

# Radiocesium in North San Francisco Bay and Baja California Coastal Surface Waters

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## **Abstract**

Radiocesium,  $^{137}\text{Cs}$ , and rare earth elements (REEs) were determined in suspended material and dissolved fractions of waters across the salinity gradient in North San Francisco Bay (estuary). We describe the variation of this conservative isotope tracer with salinity and sediment load. REE data are used to differentiate marine and terrigenous source terrains for suspended material and dissolved fractions. We estimate that about  $1\text{--}4 \times 10^{10}$  Bq of  $^{137}\text{Cs}$  migrates annually on suspended material through the North Bay. In addition,  $^{137}\text{Cs}$  concentrations were measured in surface waters off Baja California. Combined in situ water density ( $\rho_t$ ) and  $^{137}\text{Cs}$  data distinguish between California Current and Gulf of California water, and delineate areas of upwelling, where nutrient-rich, deep Pacific Intermediate water, with little or no  $^{137}\text{Cs}$ , is brought to the surface off promontories along Baja California.

*Keywords:* Radiocesium; Rare earth elements; Isotope tracer; San Francisco Bay; Baja California

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## **1. Introduction**

Man-made radioisotopes of cesium, such as  $^{137}\text{Cs}$ , are important tracers of process and transport in coastal environments for periods of years to decades. Radiocesium

behaves as a conservative element in seawater, similar to other alkali metal ions. It persists in the environment from previous atmospheric nuclear testing, and from nuclear power and processing facility discharge (i.e., Sellafield, UK). Numerous studies describe the oceanic distribution of radiocesium due to atmospheric fallout that provide insight to vertical, as well as lateral, ocean circulation (Bowen, Noshkin, Livingston, & Volchok, 1980; Livingston et al., 1985; Volchok et al., 1971; Whitehead, 1988). Radiocesium inventories and ratios ( $^{137}\text{Cs}/^{90}\text{Sr}$ , and  $^{134}\text{Cs}/^{137}\text{Cs}$ ) of nuclear facility discharge have been used to trace coastal circulation and sediment dispersion (Aarkrog, Dahlggaard, Hallstadius, Hansen, & Holm, 1983; Kupferman, Livingston, & Bowen, 1979; Livingston, 1988; Livingston, Bowen, & Kupferman, 1982; Livingston, Kupferman, Bowen, & Moore, 1984; Mackenzie, Scott, & Williams, 1987). The particle affinity of cesium in freshwater lakes and rivers, compared with its soluble nature in marine waters has been critical for understanding processes in estuaries by which many substances are transferred from land to the oceans (Evans, Alberts, & Clark, 1983; Olsen et al., 1993; Olsen, Simpson, & Trier, 1981).

We describe  $^{137}\text{Cs}$  data for mixed waters in San Francisco Bay, an industrial urbanized estuary, and for surface waters off the Southern California and Baja California coast. It has been three decades since the peak of atmospheric testing and the first investigations of radiocesium in California coastal waters (Bowen et al., 1980; Livingston et al., 1985; Noshkin, Wong, Jokela, Eagle, & Brunk, 1978; Volchok et al., 1971; Whitehead, 1988; Wong, Jokela, Eagle, Brunk, & Noshkin, 1992). One goal of this study was to examine the nature and extent of  $^{137}\text{Cs}$  transfer from the San Francisco Bay estuary offshore into the California Current. We describe  $^{137}\text{Cs}$  in coupled suspended particulate and dissolved fractions across the salinity gradient in North San Francisco Bay to characterize the migration of this isotope through the estuary. In addition, we describe the application of  $^{137}\text{Cs}$  to trace water masses in the southern California Current System, along the eastern boundary of the Pacific Ocean. Radiocesium combined with temperature and salinity data is an effective tracer of water upwelling off the Baja Peninsula. To our knowledge these are the first  $^{137}\text{Cs}$  tracer data for the southern region of the California Current System.

## 2. Field setting

### 2.1. *San Francisco Bay*

The San Francisco Bay is a large urbanized estuarine system that is heavily impacted by human activity (Abu-Saba & Flegal, 1995; Conomos, 1979; Conomos, Smith, & Gartner, 1985; Flegal et al., 1996; Flegal, Smith, Gill, Sañudo-Wilhelmy, & Anderson, 1991; Smith & Flegal, 1993; Spies, Felton, & Rice, 1987). Three subsystems delineate the Bay, and each has distinct hydrography and geography. The North Bay includes the Delta on the east, where the Sacramento and San Joaquin Rivers mix, and Suisun Bay, Carquinez Strait, and San Pablo Bay on the west, where the Napa and Petaluma Rivers enter. The Central Bay connects both the southern and northern sections with the Pacific Ocean through the Golden Gate. South Bay is a lagoon with little freshwater river input.

Salinity in the Bay is tidal driven, though storm runoff, winds, coastal upwelling and advection can dominate for periods during the year, and from year to year (Peterson et al., 1996). The principal salinity gradient in the Bay runs from the Delta area in the Northern Reach to the Central Bay and through Golden Gate to the Pacific Ocean. During low runoff periods saline water intrudes the Delta, and during high runoff periods fresh water flows into Central Bay. The shallow South Bay waters are vertically mixed with relatively uniform salinity. The mean hydraulic residence times in North Bay are 1.2 days and 60 days during high and low runoff; and in South Bay are 120 and 160 days during high and low runoff (Flegal et al., 1991; Smith, 1987).

### 2.2. *Baja California–Gulf of Mexico*

The California Current System extends from Vancouver Island to the tip of Baja California, and is divided into three geographic regions with the Baja Peninsula being the southernmost (Hickey, 1998). The California Current is a major eastern boundary current, which flows equatorward year-round bringing cold Pacific Subarctic water from the north. There have been few current measurements off Baja Peninsula (Fig. 1), so variability and forcing mechanisms are not well understood. However, the large-scale wind stress field channeled by mountains that extend the length of the Baja Peninsula

leads to surface layer offshore transport, which is replaced by upwelling of deeper water (Bakun, 1990; Bakun & Nelson, 1977). Sea surface temperature and ocean color (chlorophyll-a) satellite data provide evidence for pronounced filaments and eddies off coastal promontories, such as Punta Eugenia and Cabo San Lazaro, that are associated with upwelling of colder, less saline Pacific Intermediate water (Bakun, 1990; Bakun & Nelson, 1977; Hickey, 1998).

