**C-051**

# MARINE ORGANISMS IN THE SOUTHERN CALIFORNIA BIGHT AS INDICATORS OF POLLUTION

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## ABSTRACT

The Southern California Bight is a rather extensive ecosystem into which large quantities of urban and industrial waste are deposited. This paper presents data for two studies using the California mussel, *Mytilus californianus*, and the bay mussel, *Mytilus edulis*, to demonstrate the magnitude and extent of trace element pollution from the urban areas into the Bight. Data associating trace element levels within the tissues of the Dover Sole, *Microstomus pacificus*, as associated with the incidence of fin erosion are also presented. The highly efficient analytical system used to carry out the multielement analysis of the many biological tissues required for this type of study is described in moderate detail.

## RÉSUMÉ

La baie de la Californie du Sud est un écosystème plutôt étendu dans lequel sont déversées de grandes quantités de déchets municipaux et industriels. La présente communication comprend des données de deux études en utilisant le moule de Californie, *Mytilus californianus*, et le moule de baie, *Mytilus edulis*, pour démontrer l'importance et l'étendue de la pollution du golfe par les éléments-traces provenant des municipalités de cette région. On y présente aussi des données qui établissent une relation entre le taux des éléments-traces dans les tissus de la sole *Microstomus pacificus* et la régression des nageoires. Le système analytique très efficace essentiel pour ce type d'étude et utilisé pour le dosage simultané de plusieurs éléments dans les nombreux tissus biologiques est décrit de façon quelque peu détaillée.

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## INTRODUCTION

The Southern California Bight, extending from Point Conception to Cabo Colnett in Mexico and westward to the California Current, provides an area from which many forms of marine organisms can be selected to serve as indicators and integrators of urban and industrial pollution. Approximately eleven million people live and work in the basins adjoining the Bight. At present about 1,000,000,000 gallons of treated municipal wastewater are discharged daily through marine outfalls into the Southern California Bight. About 84% of the discharge receives only primary treatment. This discharge is more than twice the long term average for surface runoff into the Bight. An added feature of the area is the relatively large number of offshore islands which serve as natural biological sampling sites along the suspected gradients formed by the pollutants in the wastewater discharge.

The Laboratory of Nuclear Medicine, formerly the Atomic Energy Project, has had a continuing interest in the fate of certain trace elements in the environment. In past years this interest has centered on such elements as Sr, Cs, Zn, Cr, and Mn, all of which have radioisotopes resulting from the atomic energy program. Presently these interests are being extended to practically all elements as we study the many aspects of potential pollution associated with the expanding energy development program.

An important implement in these studies has been an optical emission spectrometer system (Fig. 1), which was developed to determine the levels of stable elements present in dry biological tissue. The heart of the system is an optical emission spectrometer covering the wavelength range of 210 nm to 670 nm and having a reciprocal linear dispersion of 0.68 nm/mm. Forty-four detector units (Fig. 2), are arranged on the focal circle of this instrument. Each detector is comprised of a secondary slit, a mirror to focus this slit image on the cathode of a photomultiplier tube and an interference filter to eliminate unwanted scattered light originating from the grating of the spectrometer.

In order to carry out the analysis, the sample must be volatilized, the resulting molecules disassociated into atoms, and these atoms excited by means of an electrical discharge to give off the characteristic optical spectra for the elements present within the tissue. To accomplish this, we use the special electrode configuration shown in Fig. 3. For animal tissues, a crater electrode with a 1.9 mm (.075 in.) D. stem is used. Five to fifteen milligrams of sample are weighed into the crater. No additional materials are added to the sample. A 12.5 ampere D.C. arc is drawn between the electrodes to volatilize the sample and excite the elements. During this process, each
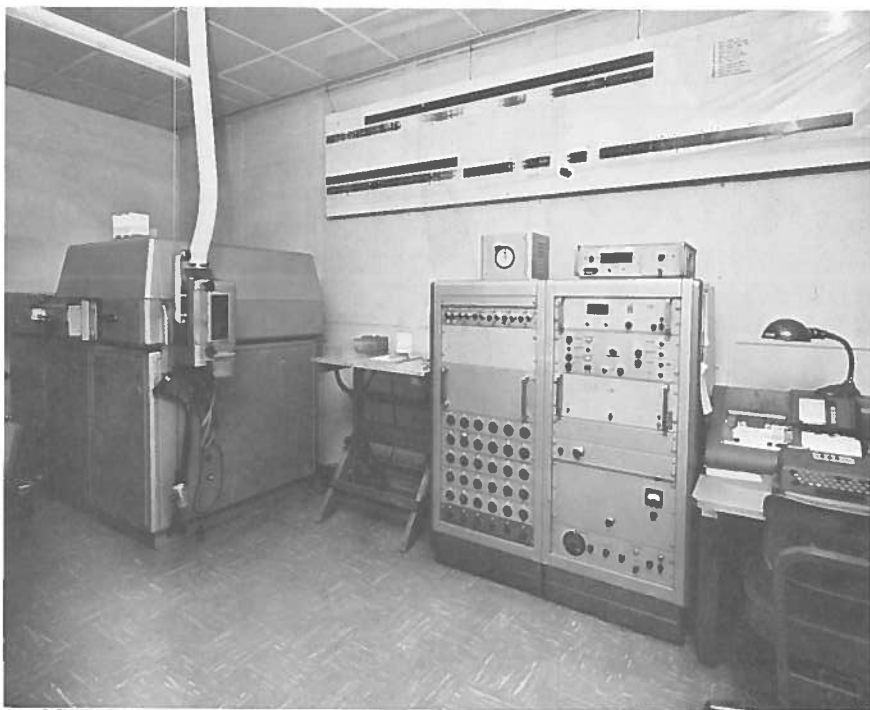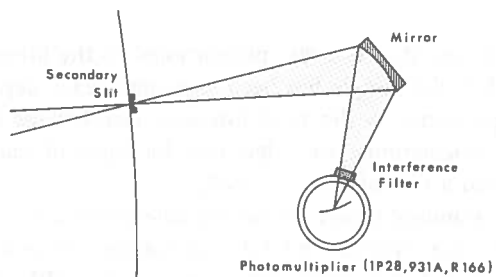
Fig. 1.  Optical Emission Spectrometer System.



