THE RELATIONSHIP OF WASTEWATER CHLORINATION ACTIVITY TO DUNGENESS CRAB LANDINGS IN THE SAN FRANCISCO BAY AREA

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January 1977

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UCB/SERL Report No. 77-1

TABLE OF CONTENTS

Page

ABSTRACT

A dramatic decline in dungeness crab landings has occurred since the early 1960's. Potential causes for the population failure suggested by the literature include physical: temperature, salinity and currents; chemical: heavy metals, petrochemicals, pesticides, polychlorinated biphenyls and chlorinated wastewater effluents; and biologic factors: disease, parasitism and competition for food. Perdurant halogenated organics are formed in the disinfection of wastewaters with chlorine. Preliminary data indicate toxicity to aquatic organisms at ug/l levels of these compounds. To establish the relationship between wastewater chlorination activity and crab landings, a survey was conducted of municipal dischargers drained by the San Francisco Bay system. Significant increases in chlorine usage in municipal wastewater treatment were found to occur immediately preceding the dungeness crab population failure.

ACKNOWLEDGMENTS

This study was made possible by Sea Grant Number NOAA-04-6-158-44021, 1976.

LIST OF TABLES

LIST OF FIGURES

I. INTRODUCTION

PURPOSE AND SCOPE

The San Francisco Bay Area dungeness crab (*Cancer magister*) population has experienced a catastrophic decline in numbers over the last 15 years as evidenced by reduction in regional crab landings. Wide cyclic variations in crab numbers are characteristic of the species; however, the prolonged low catch yields in contrast to the normal yields of the more northerly populations, indicate a population failure in the San Francisco region (see Figure 1).

This report presents a review of the literature concerning potential factors responsible for the observed crab population dynamics. Additionally, the results of a survey of the major municipal wastewater discharges in the area is given to clarify the implication of chlorinated sewage in the crab population failure.

LIFE HISTORY

C. magister is found to a depth of 30 m (100 ft) from Central California to Alaska. Dungeness crabs of the San Francisco Bay region inhabit the Gulf of the Farallons for most of their adult lives. Females mate during molting but the spermatozoa are kept viable within a special receptacle of the female until fertilization occurs several months later. The eggs are soon extruded and carried externally until hatching. During this interval the gravid female often remains burried in the upper few inches of sand. Hatching in the San Francisco Bay Area occurs in December and January, several months earlier than with populations on the northern end of the species' range. Zoeal development takes approximately four months whereupon the planktonic individuals move, probably passively, into the estuarine environment as megalopae or as first or second-stage instars. This migration through the Golden Gate occurs in late spring or early summer. Growth as juveniles in the San Francisco Bay system progresses for about a year followed by a return migration to the Gulf of the Farallons.

Sexual maturity in *C. magister* is attained in one year but large-scale breeding activity is not likely until the following year. California crabs enter the fishery at 3 years of age. It is generally believed that most of the legal size males are landed each year. Thus sub-legal size males form an important part of the breeding population [2,3].