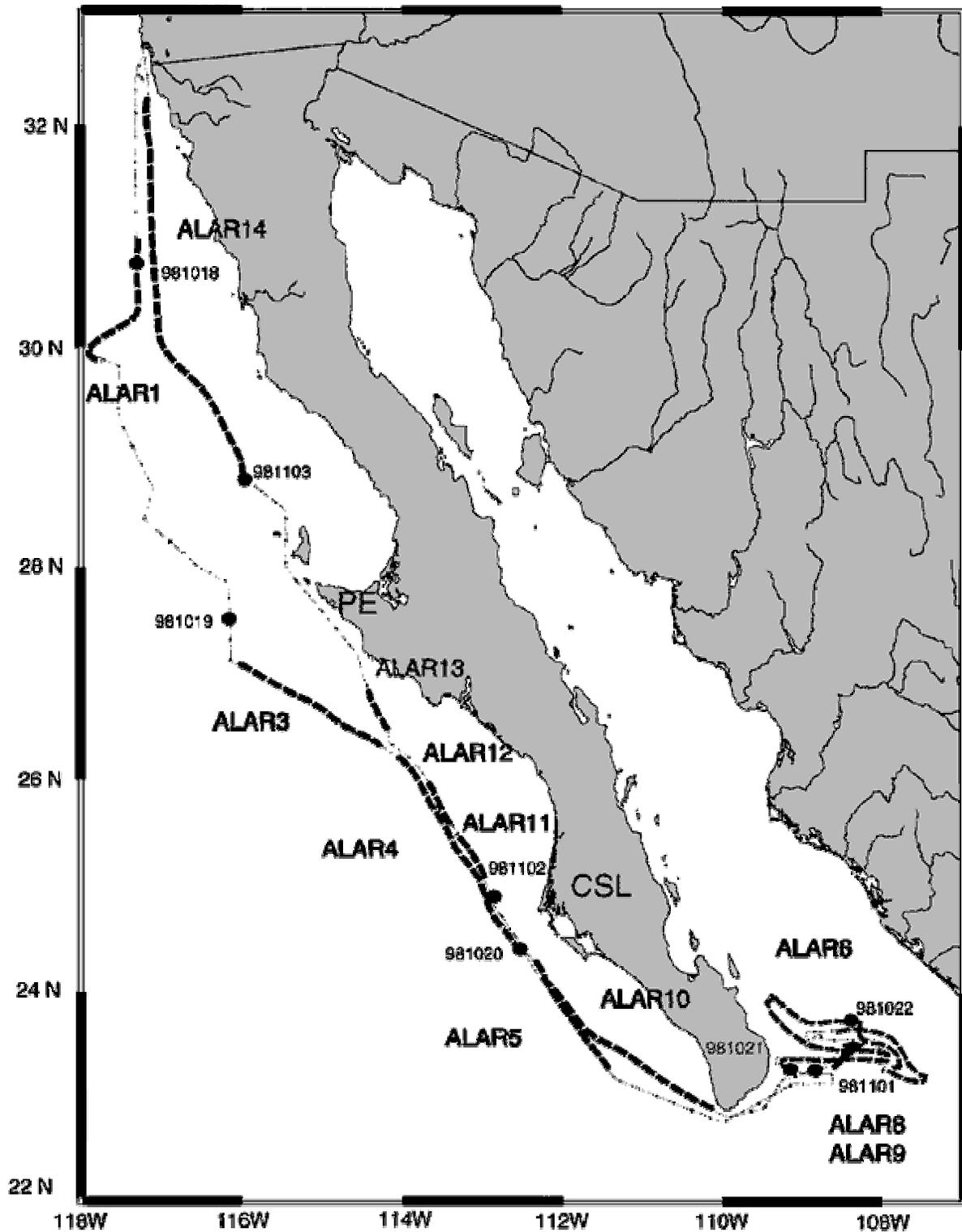
The Gulf of California is an evaporative marginal sea that loses moisture to the atmosphere, so Gulf waters are typically warm and saline. There is a net gain of heat from the atmosphere and circulation is characterized by shallow (50–250 m) outflow of Central Gulf water into the Pacific, and deep (250–500 m) inflow of Pacific Intermediate water (Bray & Robles, 1991). The California Current seldom enters the Gulf of California.

### **3. Experimental methods**

#### *3.1. Sample collection*

San Francisco Bay sample collection was conducted in April 1997 aboard the R/V *David Johnston* (UC Santa Cruz). Surface waters with highly variable salinity and turbidity from three locales in the San Francisco Bay were processed through a suspended material and dissolved ion sampling system. This system isolates coarse suspended particulate material (SPM) greater than 1  $\mu\text{m}$ , fine SPM between 0.1–1.0  $\mu\text{m}$ , and dissolved  $^{137}\text{Cs}$ , which is extracted using solid phase sorbent material, hexacyanoferrate (Bandong, Volpe, Esser, & Bianchini, 2001). All samples were returned to the lab for gamma spectroscopy analysis.

**Fig. 1.** Map showing ship track (fine line) and location of  $^{137}\text{Cs}$  samples (heavy dashed line). Start of calendar day is shown on ship track with filled circle and date (yyymmdd). PE corresponds to Punta Eugenia and CSL is Cabo San Lazaro.



The first station was in the North Bay, or Northern Reach, which extends for nearly 30 nautical miles between the confluence of the Sacramento and San Joaquin Rivers and San Pablo Bay (Table 1). About 2201 of surface water was processed

**Table 1** San Francisco Bay samples (April 1997). Samples were collected across salinity (range) gradients.  $^{137}\text{Cs}$  data are at the 95% confidence level. Fine suspended particulate material, SPM, distribution coefficients,  $K_d$ , and the fraction of  $^{137}\text{Cs}$  associated with the particulate phase,  $F_s$ , as described in text

	Salinity	Volume (l)	Fine SPM (g)	$^{137}\text{Cs}$ mBq/kg (sol) (mBq/l) (liq)	$K_d$ (l/kg)	$F_s$
<i>North Bay</i>	7.1—9.1	220	47.6		18,000	0.80
Fine SPM				4100±400		
Dissolved				0.23±0.07		
<i>San Pablo—Central Bay</i>	11.1—27.5	187	24.9		800	0.10
Fine SPM				1400±300		
Dissolved				1.7±0.2		
<i>Golden Gate</i>	28.4—30.2	396	15.2		<500	<0.02
Fine SPM				<1000		
Dissolved				2.3 ± 0.1		

through the sampler during ebb tide (outgoing) in the Carquinez Strait. The sampler was flushed, and processing continued during slack tide on the transit westward from North Bay through San Pablo and Central Bays. Finally, about 400 l of surface water was processed near the Golden Gate during flood tide (incoming).