Fig. 2.  Schematic Diagram of Detector Components.
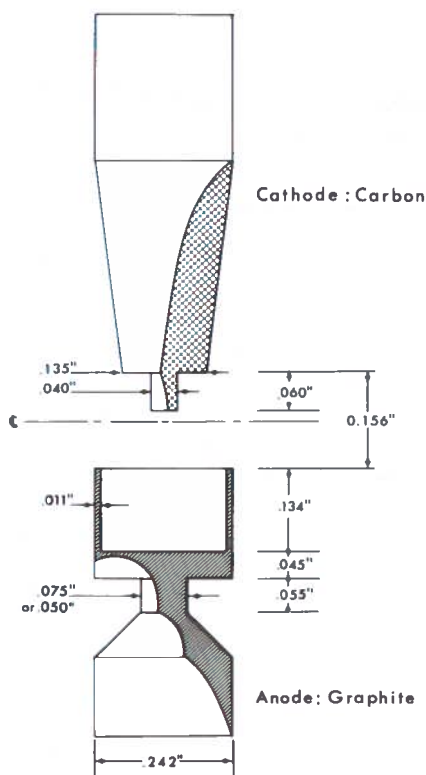
Cathode : Carbon

Anode: Graphite

Fig. 3. Electrode Configuration.

detector charges a capacitor at a rate proportional to the intensity of the light received. Thus, when the sample has been consumed, each capacitor is charged to a voltage proportional to the total intensity received by the detector. By relatively simple calculations, the electrode background can be subtracted from each signal and a net intensity derived.

By suitable standardization, these net intensities can be converted into concentrations. In this system, the total intensities are automatically transferred to IBM cards and then processed by means of an IBM 360-91 computer to concentrations. The spectral lines which are used for the analysis are shown in Fig. 4. The relative standard deviation of analysis is generally between 3% and 15% for finely ground material. The accuracy of analysis is largely dependent upon the adequacy of the calibration standards, but as judged by

comparison with the NBS Standard Reference Materials (Fig. 5), is quite good at levels above 1 ppm.

The important features of this method, beyond its simultaneous multi-element capability, are that each analysis is performed at a materials cost of less than 50¢ and, on the average, no more than five minutes of technician time, including sample preparation and analysis, is required.

This capability for determining trace elements in biological tissues and a very real interest in the element concentrating ability of certain marine tissues have led to a rather natural cooperation between the Laboratory of Nuclear Medicine and the Southern California Coastal Water Research | Project. This project, which involves a group of 30 scientists, was founded in 1969 when five local government agencies entered into a joint power agreement to sponsor environmental studies. In practice, these studies have been primarily associated with the understanding of the environmental impact of wastewater emission into the Southern California Bight.

In 1971 we initiated a study to determine whether or not marine tissues commonly available in the intertidal zone of our coastline could be used to reflect the trace element pollution resulting from this large wastewater input. In the hope of establishing a significant gradient between open ocean and the more urban related shoreline, we collected samples from coastal sites from San Diego to Gaviota and from island sites out to San Miguel and San Nicolas,

### ANALYTICAL LINES

| | | | | | |
|---|---|---|---|---|---|
| P | 2535.6 | Mn | 2576.1 | Ba | 4934.1 |
| Na | 3302.3 | Mn | 2933.1 | Li | 6103.6 |
| Na | 6154.2 | B | 2497.7 | Ag | 3280.7 |
| K | 4044.1 | Al | 3082.2 | Sn | 2840.0 |
| Ca | 3158.9 | Si | 2881.6 | Pb | 2833.1 |
| Ca | 4454.5 | Ti | 3372.8 | Be | 2348.6 |
| Mg | 5183.6 | V | 4379.2 | Be | 3131.1 |
| Zn | 2138.6 | Ni | 3414.8 | Cd | 2288.0 |
| Zn | 3282.3 | Mo | 3132.6 | As | 2780.2 |
| Cu | 3247.5 | Cr | 4254.4 | Hg | 2536.5 |
| Fe | 2488.2 | Sr | 4077.7 | Sb | 2598.1 |

### BACKGROUND POSITIONS

| | | | | | |
|---|---|---|---|---|---|
| CN | 3883.4 | Bkgd 2 | 5219.6 | SiO | 2413.8 |
| TL | 4001.0 | Bkgd 3 | 2204.0 | CaO | 6193.5 |
| $C_2$ | 4714.8 | Bkgd 4 | 2104.0 | PO | 2462.7 |

Fig. 4. Analytical Line Array—1975.