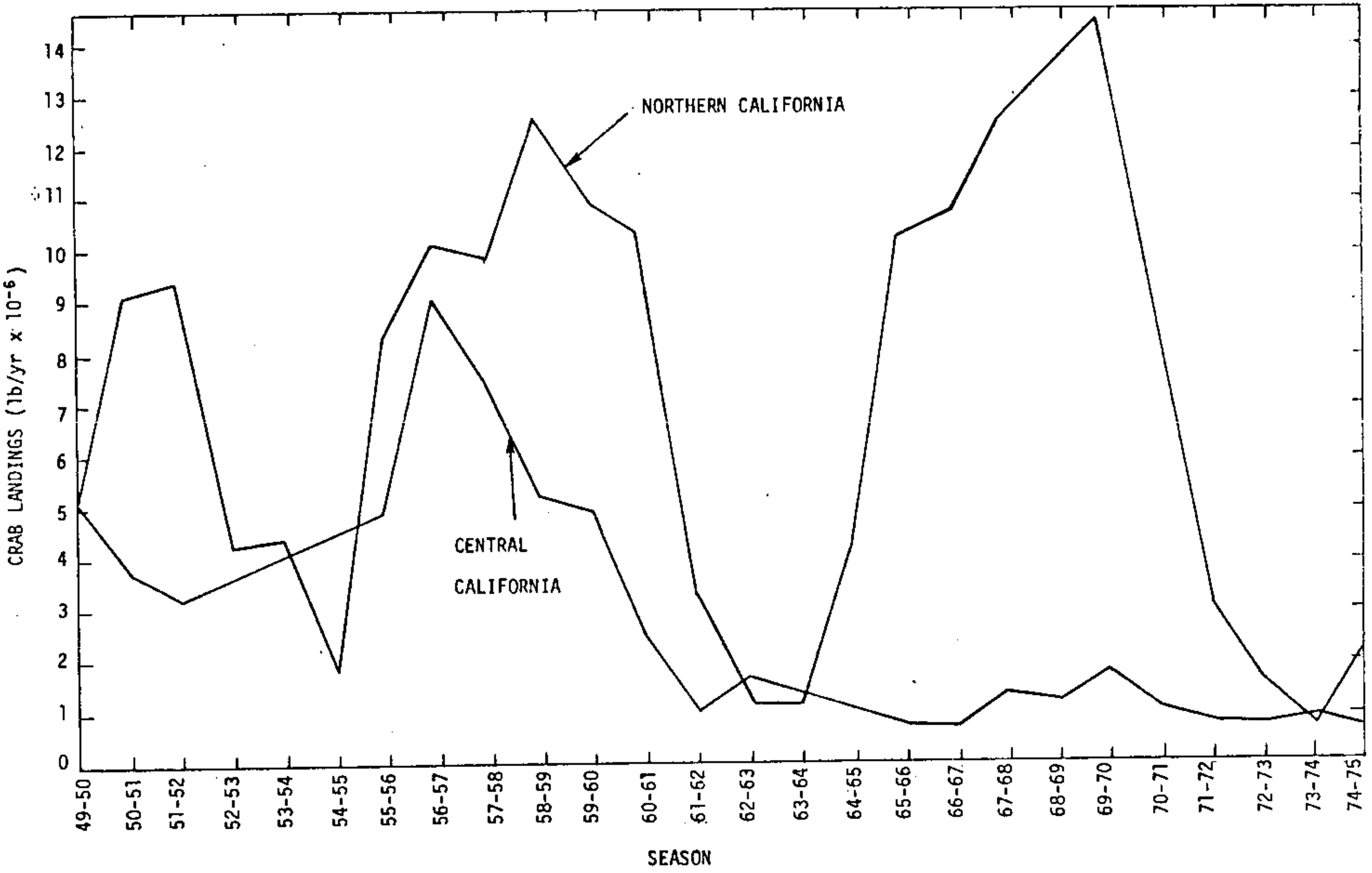


FIGURE 1. CRAB LANDINGS IN CENTRAL AND NORTHERN CALIFORNIA [1]

II. LITERATURE REVIEW

GENERAL

San Francisco is near the southern limit of dungeness crabs' range. Here conditions may be suboptimal for the crabs' survival and only a slight increase in stress, be it temperature, salinity, a pollutant, or parasitism, may be critical. In addition, the migration of dungeness crabs into and out of the San Francisco Bay system exposes them to varying environmental conditions. It is well established that an organism able to tolerate stresses such as man-made pollution in an optimal setting may be unable to survive the same stress when exposed to suboptimal physical conditions, e.g., fluctuating temperature or salinity. The changing parameters of the San Francisco estuary may form this type of suboptimal environment for dungeness crabs.

Physical, chemical, and biological factors all may influence crab population dynamics. The position of dungeness crabs high on the food chain increases their vulnerability to chemical pollutants that are biologically concentrated in passing from producers to consumers. The difficulty in identifying the reason for the population failure is further complicated by the probability that simultaneous stress are synergistic rather than additive. It may be necessary to look at combinations of variables rather than individual stresses for the solution to the problem.

PHYSICAL FACTORS

Temperature, salinity, and currents are the most obvious physical factors that could influence dungeness crab viability. Changes in these parameters in Bay waters may have resulted from diversions and impoundment of tributary streams although sufficient data to document this hypothesis are lacking. Relatively minor long-term physical changes in the Bay over the past few decades may have gone unnoticed. Physical changes to water in the Gulf of the Farallons are less likely. Dilution by coastal currents provides a high degree of stability to the physical condition of these open waters.

The salinity tolerance of dungeness crabs reflects its capacity for maintaining its internal ionic and osmotic homeostasis. Both active and passive physiological mechanisms are involved in controlling internal ionic and osmotic tonicity. Temperatures of the poikilothermic dungeness crabs are exposed to largely influence their metabolic activity. Behavioral patterns may also be dictated by the temperature regime, i.e., mobility, depth of occurrence, and cannibalism.

The temperature-salinity response of *C. magister* in the laboratory setting has been studied by several groups. Buchanan and Millemann observed greatest hatching success of dungeness crab eggs from Oregon coastal waters at 32 ppt salinity and 17.5 C (63.5 F) [4]. Also using *C. magister* from the Oregon population, Reed investigated larval development in the laboratory under various salinity and temperature regimes [5]. He reported maximum survival at salinities of 25 to 30 ppt and temperatures of 10-13.9 C (50-57 F). The shortest zoeal development time through the megalops stage, 45 days, occurred at 25 ppt salinity and 17.8 C (64 F). The osmotic and ionic regulation of Oregon *C. magister* was examined by Hunter and Rudy [6]. Smaller dungeness crabs were found to have a greater capacity for hyperosmoregulation than larger individuals. This ability to physiologically compensate for a dilute seawater environment undoubtedly plays an important role in the survival of juveniles in the San Francisco Bay system. They hypothesized that possibly gill surface area is involved, being one site of active salt regulation. The smaller the individual, the larger the gill surface area to body mass ratio. Evidently the physical environment of the Gulf of the Farallons favors survival of dungeness crabs. Full oceanic salinity and a temperature range of 10.5 C (50.9 F) in January to 15.5 C (63 F) prevail here. Lenthened zoeal development at these temperatures may permit greater current transport. The role of such migrations by *C. magister* is presently unclear.

Correlations of crab catches with coastal upwelling have been made to determine any effects nutrient import may have on productivity at *C. magister's* level in the food chain. Peterson concluded that summers of strong upwelling produce good catches 1.5 years later off Northern California and Oregon and 0.5 years later off the Washington coast [7]. He suggested that the lag in energy transport up the food chain allows the southerly populations to molt before benefiting from the increased food supply. With the Washington crabs, molting occurs later in the season permitting increased crab productivity to appear the same year. Botsford and Wickham reexamined the relationship and suggested that density dependent biologic factors rather than upwelling may be responsible for the cyclic variation in dungeness crab populations. They point out that auto-correlation of upwelling indices does not reveal significant periodicity and that the weak cycles suggested have different periods from those of the crab catches [8].