In addition, large volumes of seawater were processed through the dissolved ion sorbent extraction system during the Alarcon expedition off Baja California aboard the R/V *Roger Revelle* (UC San Diego), October 17–November 3, 1998. Total suspended material (TSM) was very low (< 1 mg/l). Data collection and analysis focussed only on  $^{137}\text{Cs}$  in the dissolved fraction, since it is a conservative, soluble element in offshore

waters. Samples were processed during transit to and from San Diego and the Gulf of California (Fig. 1).

### *3.2. Total suspended material*

Bottle samples of unfiltered San Francisco Bay water were used to measure total suspended material. Measured volumes of water were filtered through pre-weighed 47 mm diameter, 0.4 µm pore size polycarbonate screen membranes (Poretics PCTE) using a glass filter holder and funnel (Millipore) and a small vacuum pump (Air Cadet). Filters were rinsed with pH 8 water (Millipore-MQ 18.2-WI water + Seastar NH<sub>4</sub>OH), to remove residual salts without leaching or dissolving particles, and then dried overnight at 60°C in a convection oven. Sample filters were then reweighed to determine TSM by difference. To assess membrane weight loss or water retention, blank filters were processed with ultra-pure water, and then dried and reweighed.

### *3.3. Hydrography*

Temperature and salinity data were collected in the field using a SeaBird Electronics' SBE21 CT system (San Francisco Bay), and an underwater SBE 911 +CTD system (Baja California). Salinity values were calibrated with bottle samples collected during the experiments and analyzed at the Oceanographic Data Facility (SIO-UCSD). The ratios of lab to field (salinometer/CTD) salinity measurements have an average value of 1.000 with an uncertainty of 0.1% (RSE). Turbidity was measured using an infrared backscatter Seapoint turbidity meter that was set up on the SeaBird unit. Turbidity measurements show linear correlation with measurement of TSM in bottle samples ( $r = 0.992$ ). TSM values for large volume, processed waters are calculations based on the linear sensor response to suspended material ( $TSM = 0.616 \times \text{sensor value}$ ).

### *3.4. Rare earth elements*

REE concentrations in unfiltered and filtered San Francisco Bay waters were determined by isotope dilution, inductively coupled mass spectrometry using pre-concentration and purification chemistry, and enriched isotopic standards (Esser, Volpe, Kenneally, & Smith, 1994). Unfiltered and filtered (<0.1 µm) samples were acidified (pH = 2) with ultra-pure HNO<sub>3</sub>, several drops of H<sub>2</sub>O<sub>2</sub> were added to oxidize organic material, and then the samples were centrifuged. There was no residue in the filtered

samples, and minor amounts visible in the unfiltered sample dissolved readily when treated with several drops of HF acid.

### *3.5. <sup>137</sup>Cs gamma-ray measurement*

An EG&G Ortec 55% relative efficiency, p-type high-purity germanium (HPGe) coaxial detector was used for the gamma-ray spectrometric analysis of the various fractions of the water samples. The detector has a resolution of 1.95 keV full-width-at-half-maximum at 1.33 MeV (<sup>60</sup>Co gamma peak). To reduce environmental background radiation, a 10 cm thick high-performance, low-background shielding was used to contain the detector and sample during gamma-ray counting. Data acquisition was performed using Ortec's digital gamma-ray spectrometer system DSPEC™, and spectral analysis was done using GammaVision-32 (Ortec) on a Windows® 95 platform. Peak background correction was made during gamma-ray analysis to correct for instrument background, and the background activities inherent in the sorbents and filters.

Detector (counting) efficiency calibration was performed in precisely the same geometrical relationship to the calibrating sources as is used in the measurements on samples of unknown activities. A mixed gamma standard solution (Amersham QCY48) was used to produce a 12-point energy efficiency calibration curve of the detector over the 50–2000 keV region. (Bandong et al., 2001). The mixed gamma standard has <sup>137</sup>Cs as one of the reference radionuclides thus allowing for an accurate quantification of <sup>137</sup>Cs using the 662-keV gamma-ray peak (Bandong et al., 2001).

## **4. Results**

Hydrographic data collected during the April experiment, indicated that North Bay waters at Carquinez Strait had TSM between 180 and 220 mg/l, and salinity between 7.1‰ and 9.1‰. Surface waters sampled during transit across San Pablo Bay and Central Bay had TSM between 12-150 mg/l, and 11-27‰ salinity. Surface water near the Golden Gate had TSM between 4.8 and 52 mg/l, and 28-30‰ salinity, while water from several miles off the coast had TSM between 2-4 mg/l, and 32-33‰ salinity. These data are consistent with previous observations. Conomos and Peterson (1976) have shown that in the North Bay, a broad winter turbidity maximum is centered in the

Carquinez Strait with a typical TSM concentration of 90 mg/l at a salinity of 8‰, while water near the Golden Gate has 10 mg/l TSM at 30‰ salinity.

#### *4.1. <sup>137</sup>Cs in San Francisco Bay samples*

Measured <sup>137</sup>Cs data for the sample fractions are presented in Table I. Fine suspended particulate material (SPM) between 0.1 and 1.0 μm size have <sup>137</sup>Cs activity concentrations that range from 4100 to less than 1000 mBq/kg, and are positively correlated with suspended sediment load, and inversely proportional to salinity. The dissolved fractions (< 0.1 μm) have <sup>137</sup>Cs that range from 0.23 to 2.3 mBq/l, and are positively correlated with increasing salinity. Activities of <sup>137</sup>Cs in these samples are consistent with atmospheric weapons test fallout levels observed previously in coastal California waters (Bowen et al., 1980; Livingston et al., 1985; Noshkin et al., 1978; Wong et al., 1992).