Analysis of SRM # 1577

| Element | NBS | NMRB |
|---|---|---|
| K | 0.97 ± 0.06% | 0.98 ± 0.17% |
| Na | 0.243 ± 0.013% | 0.252 ± 0.096% |
| Fe | 270 ± 20 ppm | 276 ± 88 ppm |
| Cu | 193 ± 10 | 190 ± 59 |
| Zn | 130 ± 10 | 127 ± 27 |
| Mn | 10.3 ± 1.0 | 10.7 ± 2.5 |
| Pb | 0.34 ± 0.08 | 0.18 ± 0.34 |
| Cd | 0.27 ± 0.04 | 1.8 ± 1.2 |
| | 250mg Aliquotes | 10mg Aliquots |

Analysis of SRM # 1571

| Element | NBS | NMRB |
|---|---|---|
| Ca | 2.09 ± 0.03% | 2.31 ± 0.27% |
| K | 1.47 ± 0.03% | 1.36 ± 0.03% |
| Mg | 0.62 ± 0.02% | 0.64 ± 0.06% |
| P | 0.21 ± 0.01% | 0.21 ± 0.04% |
| Fe | 300 ± 20 | 281 ± 67 |
| Mn | 91 ± 4 | 90 ± 11 |
| Na | 82 ± 6 | 80 ± 20 |
| Pb | 45 ± 3 | 43 ± 9 |
| B | 33 ± 3 | 33 ± 5 |
| Zn | 25 ± 3 | 25 ± 8 |
| Cu | 12 ± 1 | 11 ± 2 |
| Ni | 1.3 ± 0.2 | 1.0 ± 0.3 |
| Cd | 0.11 ± 0.02 | 0.7 ± 0.8 |
| | 250mg Aliquotes | 10mg Aliquotes |

Fig. 5. Comparison with NBS Standard Reference Materials.

which are near the western edge of the Southern California Bight. The animals collected were starfish, abalone, goose barnacle, limpets, mussels, etc. Most of these animals were dissected into several components and then analyzed for approximately twenty-five elements. As might be expected this generated a very large amount of data. As a single tissue, the digestive gland of the intertidal mussel, *Mytilus californianus,* showed the largest number of elements within the detection range of our system and therefore shows the greatest promise in establishing suspected gradients.

Figures 6 through 9 show the concentration of Pb, Cr, Cu, and Ag found in the mussel digestive gland during this survey. Lead concentrations are generally higher near the shore than at the outer islands. In fact, a gradient of about an order of magnitude seems to exist parallel to shore with no "hot spots" in the areas of major wastewater input such as at Santa Monica Bay, Royal Palms, and Point Loma. In 1971, approximately 100 m tons of lead were introduced to the Bight by surface runoff and 200 m tons by wastewater (Young *et al.* 1973). A recent estimate of aerial transport into the Bight indicates an input of 100 m tons (Huntzicker *et al.* 1975). The lead picture, as seen by the mussel digestive gland, indicates that the major input is aerial and surface runoff.

It has been estimated (Young *et al.* 1974) that almost 200 m tons of copper are contained in the antifouling paints applied to recreational, commer-
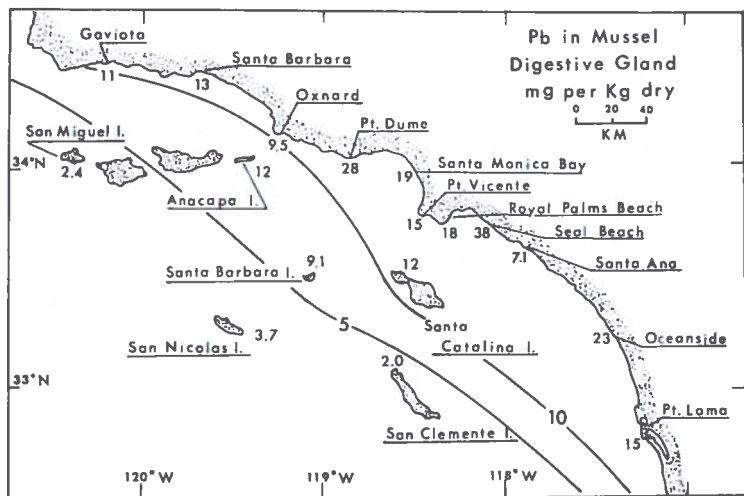


Fig. 6. Lead in Mussel Digestive Gland—1971.

cial, and naval vessels in Southern California anchorages, with about 95% being used off of the major urban centers. It is reasonable to assume that a significant portion of this copper will be released into the Bight during each year. The wastewater input of copper in 1971 was estimated at 600 m tons with an additional input of 100 m tons via rainwater. The data shown in Fig. 7 indicate major inputs of copper near Royal Palms and Point Loma, which are located adjacent to major wastewater inputs for Los Angeles and San Diego, and adjacent to the major commercial harbors of these two cities.

