesticides like lead arsenate (LA) [$Pb_5OH(AsO_4)_3$], copper acetoarsenite [$(CH_3COO)_2Cu{\cdot}3Cu(AsO_2)_2$], and calcium arsenate [$Ca_3(AsO_4)_2$]. Lead arsenate was the most extensively used of the arsenical insecticides in the first half of the 20th century, until 1947, when the organic insecticide DDT was introduced (Peryea 1998). It was in the 1920s that the intensification of agriculture and the increase in plagues in the Mexicali Valley began to require pesticides and herbices, mainly Pb, Cu, and Ca arsenates in this region (Moreno-Mena and López-Limón 2005). In New England, for example, the use of LA definitively ceased only until the 1970s (D'Angelo et al. 1996, in Robinson and Ayuso 2004). Arsenates were probably transported from cultivated soils to the CR and HR reaches via the agricultural drains and aerosols. Robinson and Ayuso (2004) studied the pollution of LA in stream sediments in New England. They found an association between Pb and As that could not solely be explained by weathering of the regional rocks (as suggested by the noncorrelation of As and Pb with Fe). According to Ayuso and Foley (2002), As and Pb in soils are retained through sorption on fine-grained Fe and Mn oxyhydroxide minerals and by remineralization as secondary mineral phases with limited solubility. Several studies on LA-polluted soils suggest that As and Pb are usually retained in the upper ~20 cm of the sediment, and that the intrasoil redistribution is limited (Peryea 1998). Furthermore, soil erosion and runoff from land into lakes and streams are important factors in the dispersion of As and Pb within drainage basins (Richardson et al. 1978; Kober et al., 1999). If the onset of As pollution (as reflected by F3 factor scores) occurred during the 1920s, an approximate time correlation with FF-1 shortly afterward can be suggested for core HR1 (Fig. 7).

An alternative source for As, Pb, Cu, and Ba to the sediments studied are the wastewaters and brines from the Cerro Prieto geothermal power station. Recent discussions on pollution issues in the Mexicali area mentioned that the groundwater at Cerro Prieto was being polluted by Ba, As, Li, and Na derived from brines (Imperial Valley/Mexicali Air Quality Taskforce 2007). Barium is used in drilling activities, and could be carried to the river channels via runoff (as was also suggested for LA). Brains and saline

waters at Cerro Prieto have relatively low As and Pb concentrations in solution compared to the nearby Salton Sea power plant (Lippmann et al. 1999). Arsenic, for example, is 0.25 μg g^{-1} in brines from Cerro Prieto but as high as 5 μg g^{-1} in the brines from Salton Sea.

Inorganic Contaminants and Toxicological Considerations

Any potential toxicological effect on benthic organisms that fed on the sediments during the time of enhanced As deposition and/or even presently burrowing organisms need to be assessed. The results are compared with the effect level concentrations for freshwater sediments (Table 1). Arsenic and Cr concentrations are well above the lowest effect level (LEL [Persaud et al. 1993]) and the consensus-based threshold effect concentrations (TECs [MacDonald et al. 2000]). Sediments with concentrations below the LEL and TEC can be considered clean or marginally polluted, as below these concentrations harmful effects on the majority of sediment-dwelling organisms are unlikely to be observed. However, compared with the probable effect level (PEL [Smith et al. 1996]) and the consensus-based probable effect concentration (PEC [MacDonald et al. 2000]), it can be seen that both As (30 μg g^{-1}) and Cr (84 μg g^{-1}) during FF2 in core HR1 are well above the PEL (9.79 and 43.4 μg g^{-1} for As and Cr, respectively) and only slightly below the PEC (33 and 111 μg g^{-1}, respectively). PEL and PEC are concentrations above which adverse effects are expected (or likely) to occur (Smith et al. 1996; MacDonald et al. 2000). Borrowing organisms and/or human disturbance of buried sediments may cause remobilization of these elements. Thus the regional extent of contamination and the potential toxic effects need to be assessed in further detail.

Conclusion

Human manipulation of the CR has caused changes in sedimentation patterns in its estuary. Due to lower sedimentation rates and lower influence by river and/or tidal flooding, the record of these changes was best preserved in sediments from the HR. Clay facies are interpreted as record of flood-flow periods that occurred after dam construction during the 20th century, similar to the massive clay layers prior to river damming. The clayey units have overall higher Fe_2O_3, Al_2O_3, and trace element concentrations, mainly controlled by the association of trace elements with clay minerals and/or their adsorption onto clay surfaces and FeOOH. However, EFs and multivariate statistics indicate that As, and probably also Pb and Cu, may have an anthropogenic source in addition to the baseline sediment geochmistry. Because of their geochemical association, the most likely source for these elements is the drainage of arsenate pesticides used intensively in the region since the 1920s and probably until the 1970s. The increase in As concentrations in one of the cores occurred at the start of flood flow FF-1 in 1939 and was highest shortly after FF-2 period started in 1979. The maximum concentration of As is well above the reported values for river suspended sediments near the NIB or those reported for Lake Powell and Navajo Canyon. Maximum concentrations of As in HR are above the PEL and only slightly below PEC toxicity levels, and Pb, Cu and Cr are partially above TEC consensus levels. The activities of $^{210}Pb_{xs}$ are low overall, probably because of its depletion in the water along the course of the CR, the low atmospheric deposition via rainfall, and/or the redeposition of million-year-old sediments. Gross average sedimentation rates estimated from the onset of DDT metabolites (Ramos-Delgado 2008), the detection limit of five to seven times the half-life of ^{210}Pb, and the proposed stratigraphic interpretation range of $\sim$0.5–2.0 cm a^{-1}.

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