Distribution coefficients ( $K_d = (\text{mBq/kg})/(\text{mBq/l})$ ) range from 18,000 l kg<sup>-1</sup> for the brackish North Bay sample to less than 500 l kg<sup>-1</sup> for the saline Golden Gate sample (Table 1). These values are in agreement with the experimentally determined range of  $K_d$  for Cs of around 10<sup>3</sup> l kg<sup>-1</sup> in seawater and 10<sup>5</sup> l kg<sup>-1</sup> in freshwater (Hamilton-Taylor, Kelly, Kershaw, & Lambert, 1993; Hilton, Nolan, & Jarvis, 1997). The fraction of <sup>137</sup>Cs in the samples associated with the particulate phase ( $F_s$ ) (Wauters & Cremers, 1996), ranges from 0.8 in brackish water to less than 0.02 in saline water (Table 1). <sup>137</sup>Cs concentration in the collected fractions shows particle affinity in freshwater, and conservative soluble behavior in saline water. Data trends suggest that nearly all desorption of <sup>137</sup>Cs from suspended material occurs before mixing in the estuary yields waters that have 10-15‰ salinity.

#### *4.2. REE in San Francisco Bay samples*

REE data for eight of the lanthanides are listed in Table 2. Unfiltered and filtered (less than 0.1 μm) samples were collected at each station while the filtration system was processing large volumes of water. Total suspended material concentrations in unfiltered bottle samples ranged from 5 mg/l in the most saline waters to 173 mg/l (Table 2) in the freshest sampled waters. TSM and REE concentrations are averages of three bottle samples at each station. Analytical uncertainty is better than 1-5%, with high

abundance REE data being more precise. The REE are very particle active, and there is a strong positive correlation ( $r > 0.99$ ) between REE and TSM concentrations across the salinity gradient. REE concentrations in the unfiltered fractions are several orders of magnitude higher than in the dissolved fractions (Table 2). There is a positive correlation between dissolved La and salinity, which suggests that the REE content in the dissolved fraction is controlled by river and seawater mixing.

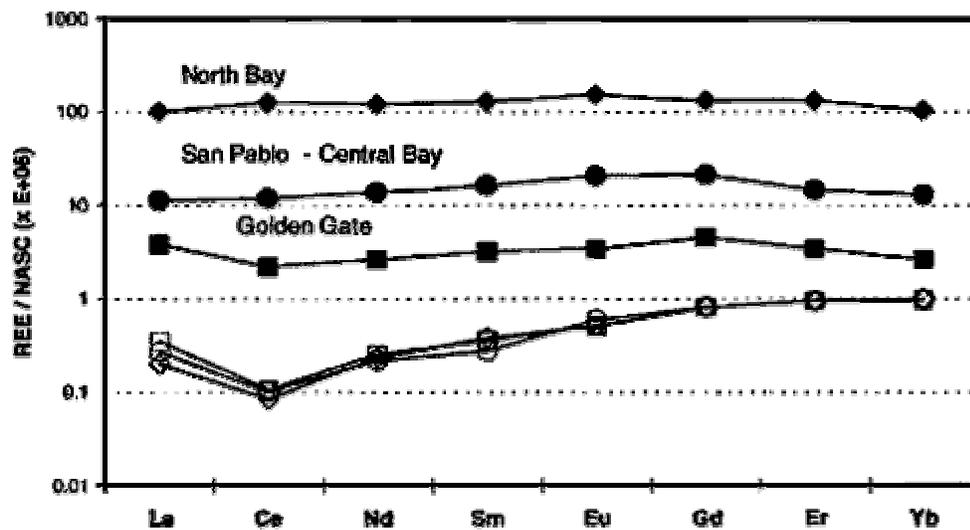
**Table 2** Rare earth elements (mg/kg), salinity and total suspended material (TSM) in San Francisco Bay bottle samples

	Salinity	TSM (mg/kg)	La	Ce	Nd	Sm	Eu	Gd	Er	Yb
<i>North Bay</i>										
Unfiltered	8.27	173	3.07	8.53	3.33	0.73	0.18	0.65	0.43	0.32
Filtered			0.006	0.006	0.006	0.002	0.001	0.004	0.003	0.003
<i>San Pablo—Central Bay</i>										
Unfiltered	20.38	25	0.35	0.80	0.37	0.09	0.02	0.10	0.05	0.04
Filtered			0.009	0.007	0.006	0.002	0.001	0.004	0.003	0.003
<i>Golden Gate</i>										
Unfiltered	29.60	5.0	0.15	0.15	0.07	0.02	0.004	0.02	0.01	0.01
Filtered			0.011	0.007	0.007	0.002	0.001	0.004	0.003	0.003

Normalized REE patterns suggest distinct source composition for the suspended material and the dissolved fraction. Fig. 2 shows REE contents in unfiltered and filtered samples normalized to those in the North America Shale Composite (NASC), which is used as an average composition for eroded silicate rocks (Gromet, Dymek, Haskin, & Korotev, 1984). Values for unfiltered water samples (solid symbols) show decreasing abundance with increasing salinity, but all exhibit a flat normalized REE pattern. These flat normalized REE suggests that the source terrain for the suspended material in North San Francisco Bay waters were eroded sediment carried by the Sacramento and San Joaquin Rivers. The total REE inventory in the suspended material was dominated

by silicate material, though highly variable amounts of suspended biologic material (plankton and algae) were present in the surface waters. In contrast, the normalized REE patterns for all filtered, or dissolved water samples show trends of light REE (La, Ce, Nd) depletion relative to heavy REE (Yb, Er, Gd), and pronounced negative Ce anomalies (oxidation state). These patterns are very similar to those for open ocean seawater (De Baar, Bacon, & Brewer, 1985; Elderfield, 1988; Elderfield, Upstill-Goddard, & Sholkovitz,

**Fig. 2.** Plot of normalized REE data for North Bay (diamond), San Pablo Central Bay (circle) and Golden Gate (square) samples. Unfiltered samples are filled symbols, while dissolved (<0.1 μm) samples are open symbols.