Due to past emission of large quantities of DDT through the Whites Point outfall near Royal Palms Beach, this wastewater plume was heavily "tagged" with DDT and related coupounds. Patterns of DDT contamination in intertidal mussels collected from islands throughout the Bight indicate that the offshore transport of contaminants from this outfall extends to Santa Barbara Island and probably beyond (SCCWRP 1973). This fact suggests that the high copper value observed at Santa Barbara Island may also result primarily from the Whites Point wastewater outfall. In contrast to lead, the copper gradient from our outermost islands to the shore is 2.5 times or less. This may be a consequence of the relatively high copper level in 'pure' sea water and the relatively large advective input of copper from outside of the Bight.

Silver (Fig. 8), exhibited a similar pattern to copper, in that the highest concentrations seem to be associated with our major wastewater outfalls. In
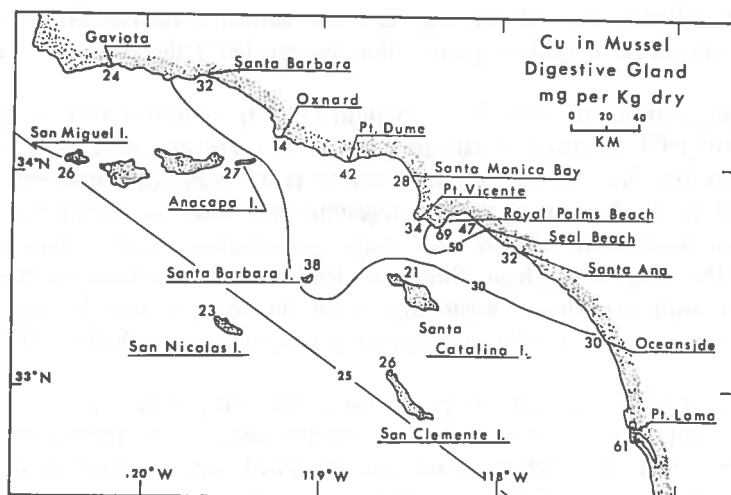


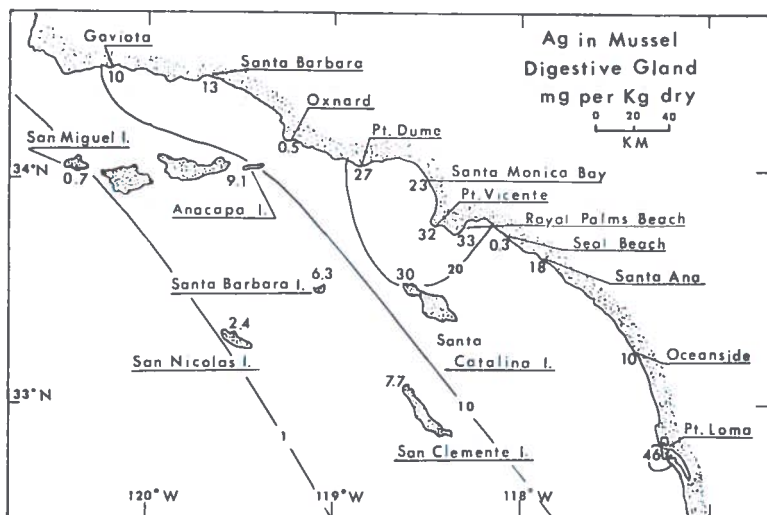Fig. 7. Copper in Mussel Digestive Gland—1971.

Fig. 8. Silver in Mussel Digestive Gland—1971.

contrast to copper, the silver values are not elevated when compared to Anacapa and San Clemente Islands, which suggests that the major source of this element was not the Whites Point outfall but rather the Hyperion outfall in Santa Monica Bay. Monitoring data for annual mass emission rates of metals via these outfalls supports this assumption (Alexander and Young 1976).

The pattern observed for chromium (Fig. 9), indicates that during the spring of 1971 relatively small amounts of this element were entering the Bight via the Point Loma outfall, whereas rather significant gradients were observed in the Los Angeles area, suggesting that both the Whites Point and Hyperion wastewater outfalls were major contributors of chromium to the Bight. The surprisingly high chromium level observed at Gaviota does not correlate with any known wastewater input; however, it may be associated with a minor input in that area resulting from oil tanker ballast (SCCWRP 1973).