CHEMICAL FACTORS

The toxicity of chemical pollutants to *C. magister* may result from direct exposure to ambient dilutions of the substance or from the effective concentration that has accumulated in the crabs diet. Pollutant concentration in organism tissues may be orders of magnitude greater than in the environment. The effect of bioconcentration becomes increasingly pronounced in successively higher levels of the food chain. *C. magister* is a carnivore high on the aquatic food chain. Its diet includes shrimp, small crabs, barnacles, amphipods, isopods, clams, and worms [9]. These food sources, which are themselves consumers, are potential routes for bioconcentration of aquatic chemical pollutants.

Organic pollutants and heavy metals are the most conspicuous chemical pollutants in the San Francisco Bay region. The organics are released to the environment principally through: 1) processing, transportation and use of petroleum products; 2) application of pesticides; 3) manufacture and use of polychlorinated biphenyls (PCB's) and 4) municipal and industrial wastewater discharges. Heavy metals are found in the hydrosphere at elevated levels mainly from the discharge of municipal and industrial wastewaters.

Many organic compounds are characterized by poor solubility in water. The degree of solvation depends on the number and arrangement of nonpolar versus polar groups on the molecule. Organic compounds in the water tend to accumulate at interfaces and orient their nonpolar moieties toward the hydrophobic phase. Thus, the organics may be selectively absorbed and accumulated in the organisms lipid tissues. Transportation and metabolism of lipid reserves occurs at high rates in discrete developmental stages of the crab's life cycle. Allen noted that in *C. magister* hemolymph lipid concentrations are highest in females with developing ovaries [10]. Toxic organic pollutants accumulated in fatty tissue of the adult female may be transported and further concentrated in the eggs by this mechanism. Further study is indicated to assess the significance of this phenomenon in dungeness crabs.

Petrochemicals

Oil and related petrochemicals may enter the estuarine and marine environments through spills, ballast tank flushing, transfer operations, industrial waste discharges, and natural geological processes. Petroleum fractions released to the hydrosphere disperse by emulsification, solubilization, evaporation, and precipitation. The less stable components are subject to biological and photochemical degradation. Probably most of the oil entering the water ends up in a relatively narrow zone along the coastline. Some of the oil settles to the bottom here where certain fractions continue to be released to the shallow overlying waters for months or years [11].

Petrochemicals include a diverse mixture of hydrocarbons with variable solubilities, persistences and toxicities. The potential impact of these substances on aquatic organisms may be direct kill by coating, asphyxiation, or contact poisoning; direct kill by exposure to water soluble fractions; destruction of food sources; or sublethal effects such as reproductive failure and reduced resistance to infection and other stresses [11]. The literature is lacking in reports of petrochemical toxicity to *C. magister*, however studies using related marine invertebrates offer some quantitative indication of the probable significance to these crabs.

Using *Homarus americanus*, Atema and Stein found that 10 ppm crude oil stirred in seawater caused increased reliance on tactile sensing [12]. They hypothesized that the oil imparted a disagreeable taste or odor to the water masking the attractive properties of the food. Exposure of the lobster to the water soluble fraction alone produced no discernible effect.

Crude oil at 0.56 mℓ/ℓ resulted in 50% mortality within 48 hr (48 hr LD_{50}) to juvenile Prudhoe Bay tanner crabs, *Chionoecetes bairdi* [13]. Sublethal effects included reduced molting success at 0.32 mℓ/ℓ crude oil and extensive autotomy in recently molted juveniles who survived acute oil exposure. Undoubtedly crabs with fewer legs and chelae would be less competitive in feeding, defending, migrating and mating. Since molting is under hormonal control, they suggest the oil disrupts the crab's internal hormonal homeostasis.

Anderson, *et al.* studied the toxicities of oil-water despersions (OWD) and water soluble fractions (WSF) of various crude and refined oils on brown shrimp postlarvae, *Penaeus aztecus*, and the grass shrimp, *Palaemonetes pugio* [14]. Table I summarizes their findings.

TABLE I

TOXICITY OF CRUDE AND REFINED OILS TO SHRIMP

Oil Type	96-hr LD_{50} (ppm)	
	Penaeus aztecus	*Palaemonetes pugio*
Crude OWD	>1000	200 - 6000
Crude WSF	>19.8	>10.2 - >16.8
Refined OWD	9.4	3.0
Refined WSF	1.9 - 4.9	2.6 - 3.5

SOURCE: Anderson, *et al.* [14]

Although the concentrations of the WSF of the refined oils were lower than those of the crude oils, significantly higher concentrations of di- and tri-aromatic hydrocarbons are found in the former. They suggest the toxicity of oil is a function of its content of those hydrocarbons. The greater toxicity of crude WSF than crude OWD here contrasts the opposite results of Atema and Stein's lobster experiments. Direct investigation of the effects of oil on dungeness crabs is preferred over extrapolation from these apparently conflicting reports.