#### 4.3. <sup>137</sup>Cs in Baja California waters

Fig. 1 shows ship transit to and from San Diego and the Gulf of California. The dashed lines correspond to locations and periods during which <sup>137</sup>Cs in surface water was pre-concentrated and isolated on sorbent material. Table 3 shows measured temperature, salinity, calculated density (σ<sub>t</sub>), and <sup>137</sup>Cs for the seawater processed during the expedition. Temperature and salinity data were collected every 1.5 s during the survey, so the values in Table 3 correspond to average compositions (and relative standard deviation) of the water processed while the ship was underway.

California Current water (ALAR1, ALAR14) was typically colder (18.9—19.2°C), less saline (33.5‰) and denser, with <sup>137</sup>Cs values (2.33—2.44mBq/l) higher than other surface waters off Baja (Fig. I). These <sup>137</sup>Cs values were similar to those observed in California Current water off San Francisco Bay. Gulf Surface water (ALAR6, ALAR8, ALAR9) was extremely uniform and showed little, if any, variation in salinity, temperature and TSM during the survey. The uniform <sup>137</sup>Cs concentration, 1.48 ± 0.04 mBq/l, independent of the volume processed (1096, 5037 and 106501) reflects both the homogeneity of surface water in the southern part of the basin, and the reproducibility of the sampler system. Surface water off Baja Peninsula, between Punta Eugenia (PE) and Cabo San Lazaro (CSL) collected on the transit south (19 October: ALAR3, ALAR4) had <sup>137</sup>Cs between 1.55 and 1.59 mBq/l, while water collected on the transit north, (2 November ALAR11, ALAR12, ALAR13) had <sup>137</sup>Cs between 0.85 and 1.07 mBq/l.

**Table 3** Pacific surface water off Baja California. Analytical uncertainty is one standard deviation

	<b>Collection date</b>	<b>Volume (l)</b>	<b>Temp (°C)</b>	<b>Salinity</b>	<b>Density (kg/m<sup>3</sup>)</b>	<b>sigma-t</b>	<b><sup>137</sup>Cs (mBq/l)</b>
ALAR-1	10/18/98	688	19.21	33.50	23.82		2.44
±			0.07	0.02	0.06		0.11
ALAR-3	10/19/98	5074	20.57	33.76	23.67		1.59
±			0.86	0.11	0.32		0.07
ALAR-4	10/19/98	5040	21.49	33.89	23.52		1.55
±			0.22	0.05	0.10		0.07
ALAR-5	10/20/98	685	24.19	34.53	23.23		2.48
±			0.45	0.13	0.23		0.11
ALAR-6	10/21/98	10648	29.44	34.97	21.90		1.44
±			0.19	0.02	0.07		0.07
ALAR-8	10/31/98	5037	29.70	35.03	21.85		1.55
±			0.10	0.02	0.02		0.07

ALAR-9	10/31/98	1062	29.61	35.01	21.87	1.44
±			0.02	0.01	0.02	0.07
ALAR-	11/1/98	4989	25.32	34.72	23.03	1.85
10						
±			0.98	0.09	0.37	0.07
ALAR-	11/2/98	1196	21.77	34.11	23.61	0.85
11						
±			0.66	0.16	0.30	0.04
ALAR-	11/2/98	1080	20.41	33.81	23.75	1.03
12						
±			0.28	0.02	0.09	0.07
ALAR-	11/2/98	1148	20.94	33.88	23.66	1.07
13						
±			0.29	0.02	0.09	0.07
ALAR-	11/3/98	4429	18.89	33.51	23.91	2.33
14						
±			0.34	0.04	0.12	0.15

## 5. Discussion

### 5.1. Estuarine <sup>137</sup>Cs transport

Cesium is fixed in the lattice sites of weathered silicate minerals (micas). Similar to other alkali elements (e.g., Na, K), it is strongly bound and can only be displaced by ions of similar size and charge. As a result, water-mineral exchange in freshwater is limited (Evans et al., 1983; McLean & Summers, 1990). In estuaries, like Northern San Francisco Bay, desorption of <sup>137</sup>Cs from fine suspended particle surfaces occurs during exchange and mixing with saline waters. Changes in the salinity and hydrodynamic conditions of the water cause large changes in the concentrations and properties of particulate and colloidal forms of radionuclides in estuaries due to coagulation, sedimentation and desorption (Benes, Cernik, & Ramos, 1992; Olsen et al., 1993;

Olsen et al., 1981). This is reflected in our data, where there was an 8-fold decrease in concentration of particulate-bound  $^{137}\text{Cs}$  with only a 2-fold decrease in the suspended material load and a 3-fold salinity increase.

Clearly, the transport of  $^{137}\text{Cs}$  depends on the migration of suspended material and bottom sediments carried by rivers into the estuary. Evidence for desorption based on particulate phase activities alone has to take into account the possibility that the observed trends could be produced as a result of mixing of particles from different sources, including marine environments (Beasley & Jennings, 1984; Cook, MacKenzie, McDonald, & Jones, 1997; Harvey, Leonard, & Lovett, 1990). Suspended terrigenous sediment transport in San Francisco Bay is in one direction, from the Delta, across North Bay and into Central Bay, and South Bay to a lesser extent (Krone, 1996; Schemel, Hager, & Childers, 1996). At times, increased wind velocity leads to resuspension of bottom sediments in the shallow North and South Bay. Even though terrigenous suspended sediments persist in surface water at Golden Gate (flat REE patterns), the low suspended material load and the 30–50-fold decrease in REE abundance suggest that fine material settles in deep water basins of North Central Bay. However, it is noteworthy that while fine suspended material of terrigenous origin is present in waters near the entrance to San Francisco Bay, there is little, if any  $^{137}\text{Cs}$  present. This suggests that Cs desorption from terrigenous suspended material in saline water may be rapid (Beasley & Jennings, 1984; Mackenzie et al., 1987; Olsen et al., 1993).