No clear-cut regional differences were observed for zinc and nickel. The range of values was but twofold for zinc (46 ppm to 110 ppm) with little suggestion that the higher values are associated with wastewater outfalls. Nickel exhibited a sixfold range (3.3 ppm to 20 ppm) with no significant difference being noted between the islands and the coastal stations. In the
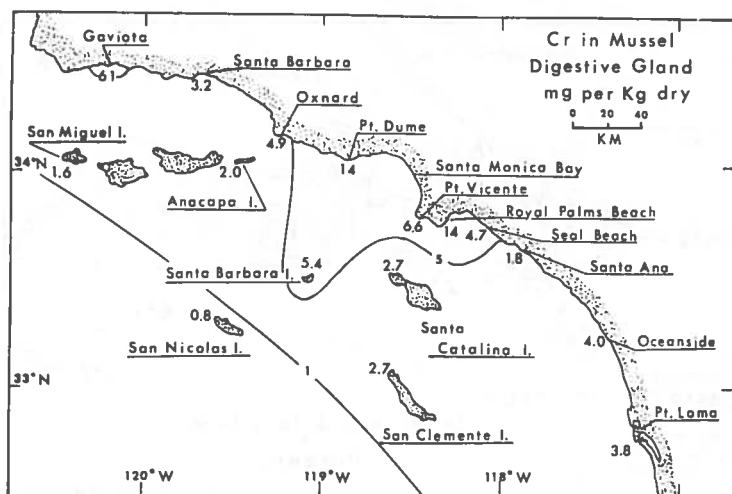
Fig. 9. Chromium in Mussel Digestive Gland—1971.

case of both of these elements, it would appear that the relatively high advective input may contribute to the inability to find regional differences.

In general, it appears that the digestive gland of *M. californianus* can be a useful tool in defining levels of marine pollution in cases where the pollution input is significantly higher than the advective input. In our 1971 survey, this certainly was the case for the elements Cu, Cr, Pb, and Ag.

In 1974 a similar study was initiated to determine the contributions of 'within harbor' sources to the pollution of the Bight. The harbor mussel, *Mytilus edulis*, was used to monitor the elemental levels. Two commercial harbors, Los Angeles-Long Beach and San Diego and one recreational harbor, Newport, were chosen for study. Fifteen monitoring sites were selected within and near these three harbors. Six 4-6 cm specimens were collected from each of these sites and dissected into digestive gland, gonad, adductor muscle and remaining tissue. Each sample was freeze-dried and analyzed for twenty-five elements.

Figure 10 shows the level of chromium observed in the digestive gland of samples collected in and near the Los Angeles-Long Beach harbor. A comparison of the 8 ppm level in the main Los Angeles channel with the 2 ppm level observed in Fish Harbor, the site of several canneries, suggests that a significant amount of chromium comes into the harbor by way of the Dominguez Channel. However, as found in 1971, the level near the Whites
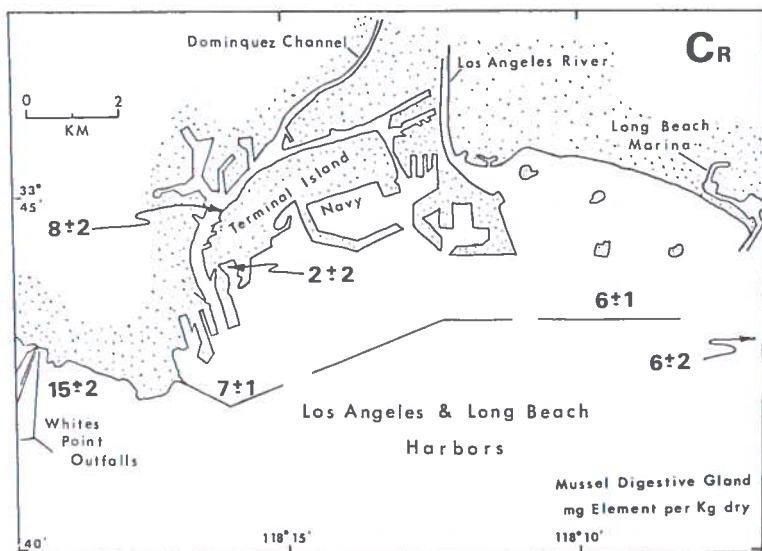
Fig. 10. Chromium in Mussel Digestive Gland in Los Angeles-Long Beach Harbor—1974.

Point outfalls is about two to three times the level found for nearby Seal Beach and along the breakwater. Nickel gave similar results with the outfall area being about twice the values for the harbor area (10 ppm *versus* 3-5 ppm). Copper showed only a slight elevation at the outfall in comparison to the harbor (47 ppm *versus* 29-36 ppm). Lead and zinc showed about the same values for the entire area (9-15 ppm Pb and 100-160 ppm Zn).