The effect of outboard motor effluent on the bivalve molluscs *Mytilus edulis* and *Ostrea lurida* was examined by Clark, *et al.* [15]. Fifty ppb n-paraffin hydrocarbon was measured in their test effluent versus 7.3 ppb in industrialized harbors. They suggest their experimental conditions represent the upper bound of concentrations expected in shallow estuaries with limited flushing set in the context of sediment-incorporated petroleum components in constant flux with the overlying waters. Although oyster mortality was 14% after 10 days continuous exposure, the mussel mortality reached 66% following only 1 day exposure to effluent and 9 days in fresh running sea water and 75% after 45 days. Both the 10-day oysters and 1-day mussels showed extensive gill tissue degeneration. In addition, the mussels experienced some dilation of the tubules of the digestive diverticulum.

Pesticides

Pesticide defines a broad spectrum of compounds including insecticides, herbicides, fungicides, molluscacides, nematocides and rodenticides. The persistence of pesticides varies with the type compound. Chlorinated compounds are usually quite stable, i.e., DDT, DDE, Methoxychlor and 2,4-D. Organophosphorus compounds such as Malathion, however, are relatively shortlived in the hydrosphere. Pesticide decomposition in the aquatic environment occurs through the processes of photochemical degradation, anaerobic and aerobic microbial degradation, and degradation due to metabolic processes within higher organisms [16].

The very slightly soluble DDT probably concentrates at the air-water interface. Algal and bacterial assimilation may effectively distribute DDT throughout the water column and introduce it to the food chain [11].

Sevin is a wide spectrum carbonate insecticide with a half life in natural sea water at 20 C (68 F) of 82 hr [17]. It acts as a cholinesterase inhibitor in arthropods. The effect of Sevin on the forms of *C. magister* has been reported by Stewart *et al.* [18, 19]. One mg/ℓ Sevin had no effect on hatching success but the effective concentration (EC_{50}) for disruption of molting from prezoeae to zoeae was found to be 0.006 mg/ℓ. The 96-hr LD_{50} for first-stage zoeae was 0.01 mg/ℓ and for late juveniles 0.25 mg/ℓ. Zoeae experienced delayed mortalities when held in clean water following Sevin exposure whereas the juveniles did not. Sixty percent survival of zoeae in concentrations as low as 0.0001 mg/ℓ for 25 days was obtained however molting was delayed. Larval molting was prevented by 0.01 mg/ℓ Sevin. The 24-hr LD_{50} for second instar juveniles was found to be 0.076 mg/ℓ , for ninth instar juveniles 0.35 - 0.62 mg/ℓ, and for adults 0.49 - 0.70 mg/ℓ. In 96-hours, 0.26 mg/ℓ Sevin resulted in 50% adult mortality. The byproduct of Sevin degradation, 1-naphthol was 30-300 times less toxic to *C. magister* than Sevin itself.

Chlorinated pesticides, especially DDT and DDE, are believed to act through breakdown of enzymatic pathways [11]. Little information is available on the effect of this type of pesticide on dungeness crabs, however some other crab species have been studied. Xanthid crab larvae, *Leptodius floridanus* and *Panopeus herbstii*, exhibited acute toxicity to 5 ppb Dieldrin [20]. Development of *L. floridanus* larvae was significantly delayed by 0.5 ppb Dieldrin with the first stage larvae being more sensitive than later forms.

Bookhout and coworkers observed sublethal effects with exposure to 0.01 ppb Mirex in larvae of *Callinectes sapidus* (blue crab) and *Rithropanopeus harrisii* (mud crab) [21,22]. One ppb was found to be acutely toxic to *C. sapidus* larvae. Residue analysis of Menippe mercenaria (stone crab) larvae reared in 0.01 ppb Mirex indicated 13,000 fold concentration of this substance in the crab's tissues. *C. sapidus* and *R. harrisii* were slightly less efficient concentrators of this very lipid-soluble chemical.

The pronounced effects of minute concentrations of insecticides on crabs is not surprising. Crustaceans and insects are phylogenetically related in Arthropoda and have many physiological processes in common. Thus chemical agents effective in disrupting the metabolic machinery of insects would likely be toxic to crabs as well.

Polychlorinated Biphenyls

Like some pesticides, PCB's are extremely stable chlorinated hydrocarbons and have similar chemical and toxic properties. Although not deliberately released to the environment, they are found in sewage discharges of regions where used in the manufacture of electrical components, paints and insulation pastes. There is evidence that degradation of polychlorinated pesticides produces PCB's but the extent of this accumulation is presently unclear. When adsorbed to marine sediments, PCB's are recycled to the water and aquatic organisms.

Pink shrimp larvae, *Penaeus duorarum*, are killed by 15 days exposure to 1 ppb Aroclor 1254 (54% degree of chlorination) and adults by 35 days exposure to 3.5 ppb [16]. Growth of *P. duorarum* in 2.5 ppb for 20-25 days gives a concentration of 25 ppm in the whole body and 500 ppm in the hepatopancreas. The later figure represents a 200,000 fold increase over the ambient levels.

Heavy Metals

Approximately two dozen metals are highly toxic to aquatic biota, most notably mercury, lead, arsenic, cadmium, chromium and nickel. In the water column they may occur in solution or in the chelated form. Precipitation is the most significant route of removal from the aqueous phase, but heavy metals may become reavailable through exchange with the sediment.

Most heavy metals are bioconcentrated and exert cumulative toxicity over a period of time. The effect of these elements is often dependent on their chemical form. Organic mercury compounds for example, are more toxic than elemental mercury or its inorganic compounds [11].