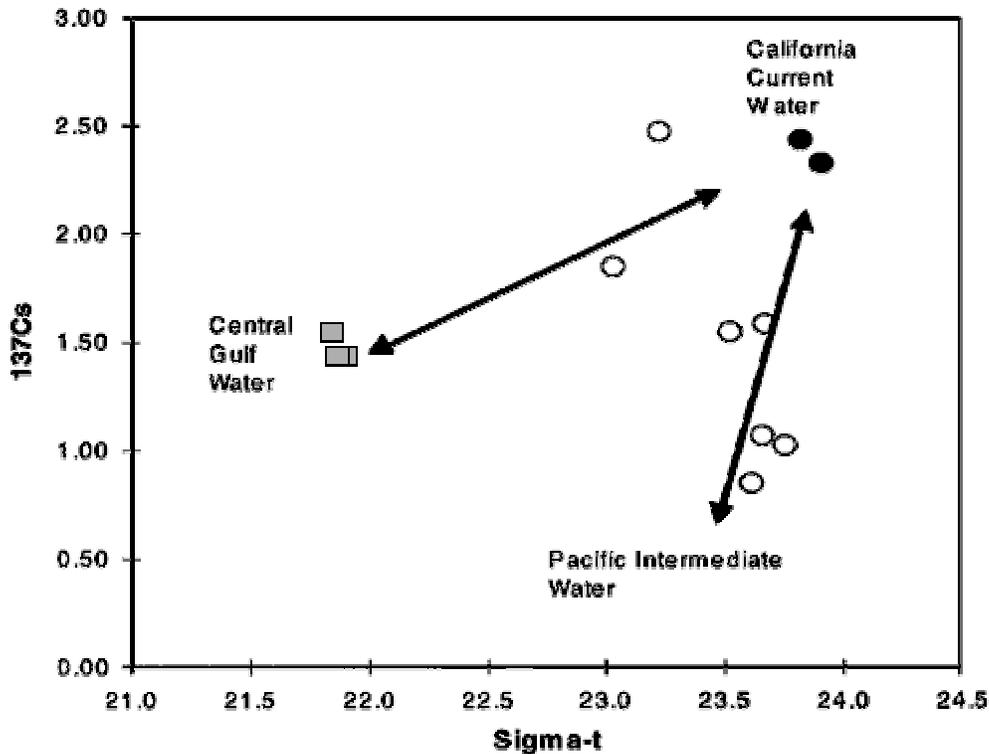
An estimate can be made of the fallout  $^{137}\text{Cs}$  bound on freshwater solids in the Sacramento and San Joaquin drainage areas that migrates into the San Francisco estuary. Flow of these rivers into the estuary is modified by man (water resource management) and nature (storms/droughts), so suspended sediment loads and discharge are highly variable on seasonal to interannual periods. However, average annual estimates of sediment delivered to the Delta and accumulated in North and Central Bay are between  $3\text{--}9 \times 10^9$  kg/yr (Krone, 1996; Schemel et al., 1996). Assuming the North Bay fine SPM concentration (4 Bq/kg) is a lower limit for particle bound  $^{137}\text{Cs}$ , then about  $1\text{--}4 \times 10^{10}$  Bq of  $^{137}\text{Cs}$  migrates into the Bay estuary per year on suspended

material from the Delta. This level is nearly an order of magnitude higher than the  $^{137}\text{Cs}$  present,  $4 \times 10^9$  Bq, in an average tidal prism volume ( $1.6 \times 10^{12}$ ) flushing through the channel at Golden Gate (2.2 mBq/l). In this case, a pronounced spike of dissolved  $^{137}\text{Cs}$  (conservative tracer) may be observable in the California Current offshore of San Francisco Bay during periods of high river runoff.

### *5.2. Tracing offshore circulation*

$^{137}\text{Cs}$  is an effective tracer of water mass circulation, mixing, and upwelling especially in combination with temperature and salinity data. Fig. 3 shows  $^{137}\text{Cs}$  data plotted versus in situ water density, or sigma-t ( $\sigma_t$ ). There is an equation of state for the in situ density of seawater defined by its salinity, temperature and pressure, and expressed as the quantity  $\sigma_{\text{t}}$ , which is equal to the density minus 1000 ( $\text{kg/m}^3$ ). California Current and Central Gulf water had distinct density (23.87 vs. 21.87  $\text{kg/m}^3$ ) and  $^{137}\text{Cs}$  (2.38 vs. 1.48 mBq/L) values. Gulf surface waters had low  $^{137}\text{Cs}$  compared with California Current waters, in spite of high salinity due to evaporation. This is probably due to latitudinal differences in atmospheric fallout, and little river runoff from the surrounding landmass. Surface water processed on the northern transit near the tip of the Baja Peninsula (ALAR10) had intermediate  $^{137}\text{Cs}$  and density values that reflected mixing of California Current and Gulf surface water (Fig. 3).

**Fig. 3.** Plot of  $^{137}\text{Cs}$  (mBq/l) versus sigma-t ( $\text{kg/m}^3$ ), or water density showing mixing between California Current water (solid circles) and Central Gulf surface waters (shaded squares), and trend for upwelling Pacific Intermediate water, with little or no  $^{137}\text{Cs}$  compared to upper surface waters.



Wind-driven upwelling of deep water occurs along the Baja Peninsula that brings cold, saline and nutrient rich water to the surface near the coast. The shelf along Baja is narrow, and upwelling brings Pacific Intermediate water to the surface. There are few hydrographic data (or  $^{137}\text{Cs}$ ) showing vertical distribution in the water column offshore Baja California. Previous profiles of  $^{137}\text{Cs}$  abundance with depth in the Eastern Pacific from GEOSECS (stations 201, 347), the Farallons, and Santa Monica Basin (Bowen et al., 1980; Livingston et al., 1985; Noshkin et al., 1978; Wong et al., 1992) showed marked  $^{137}\text{Cs}$  decreases in the water column at 400–600 m depths. Values approach method detection limits at these water depths, which suggested that  $^{137}\text{Cs}$  was confined to the mixed layer in the upper surface above the thermocline. Hydrographic data and satellite imagery (SeaWiFS) collected during this survey showed significant upwelling of nutrient-rich (increased chlorophyll-a content), cold water along Punta Eugenia and Cabo San Lazaro promontories. The coupled  $^{137}\text{Cs}$  and water density data (Fig. 3) suggest these areas of increased biologic activity reflect mixing of deep Pacific Intermediate water, with little or no  $^{137}\text{Cs}$ , and California Current surface water. There was a 2-week period between the southern (19 October) and northern (2 November)

transects along these promontories. The lower  $^{137}\text{Cs}$  concentrations during the northern transect (ALAR11-13) compared with the southern transect (ALAR3-4) suggest that upwelling was more pronounced during this period. That is, there was increased lateral offshore transport of California Current surface water, which was replaced by deep upwelling Pacific Intermediate water. These data also support physical ocean models and satellite observations, which suggest that upwelling near promontories along the Baja coast is a highly fluctuating process.