Figure 11 shows the level of copper in the mussel digestive glands at six stations near and in San Diego harbor. On the average, the harbor values are over twice the values found for sites outside of the harbor. The highest value, 73 ± 21 mg/Kg dry weight, was observed in the northern portion of the bay along the main channel. This site is at the mouth of a basin harboring several major yacht repair facilities, and it is reasonable to assume that this high copper level may be associated with the removal and application of large quantities of antifouling paint. Chromium and lead were higher in the commercial dock and large shipyard area than at other locations in the harbor or outer sites. The chromium level was 15 ppm *versus* 4-7 ppm while the lead level was 22 ppm *versus* 6-13 ppm. Nickel was higher at the commercial dock and yacht harbor areas and at the northern coastal point than at Point Loma
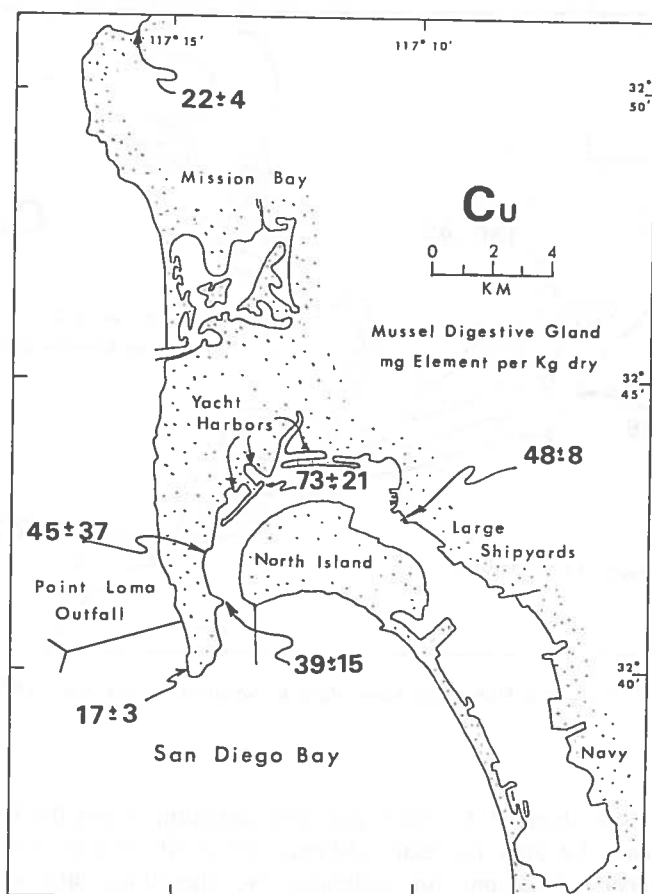
Fig. 11. Copper in Mussel Digestive Gland in the San Diego Harbor Area—1974.

and the remaining harbor locations (5-8 ppm *versus* 1-3 ppm). Zinc was slightly higher at the Ballast Point location in the entrance channel to San Diego Harbor than at any other location (210 ppm *versus* 120-160 ppm). The major activity in this area is associated with marine research vessels and submarines.

Figure 12 shows the copper level in the digestive gland of the *M. edulis* at one site within the recreational yacht harbor at Newport, Calif. and two sites outside of the harbor. This approximately sevenfold elevation within the harbor was also observed in other tissues of these samples. This large
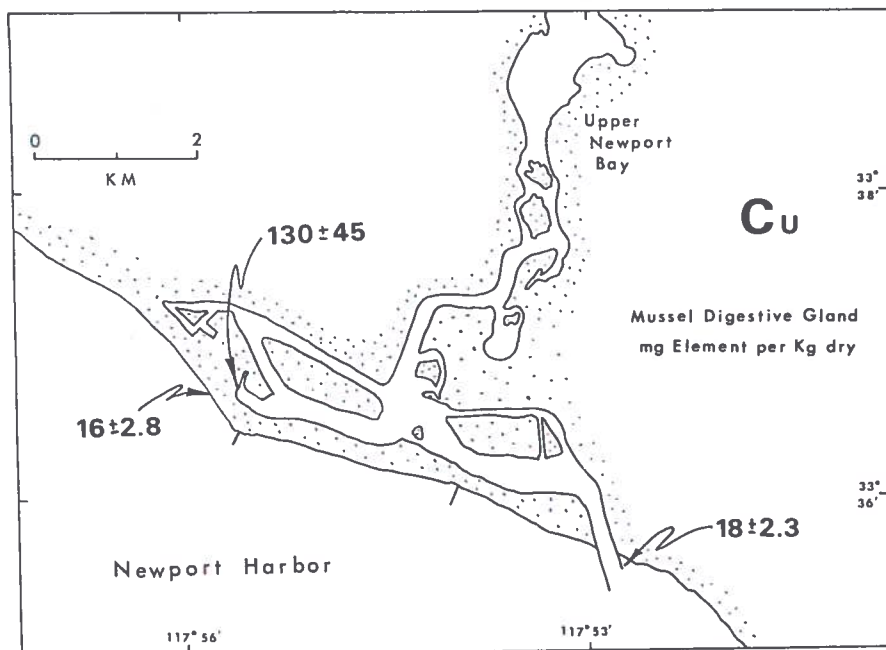
Fig. 12. Copper in Mussel Digestive Gland in Newport Harbor Area—1974.

differential was observed for lead, zinc and cadmium where the levels were 19 ppm *versus* 5-6 ppm for lead, 240 ppm *versus* 80-120 ppm for zinc, and 9.3 ppm *versus* 0-0.3 ppm for cadmium. No significant differences were observed for chromium, 3.7-4.1 ppm, or nickel 1-3 ppm. Measurements of the PCB 1254 levels in these tissues also indicated this high differential (880 *versus* 100 $\mu$g/Kg) (Young and Heesen 1974). It seems likely that these high levels of PCB, copper, lead, and zinc are associated with the antifouling paints used on yachts. In fact, this harbor site is quite close to one of the major yacht repair facilities.