Rehwoldt *et al.* studied the effects of various heavy metals on benthic organisms of the Hudson River near Poughkeepsie [23]. In general, sensitivity of the scud, *Gammarus* sp., to these elements was found to be greater than that typical of fish. The scud 96-hr LD_{50} values reported were: Cu^{2+} - 0.91 mg/ℓ, Zn^{2+} - 8.1 mg/ℓ, Ni^{2+} - 13.0 mg/ℓ, Cd^{2+} - 0.07 mg/ℓ, Hg^{2+} - 0.01 mg/ℓ and Cr^{3+} - 3.2 mg/ℓ.

Low level exposure of fiddler crab larvae, *Uca pugilator*, to mercuric chloride has been shown to result in reduction of metabolic activity and swimming capability [24, 25]. Concentrations as low as 9×10^{-11} M mercuric chloride (18 ppb Hg^{2+}) produced limited mortality in the larvae. The observed sublethal effects on *U. pugilator* were found to be temperature dependent. Above 25 C (77 F) added mercury stress depressed metabolic rates, but at 20 C (68 F) oxygen uptake was increased. The clearing rate of inorganic mercury from dungeness crabs was measured following a 2-hour exposure to 500 ppb Hg^{2+} [26]. Twenty to 25 days was the half-life calculated here. No toxic response was reported.

Cadmium at 1.0 ppm and copper at 5.0 ppm were shown to lethal to the rock crab, *Cancer irroratus*, within 48 hours [27]. At 0.6 ppm copper osmoregulatory activity disappeared, but a similar effect was not found with cadmium concentrations as high as 2.0 ppm. Disruption of hyperosmoregulation became more pronounced at lower salinities. Although only cadmium depressed gill tissue oxygen consumption, copper turned the gills green indicating probable respiratory damage.

Wastewater

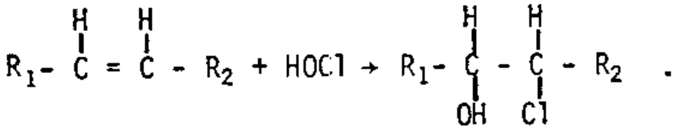
An array of compounds potentially toxic to dungeness crabs are released in municipal wastewater discharges. Many of these substances have been described above but special attention must be given to added toxicity resulting from the application of chlorine to wastewater. The bulk of chlorine added in sewage treatment serves as a disinfectant in the control of pathogenic microorganisms. Chlorine is also used in treatment processes for odor control, elimination of nuisance filamentous bacteria and in-house reuse of the treated wastewater. Many industrial wastewater discharges contain chlorine used both as a reagent and for maintenance of cooling and process water quality.

There are four principal reactions of chlorine with organic matter in dilute aqueous solutions [28]. These are: 1) addition of olefinic bonds, 2) activated ionic substitution, 3) oxidation with reduction of the hypochlorite chlorine to chloride and 4) substitution of chlorine for hydrogen on a nitrogen atom. Only in the last reaction does chlorine retain some of its oxidizing power. The other three reaction types convert the free chlorine either to chloride or to a covalently bonded state with carbon. Simple aliphatic substitutions such as

$$CH_3 - CH_3 + HOCl \rightarrow CH_3 - CH_2Cl + H_2O$$

are not expected. This type of reaction proceeds best in nonionizing media and usually requires much thermal energy or an initiator, and light.

Addition to olefinic bonds can be represented by



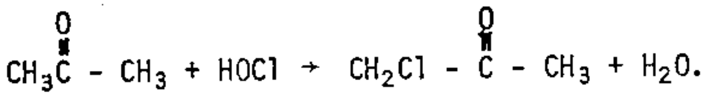
$$R_1-\overset{\mathrm{H}}{\mathrm{C}}=\overset{\mathrm{H}}{\mathrm{C}}-R_2 + HOCl \rightarrow R_1-\underset{\mathrm{OH}}{\overset{\mathrm{H}}{\mathrm{C}}}-\underset{\mathrm{Cl}}{\overset{\mathrm{H}}{\mathrm{C}}}-R_2 \; .$$

chlorinated organic compounds, and sometimes dichloro compounds, result from this type of reaction. These compounds are often quite stable in neutral aqueous environments and their toxicities are for the most part unknown.

Activated ionic substitution reactions can occur with aromatic compounds, exemplified by

$$C_6H_5OH + HOCl \rightarrow C_6H_4ClOH + H_2O$$

or as the haloform reaction, for example



$$CH_3\overset{\mathrm{O}}{\overset{\|}{\mathrm{C}}} - CH_3 + HOCl \rightarrow CH_2Cl - \overset{\mathrm{O}}{\overset{\|}{\mathrm{C}}} - CH_3 + H_2O.$$

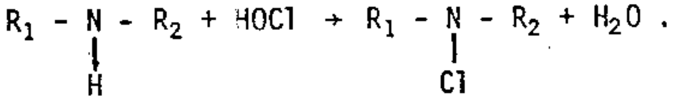
Substitution on aromatic compounds may involve phenol, its homologs and analogs, and possibly aromatic aldehydes and acids. The haloform reaction is a substitution of chlorine on organic compounds having an acetyl group or structures that may be oxidized to the acetyl group. Chloroform is the final product of exhaustive substitutions of the latter type. As with the addition reaction, substitution reactions produce chlorinated organic compounds which may persist after discharge.