## **6. Conclusions**

More than three decades after the peak of atmospheric nuclear weapons testing,  $^{137}\text{Cs}$  is still migrating into the San Francisco Bay estuary and the coastal ocean as freshwater sediments traverse the regional watersheds. We estimate the level of  $^{137}\text{Cs}$  borne on terrigenous suspended material and released upon mixing with saline waters in the San Francisco Bay estuary is at least an order of magnitude more than that entering from the coastal ocean on the incoming tide. The rapid solubility of  $^{137}\text{Cs}$  on suspended material with increasing water salinity suggests that this isotope tracer is useful for understanding the rate of sediment transport and chemical flux at the ocean boundary. The conservative behavior and short-lived activity of  $^{137}\text{Cs}$  in open ocean water makes this isotope a powerful tracer of water mass movement and mixing. Coastal regions exhibiting large-scale vertical upwelling of deeper, nutrient-rich waters (i.e., eastern boundary of the Northern and Southern Pacific Ocean) are critical to biologic productivity. The integration of this tracer with other water properties, like salinity and density, provide insight into the extent of vertical transport and mixing.

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## **References**

Aarkrog, A., Dahlgard, H., Hallstadius, L., Hansen, H., & Holm, E. (1983). Radiocaesium from Sellafield effluents in Greenland waters. *Nature*, *304*(7), 49-51.

Abu-Saba, K., & Flegal, A. R. (1995). Chromium in San Francisco Bay: Superposition of geochemical processes causes complex spatial distributions of redox species. *Marine Chemistry*, *49*, 189-199.

Bakun, A. (1990). Global climate change and intensification of coastal ocean upwelling. *Science*, *247*(4939), 198-201.

Bakun, A., & Nelson, C. S. (1977). *Climatology of upwelling related processes off Baja California*. California Cooperative Oceanic Fisheries Investigations Report, XIX (pp. 107-127).

Bandong, B. B., Volpe, A. M., Esser, B. K., & Bianchini, G. M. (2001). Pre-concentration and measurement of low levels of gamma-ray emitting radioisotopes in coastal waters. *Applied Radiation and Isotopes*, in press.

Beasley, T. M., & Jennings, C. D. (1984). Inventories of  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  in Columbia River sediments from Hanford to the Columbia River Estuary. *Environmental Science and Technology*, *18*, 207-212.

Benes, P., Cernik, M., & Ramos, P. L. (1992). Factors affecting interaction of radiocesium with freshwater solids; 2. contact time, concentration of the solid and temperature. *Journal of Radioanalytical and Nuclear Chemistry*, *159*, 201-218.

Bowen, V. T., Noshkin, V. E., Livingston, H. D., & Volchok, H. L. (1980). Fallout radionuclides in the Pacific Ocean: vertical and horizontal distributions largely from GEOSECS stations. *Earth and Planetary Science Letters*, 49, 41-434.

Bray, N. A., & Robles, J. M. (1991). Physical oceanography of the Gulf of California. In J. P. Dauphin, & B. R. T. Simoneit (Eds.), *The Gulf and Peninsular Province of the Californias* (pp. 511-553). Tulsa, Okla., USA: American Association of Petroleum Geologists.

Conomos, T. J. (1979). Properties and circulation of San Francisco Bay waters. In T. J. Conomos (Ed.), *San Francisco Bay: The urbanized estuary* (pp. 47-84). San Francisco: American Association for the Advancement of Science.

Conomos, T. J., & Peterson, D. H. (1976). Suspended-particle transport and circulation in San Francisco Bay: an overview. In M. Wiley (Ed.), *Estuarine processes: Circulation, Sediments, and transfer of material in the estuary*, Vol. II (pp. 82-97). New York: Academic Press.

Conomos, T. J., Smith, R. E., & Gartner, J. W. (1985). Environmental setting of San Francisco Bay. *Hydrobiologia*, 129, 1-12.

Cook, G. T., MacKenzie, A. B., McDonald, P., & Jones, S. R. (1997). Remobilization of Sellafield-derived radionuclides and transport from the North-East Irish Sea. *Journal of Environmental Radioactivity*, 35(3), 227-241.

De Baar, H. J. W., Bacon, M. P., & Brewer, P. G. (1985). Rare earth elements in the Pacific and Atlantic Oceans. *Geochimica et Cosmochimica Acta*, *49*(9), 1943-1959.

Elderfield, H. (1988). The oceanic chemistry of the rare-earth elements. *Philosophical Transactions of the Royal Society of London A*, *325*(1583), 105-126.

Elderfield, H., Upstill-Goddard, R., & Sholkovitz, E. R. (1990). The rare earth elements in rivers, estuaries, and coastal seas and their significance to the composition of ocean waters. *Geochimica et Cosmochimica Acta*, *54*, 971-991.

Esser, B. K., Volpe, A., Kenneally, J. M., & Smith, D. K. (1994). Preconcentration and purification of rare earth elements in natural waters using silica-immobilized 8-hydroxyquinoline and a supported organophosphorus extractant. *Analytical Chemistry*, *66*(10), 1736-1742.

Evans, D. W., Alberts, J. J., & Clark, R. A. (1983). Reversible ion-exchange fixation of cesium-137 leading to mobilization from reservoir sediments. *Geochimica et Cosmochimica Acta*, *47*, 1041-1049.

Flegal, A. R., Rivera-Duarte, I., Ritson, P. I., Scelfo, G. M., Smith, G. J., Gordon, M. R., & Sanudo-Wilhemly, S. A. (1996). Metal contamination in San Francisco Bay waters: historic perturbations, contemporary concentrations, and future considerations. In J. T. Hollibaugh (Ed.), *San Francisco Bay: The ecosystem* (pp. 173-188). San Francisco, CA: AAAS.

Flegal, A. R., Smith, G. J., Gill, G. A., Sañudo-Wilhelmy, S., & Anderson, L. C. D. (1991). Dissolved trace element cycles in the San Francisco Bay estuary. *Marine Chemistry*, *36*, 329-363.