In 1971 it became apparent from regional trawling surveys that there were areas within the Bight where the prevalence of fin erosion in a flatfish, the Dover Sole (*Microstomus pacificus*), was quite high (SCCWRP 1973; Mearns and Sherwood 1974). As can be seen from Fig. 13, this high incidence appeared to be related to the wastewater emission from the Los Angeles area. Repeated trawl surveys through 1975 confirmed this relationship and emphasised that a very high incidence was associated with the Palos Verdes shelf and
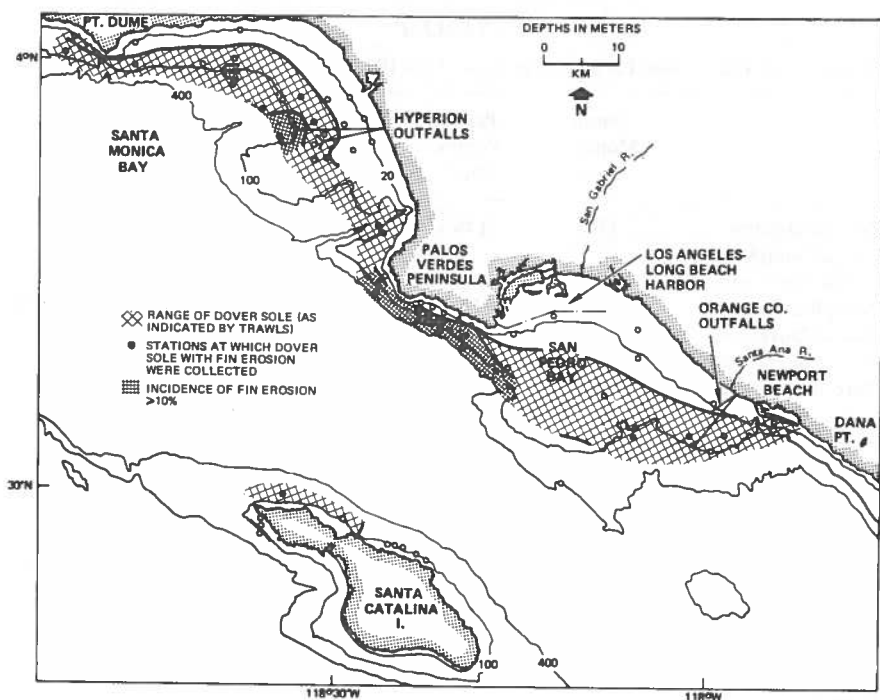
Fig. 13. Incidence of Fin Erosion in Dover Sole Collected in the Trawling Surveys of Southern California Coastal Waters, August Through October, 1971.

quite possibly the wastewater from the Whites Point outfalls, Table 1 (SCCWRP 1975).

The chance that trace metal pollutants might be associated with this high incidence prompted the following study. Diseased and normal Dover Sole samples were collected during March of 1973 from the Palos Verdes shelf. The samples were dissected into muscle, heart, liver, gonads and kidney and then freeze-dried. As before, each tissue was analyzed for approximately 25 elements. In most cases, the tissue levels were essentially the same; however, as shown in Table 2, chromium and lead gave an indication of being related to the fin rot disease (SCCWRP 1974).

In the hope of confirming these observations, another group of samples was collected between February and March of 1975. Dover Sole samples from the Dana Point area were included to serve as a basis for a regional comparison. In this comparison, differences in trace metal or major cation

TABLE 1

Summary of Fin Erosion Prevalence in Dover Sole 1972 - 1975.

| | Santa Monica Bay | Palos Verdes Shelf | San Pedro Bay | Dana Point | Point Loma | Santa Catalina Island |
|---|---|---|---|---|---|---|
| No. of Samples | 60 | 144 | 73 | 78 | 8 | 28 |
| No. of Samples with Dover sole | 39 | 106 | 44 | 45 | 7 | 16 |
| Sampling Period | 1972-73 | 1972-73 | 1972-73 | 1972-74 | 1975 | 1972-74 |
| No. of Dover sole | 516 | 14,277 | 3,842 | 881 | 64 | 135 |
| No. with eroded fins | 9 | 5,594 | 88 | 6 | 0 | 0 |
| Percent with eroded fins | 1.7 | 39 | 2.3 | 0.68 | 0 | 0 |

TABLE 2

Chromium and lead (mg/dry kg) in the tissues of diseased and apparently healthy Dover Sole from the Palos Verdes shelf.