Compounds oxidized by chlorine with the production of chloride include alcohols, aldehydes, carbohydrates, substances hydrolyzable to the hydroxylated form and those with sulfhydryl groups or other reduced sulfur linkages. The reaction type can be illustrated by

$$R - CHO + HOCl \rightarrow RCOOH + H^+ + Cl^- .$$

Since chloride rather than chlorinated organics is obtained, toxicity to aquatic organisms is of little concern in most cases. It is felt that these oxidation reactions are responsible for most of the chlorine demand in municipal wastewaters.

The fourth reaction type, leading to the formation of N-chlorinated compounds, may occur with amines, amides, amino acids, proteins, and heterocyclic compounds. The generalized type form is



$$R_1 - \underset{H}{\underset{|}{N}} - R_2 + HOCl \rightarrow R_1 - \underset{Cl}{\underset{|}{N}} - R_2 + H_2O .$$

These reactions may be quite rapid, particularly with the more basic nitrogen atoms. In the case of ammonia, milligram per liter concentrations of chlorine react to form chloramine in about a minute. Unless oxidized by other wastewater components, N-chlorinated products of reactions with simple primary or secondary amines may persist for several days after discharge.

Traditionally, wastewaters have been chlorinated to a measured residual of 0.5 to 2.0 mg/ℓ. A change in treatment standards now requires dechlorination to 0 mg/ℓ residual before discharge although many facilities have yet to implement this process modification. Chlorine residual, the capacity for oxidizing KI_2 to I_2, measures only unreacted hypochlorite and the products of substitution for hydrogen on nitrogenous compounds. Dechlorination to 0 mg/ℓ residual chlorine does not imply that compounds with the C-Cl bond have been destroyed, nor that any toxicity associated with them has been mitigated.

Identifications of chlorinated organics produced in wastewater treatment have been made. Table II lists a set of compounds identified from superchlorinated wastewater samples by Glaze and Henderson [29]. Samples were dosed at 1500 mg/ℓ chlorine and extracts from Amberlite XAD-2 resin were gas chromatographed and compared with unchlorinated samples. Tentative identification of the 30 chlorinated compounds found indicated chlorine substitution on aromatic derivatives such as benzene, toluene, and benzyl alcohol. Three chlorinated acetone derivatives were also isolated which may be precursors of chloroform. Based on microcoulometric data, they estimated that only 20 to 25% of the total chlorinated organic concentration was measured.

Jolley used ^{36}Cl labelling and anion exchange chromatography to resolve chlorinated organics from primary and secondary treatment effluents [30]. Forty-seven and 44 compounds, respectively, were found with 35 compounds

TABLE II

QUALITATIVE-QUANTITATIVE ANALYSIS OF CHLORINATED ORGANICS IN WASTEWATER EFFLUENT

Compound Name	Concentration (μg/ℓ)
Chloroform	--
Dibromochloromethane	--
Dichlorobutane (—)	27
3-chloro-2-methylbut-1-ene	285
Chlorocyclohexane (118)	20
Chloroalkyl acetate (—)	--
O-dichlorobenzene	10
Tetrachloroacetone	11
P-dichlorobenzene	10
Chloroethylbenzene	21
Pentachloroacetone	30
Hexachloroacetone	30
Trichlorobenzene	--
Dichloroethyl benzene	20
Chlorocumene (154)	--
N-methyl-trichloroaniline	10
Dichlorotoluene	--
Trichlorophenol	--
Chloro-α-methyl benzyl alcohol	--
Dichloromethoxytoluene	32
Trichloromethylstyrene (220)	10
Trichloroethyl benzene (208)	12
Dichloro-α-methyl benzyl alcohol (190)	10
Dichloro-bis(ethoxy)benzene (220)	30
Dichloro-α-methyl benzyl alcohol (190)	--
Trichloro-N-methylanisole	--
Trichloro-α-methyl benzyl alcohol	25
Tetrachlorophenol	30
Trichloro-α-methyl benzyl alcohol	50
Trichlorocumene (222)	--
Tetrachloroethylstyrene (268)	--
Trichlorodimethoxybenzene (240)	--
Tetrachloromethoxytoluene (258)	40
Dichloroaniline derivative (205)	13
Dichloroaromatic derivative (249)	15
Dichloroacetate derivative (203)	20
Trichlorophthalate derivative (296)	--
Tetrachlorophthalate derivative (340)	--

SOURCE: Glaze and Henderson [29].

occurring in common to both types of wastewater. One of the peaks resolved was chloride in each case. Table III lists tentative identifications and concentrations of 17 of the compounds. All of the compounds identified contain only a single chlorine atom making them generally more biodegradable than polychlorinated materials. Jolley dechlorinated one set of samples with thiosulfate before concentration and chromatographing. Fifty-two chlorine containing constituents were then separated. Thirty-nine of these, including all the major constituents, were in common with the undechlorinated secondary effluent samples.

The toxicity of these exotic chlorinated organics is largely unknown. Two of Jolley's compounds however, 4 chlororesorcinol and 5-chlorouracil, were bioassayed for carp egg hatchability (*Cyprinus carpio*) [31]. The lowest concentration tested, 1 μg/ℓ, significantly lowered hatchability at 21 C in both cases. Eggs fertilized and hardened in clean water for 30 minutes before addition of the test solution fared better as did eggs exposed to chlorouracil at 26 C. Possible effects of lower concentrations were not examined.