Gromet, L. P., Dymek, R. F., Haskin, L. A., & Korotev, R. L. (1984). The North American Shale Composite: its compilation, major and trace element characteristics. *Geochimica et Cosmochimica Acta*, *48*, 2469-2482.

Hamilton-Taylor, J., Kelly, M., Kershaw, P., & Lambert, C. E. (1993). Radionuclide aquatic pathways. In F. Warner, & R. M. Harrison (Eds.), *Radioecology after Chernobyl: Biogeochemical pathways of artificial radionuclides* (pp. 222-233). New York: Wiley.

Harvey, B. R., Leonard, K. S., & Lovett, M. B. (1990). Some experiences with chemical and physical techniques for the separation of different species of man-made radionuclides in environmental waters. In M. Garcia-Leon, & G. Madurga (Eds.), *Low-level measurements of man-made radionuclides in the environment* (pp. 263-292). New York: World Scientific.

Hickey, B. M. (1998). Coastal oceanography of Western North America from the tip of Baja California to Vancouver Island. In A. R. Robinson, & K. H. Brink (Eds.), *The sea* (pp. 345-393). New York: Wiley.

Hilton, J., Nolan, L., & Jarvis, K. E. (1997). Concentrations of stable isotopes of cesium and strontium in freshwaters in northern England and their effect on estimates of sorption

coefficients (Kd). *Geochimica et Cosmochimica Acta*, 61(6), 1115-1124.

Krone, R. B. (1996). Recent sedimentation in the San Francisco Bay system. In J. T. Hollibaugh (Ed.), *San Francisco Bay: The ecosystem*. San Francisco: Pacific Division AAAS.

Kupferman, S. L., Livingston, H. D., & Bowen, V. T. (1979). A mass balance for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in the North Atlantic Ocean. *Journal of Marine Research*, 37, 157-199.

Livingston, H. D. (1988). The use of Cs and Sr isotopes as tracers in the Arctic Mediterranean Seas. *Philosophical Transactions of the Royal Society of London A*, 325, 161-176.

Livingston, H. D., Bowen, V. T., Casso, S. A., Volchok, H. L., Noshkin, V. E., Wong, K. M., & Beasley, T. M. (1985). *Fallout nuclides in Atlantic and Pacific water columns: GEOSECS data*. Woods Hole Oceanographic Institution Technical Report, WHOI-85-19.

Livingston, H. D., Bowen, V. T., & Kupferman, S. L. (1982). Radionuclides from Windscale discharges I: their dispersion in Scottish and Norwegian coastal circulation. *Journal of Marine Research*, 40(4), 1227-1258.

Livingston, H. D., Kupferman, S. L., Bowen, V. T., & Moore, R. M. (1984). Vertical profile of artificial radionuclide concentration in the central Arctic Ocean. *Geochimica et Cosmochimica Acta*, 48(11), 2195-2203.

Mackenzie, A. B., Scott, R. D., & Williams, T. M. (1987). Mechanisms for northwards dispersal of Sellafield waste. *Nature*, *329*, 42-45.

McLean, R. I., & Summers, J. K. (1990). Evaluation of transport and storage of Co-60, Cs-134, Cs-137 and Zn-65 by river sediments in the lower Susquehanna River. *Environmental Pollution*, *63(N2)*, 137-153.

Noshkin, V. E., Wong, K. M., Jokela, T. A., Eagle, R. J., & Brunk, J. L. (1978). *Radionuclides in the marine environment near the Farallon Islands*. Lawrence Livermore National Laboratory, UCRL-52381.

Olsen, C. R., Larsen, I. L., Mulholland, P. J., Damm, K. L. V., Grebmeier, J. M., Schaffner, L. C., Diaz, R. J., & Nichols, M. M. (1993). The concept of an equilibrium surface applied to particle sources and contaminant distributions in estuarine sediments. *Estuaries*, *16*, 683-696.

Olsen, C. R., Simpson, H. J., & Trier, R. M. (1981). Plutonium, radiocesium and radiocobalt in sediments of the Hudson River estuary. *Earth and Planetary Science Letters*, *55*, 292-377.

Peterson, D. H., Cayan, D. R., Dettinger, M. D., Noble, M. A., Riddle, L. G., Scheml, L. E., Smith, R. E., Uncles, R. J., & Walters, R. A. (1996). San Francisco Bay salinity: observations, numerical simulation and statistical models. In J. T. Hollibaugh (Ed.), *San Francisco Bay: The ecosystem* (pp. 9-34). San Francisco, CA: AAAS.

Schemel, L. E., Hager, S. W., & Childers, D. (1996). The supply and carbon content of suspended sediment from the Sacramento River to San Francisco Bay. In J. T. Hollibaugh (Ed.), *San Francisco Bay: The ecosystem* (pp. 237-260). San Francisco: Pacific Division AAAS.

Smith, G. J., & Flegal, A. R. (1993). Silver in San Francisco Bay estuarine waters. *Estuaries*, *16*, 547-558.

Smith, L. H. (1987). *A review of circulation and mixing studies of San Francisco Bay, California*. U.S. Geological Survey, Circ. 1015.

Spies, R. B., Felton, J. S., & Rice, D. W. (1987). Benthioles in estuarine sediments as indicators of street runoff. *Nature*, *327*, 697-699.

Volchok, H. L., Bowen, V. T., Folsom, T. R., Broecker, W. S., Schuert, E. A., & Bien, G. S. (1971). Oceanic distributions of radionuclides from nuclear explosions. In *Radioactivity in the marine environment* (pp. 42-89). Washington, D.C: National Academy Science.

Wauters, J., & Cremers, A. (1996). Effect of particle concentration and fixation on radiocesium sorption. *Environmental Science and Technology*, *30*, 2892-2898.

Whitehead, N. E. (1988) *Inventory of <sup>137</sup>Cs and <sup>90</sup>Sr in the world oceans*. International Lab Marine Radioactivity, IAEA-TECDOC-481.

Wong, K. M., Jokela, T. A., Eagle, R. J., Brunk, J. L., & Noshkin, V. E. (1992). Radionuclide concentrations, fluxes,

and residence times at Santa Monica and San Pedro Basins.  
*Progress in Oceanography, 20, 353-391.*