| | Chromium | Lead |
|---|---|---|
| Gonads | | |
| Diseased | < 0.2 | — |
| Healthy | 0.9 | — |
| Heart | | |
| Diseased | < 0.2 | — |
| Healthy | 0.4 | — |
| Liver* | | |
| Diseased | 0.6 | — |
| Healthy | 0.2 | — |
| Kidney | | |
| Diseased | 0.5 | 6.0 |
| Healthy | 0.2 | 3.1 |

*Difference is significant at the 90% confidence level. All other values are significant at the 95% confidence level.

levels were considered to be associated with fin erosion if, for a given element and tissue, significant statistical differences ($p \leqslant 0.05$) were observed between the concentrations of elements in the diseased Palos Verdes fish and the unaffected fish from both Palos Verdes and Dana Point. On the other hand,

TABLE 3

Concentrations of trace metals and major cations (mg/dry kg) in tissues of fin eroded and apparently unaffected Dover Sole collected off Palos Verdes and Dana Point, February-March, 1975.*

| | Muscle | Kidney | Gonad | Skin Eyed Side | Skin Blind Side |
|---|---|---|---|---|---|
| **Trace Metal** | | | | | |
| **Cadmium** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | —** | <3.2 | <5.2 | <3.1 | <3.6 |
| Unaffected | <1.8 | <3.2 | <5.0 | <3.9 | <3.0 |
| Dana Point | | | | | |
| Unaffected | <3.0 | <4.0 | <4.2 | <4.8 | <4.6 |
| **Chromium** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | —** | 0.2 | 1.3 | 1.0 | 0.8 |
| Unaffected | —** | 0.2 | 1.5 | 0.9 | 0.7 |
| Dana Point | | | | | |
| Unaffected | —** | —** | —** | 0.3 | 0.4 |
| **Copper** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | 0.9 | 19 | 7.2 | 1.5 | 2.2 |
| Unaffected | 1.0 | 19 | 12 | 1.4 | 1.6 |
| Dana Point | | | | | |
| Unaffected | 1.0 | 5.9 | 13 | 1.4 | 1.6 |
| **Lead** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | —** | <1.7 | <1.2 | <1.0 | <0.5 |
| Unaffected | —** | 1.4 | <1.5 | <1.3 | <1.1 |
| Dana Point | | | | | |
| Unaffected | —** | <1.2 | <0.8 | <1.8 | <0.4 |
| **Major Cation** | | | | | |
| **Calcium** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | 640 | 1,300 † | 800 | 14,000 | 8,800 |
| Unaffected | 590 | 960 | 1,000 | 8,300 | 7,800 |
| Dana Point | | | | | |
| Unaffected | 370 | 450 | 870 | 9,600 | 4,900 |
| **Magnesium** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | 1,800 | 2,600 | 2,600 | 2,000 | 1,700 |
| Unaffected | 2,000 | 2,100 | 5,200 | 2,200 | 1,600 |
| Dana Point | | | | | |
| Unaffected | 2,100 | 2,200 | 5,000 | 2,700 | 2,100 |
| **Sodium** | | | | | |
| Palos Verdes | | | | | |
| Fin eroded | 18,000 | 22,000 | 19,000 | 6,900 | 8,400 |
| Unaffected | 17,000 | 22,000 | 24,000 | 7,700 | 5,400 |
| Dana Point | | | | | |
| Unaffected | 15,000 | 25,000 | 33,000 | 10,000 | 14,000 |

*Solid-line boxes mark regionally associated significant differences, $p \leqslant 0.05$; dashed-line boxes mark fin erosion associated significant differences, $p \leqslant 0.05$.

**Below the limit of detectability.

†Both regional and fin erosion associated significant differences, $p \leqslant 0.05$.

no statistical differences should be observed between the unaffected specimens from the two regions.

On this basis, only three significant differences associated with the occurrence of fin erosion were observed (see Table 3). These were in the gonad for copper and magnesium and in the kidney for calcium. The diseased Palos Verdes fish had significantly higher levels of these elements in the kidney and significantly lower levels in the gonad. The previous apparent correlation between fin rot and chromium and lead levels was not supported.

The rather large, twofold to threefold difference between the calcium level of the kidney at Palos Verdes and Dana Point leads to another possible relationship to fin erosion. It may be that this exceedingly high calcium level is indicative of high kidney stress or malfunction which, in a high proportion of the cases, results in fin erosion. These high levels are not observed in sport fish from the area such as white croaker, boccacio, Pacific sanddab, and kelp bass; where the range of calcium values is from 200 to 950 $\mu$g/g dry tissue. It is interesting to note that in chronic or acute renal failure in humans, the calcium level of the kidney increases from a mean of 515 to a mean of 630 $\mu$g/g dry tissue, while most of the other elements exhibit no significant change (Indraprasit *et al.* 1974).

This possibility, as well as others, continues to be investigated in the hope of eventually isolating the agent or agents causing this high incidence of fin erosion in the Dover Sole.

## ACKNOWLEDGEMENTS

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