The biocidal character of chlorinated wastewater effluents has been well documented. The example of Sacramento, California is a good case in point. One of the city's treatment plants discharging to the Sacramento River dechlorinates its effluent, but another plant whose outfall is about 8 km (5 mi) downstream does not. Aufwuchs growth below the dechlorinated discharge is plentiful and is apparently being promoted by organic matter and nutrients in the effluent. Attached growth at the lower outfall is virtually absent as was the case at the upstream site before dechlorination was initiated. The implication is clear that compounds associated with chlorine residual are toxic to attached river microbiota.

A similar situation in the Little Patuxent River, Maryland was described by Tsai [32]. Increases in chlorinated wastewater discharged to the river between 1958 and 1967 were found to limit fish migration and alter the number and species of resident fish. Reaches of the River directly below chlorinated effluent outfalls showed the acute effects while farther downstream, where the chlorine had become sufficiently dissipated, the effects were much less pronounced. DO, temperature, dissolved and suspended solids, BOD, and pH were similar in both types of reaches.

Several bioassays of toxicity from chlorinated municipal effluents have been conducted at the University of California, Berkeley, Sanitary Engineerin Research Laboratory [33,34]. Esvelt's group measured the 96-hr LD_{50} for chlorine residual at 0.2 mg/ℓ for golden shiners. More recently Stone, Kaufman, and Horne found that 0.06 mg/ℓ of chlorine residual reduced aufwuchs biomass to 30% of the controls. Phytoplankton chlorophyll *a* was reduced by about 50%. At higher levels of residual chlorine, no aufwuchs growth was observed after 14 days exposure. In both studies dechlorination removed all observed toxic effects.

Muchmore and Epel examined fertilization success of the sea urchin, *Strongylocentrotus purpuratus* exposed to chlorinated sewage. A concentration of 0.055 ppm chlorine significantly reduced fertilization success after 5 minutes but it is unclear whether they were referring to residual chlorine or the amount applied. Similar results were obtained when seawater was chlorinated to the same degree indicating that chlorine rather than some other

TABLE III

TENTATIVE IDENTIFICATIONS AND CONCENTRATIONS OF CHLORINE-CONTAINING CONSTITUENTS IN CHLORINATED EFFLUENTS

Identification	Concentration of Organic Compound (μg/ℓ)
5-Chlorouracil	4.3
5-Chlorouridine	1.7
8-Chlorocaffeine	1.7
6-Chloroguanine	0.9
8-Chloroxanthine	1.5
2-Chlorobenzoic acid	0.26
5-Chlorosalicylic acid	0.24
4-Chloromandelic acid	1.1
2-Chlorophenol	1.7
4-Chlorophenylacetic acid	0.38
4-Chlorobenzoic acid	1.1
4-Chlorophenol	0.69
3-Chlorobenzoic acid	0.62
3-Chlorophenol	0.51
4-Chlororesorcinol	1.2
3-Chloro-4-hydroxybenzoic acid	1.3
4-Chloro-3-methylphenol	1.5

SOURCE: Jolley, [30].

wastewater component was responsible. Dechlorination with sodium thiosulfate removed the toxic effect. Sperm susceptibility proved to be the critical factor since fertilization failure occurred when the sperm alone were exposed to test solutions but not with the eggs. A tendency for flagella detachment was observed suggesting that necessary sperm motility had been impaired. Muchmore and Epel report similar results in fertilization experiments with the echiuroid *Urechis caupo* and the polychaetous annelid *Phragmatopoma californica.*

It must be emphasized that the data presented here on chlorinated wastewater effluent toxicity were derived from studies designed to demonstrate the short-term effects. One can expect to document chronic, long-term effects only when observations are conducted in terms of a generation time or longer. Little information is available in the literature concerning the latter phenomena.

BIOLOGICAL FACTORS

Dungeness crab population dynamics would reflect any changes in its relationship with other species of organisms. Disease, parasitism, predation, and competition for food and space may all be altered in response to an environmental change. Indeed, the characteristic cyclic nature of the population levels of *C. magister* may be an artifact of the numbers or activity of another species. The effect of pollution on other organisms in the dungeness crab's community could as likely alter the crab population balance as direct toxicity to the crab itself.

Crab diseases have been attributed to viruses, bacteria, fungi, protozoa (gregarines, microsporidans and citiates) and helminths (trematodes, cestodes, nemerteans and leeches). Infestations of other crustaceans including rhizocephalans, isopods and copepods have also been reported on crabs. The rock crabs *Cancer borealis* and *C. irroratus*, have been observed harboring the copepod *Choniosphaera cancrorum* [36]. The bacterium *Leucothrix mucor* is known to infest the eggs and bodies of the crabs *Carcinus maenas, Cancer borealis* and *C. irroratus*. The planktonic forms of these crabs seem to be less susceptible to *L. mucor* than benthic forms [37]. Nemertean worms are known to inhabit the adults and egg masses of a variety of crab species [38–40]. On the Atlantic coast of the United States, *Carcinonemertes carcinophila* is associated with lady crabs (*Plationichus ocellatus*) and blue crabs (*Callinectes sapidus*) among others. A different nemertean species, *C. epialti*, has been found in the Pacific on the common kelp crab, *Epialtus productus*.

Fisher and Wickham the University of California, Bodega Marine Laboratory have reported several organisms living within the egg masses of *C. magister* [41]. *C. epialti* was the only species identified but also present were a vorticellid protozoan and numbers of filamentous organisms resembling the blue-green alga *Oscillatoria* and the bacterium *Leucothrix*. Their data suggest that epibiotic fouling decreases in the more northerly dungeness crab populations.

III. METHODS

The chlorination histories of 27 municipal wastewater treatment plants discharging to waters drained by the San Francisco Bay estuary were obtained. Twenty-five treatment plants within Region 2 of the California Water Quality Board were included representing approximately 83% of the dry weather flow currently being discharged in the jurisdiction. Municipal dischargers in Sacramento and Stockton were also surveyed. The City of Fresno has traditionally disposed of its wastewater effluent in terminal ponds and is excluded from consideration here. The data resulting from the survey is presented in the Appendix. The locations of the 25 Region 2 Water Quality Board discharges are illustrated on Figure 2 and keyed with Table IV.

Some facilities, particularly those with less than 10 mgd flow, kept incomplete records in which case recollections of superintendents and operators were used. Interpolation on the basis of flow data was necessary in some instances.

The annual quantity of chlorine applied was chosen as the independent variable rather than measured residual. The chlorine residual test is often inaccurate due to interferring substances and compounds having the C-Cl bond are not necessarily detected. Chlorine residual data is not felt to be the best indicator of chlorine loading to the hydrosphere. Chlorine consumption for treatment purposes other than disinfection is included in the computations although this quantity is usually less than 10-20% of the total usage.

FIGURE 2. LOCATION OF MUNICIPAL DISCHARGERS SURVEYED IN REGION 2 OF THE CALIFORNIA WATER QUALITY BOARD

TABLE IV

KEY TO MUNICIPAL DISCHARGERS SURVEYED IN REGION 2 OF THE CALIFORNIA WATER QUALITY BOARD

Number	Region
1.	San Francisco, Richmond-Sunset Sewage Treatment Plant[a]
2.	San Francisco, North Point Sewage Treatment Plant[a]
3.	San Francisco, Southeast Sewage Treatment Plant
4.	City of San Mateo
5.	City of Palo Alto
6.	City of Sunnyvale
7.	San Jose - Santa Clara Sanitary District
8.	City of Hayward
9.	Ora Loma Sanitary District
10.	East Bay Municipal Utilities District[a]
11.	City of Richmond[a]
12.	San Pablo Sanitary District[a]
13.	Town of Pinole[a]
14.	Central Contra Costa Sanitary District
15.	Vallejo Sanitation and Flood Control District[a]
16.	Napa Sanitation District[a]
17.	Sonoma Valley County Sanitation District[a]
18.	City of Petaluma[a]
19.	Sanitary District No. 6 of Marin County, Ignacio[a]
20.	Sanitary District No. 6 of Marin County, Novato[a]
21.	Las Gallinas Valley Sanitary District, San Rafael[a]
22.	San Rafael Sanitary District[a]
23.	Sanitary District No. 1 of Marin County[a]
24.	City of Mill Valley[a]
25.	Sausalito - Marin City Sanitary District[a]

[a] Facility discharging directly to San Pablo and North San Francisco Bays.

IV. RESULTS AND DISCUSSION

The annual chlorine usage in treatment plants drained by the San Francisco Bay estuary since the 1949-50 season is presented in Table V. Also included is a tabulation of the quantities attributable to facilities discharging directly to the shoreline of and smaller tributaries of San Pablo and North San Francisco Bays. Available data indicate this portion of the estuary provides nearly 80% of the recruits to commercial stocks in the Gulf of the Farallons [42]. Figure 3 illustrates the survey data and includes data on crab landings in the San Francisco area for comparison. Since all treatment plants drained by the Golden Gate were not contacted, the absolute quantities of chlorine discharged are greater than is shown in Table V and Figure 3 but the percentage unaccounted for is believed to be relatively constant over the study period.

To clarify the relationship between crab landings and chlorination activity the percentage increase in chlorine usage was calculated between the 1956-57 season, the most recent period of probable normal landings, and the 1962-63 season, when disruption of the characteristic population cycle becomes apparent. The exposure of dungeness crabs to chlorinated municipal effluent is greatest to juveniles residing in the San Francisco estuary. Any population decrease to these forms would not be reflected in crab landings until the age class reaches the minimum legal size. Hence, the percentage increase in chlorine usage was also calculated for lag periods of up to 6 years. the results of these computations are given in Table VI.

The increase in chlorination activity in a framework of 1 to 6 years lag time to maturity from the juvenile form is 150% to 330% directly to San Pablo and North San Francisco Bay waters and 140% to 370% considering all facilities. Clearly these increases are significant. Whether these elevated levels of chlorine loading are implicated in the crab population dynamics cannot be demonstrated here. The fact remains however, that the hypothesis that increased municipal wastewater chlorination activity precipitated a failure of the regional crab population is not eliminated from consideration. The data strongly suggest that a cause and effect relationship exists. Many pollution parameters of the San Francisco region, particularly those that are a function of human population, have increased in the same time interval. This and other aspects of the San Francisco region's water pollution history in perspective of the dungeness crab population failure demand further study.

TABLE V

CHLORINE USAGE BY DISCHARGERS DRAINED BY THE SAN FRANCISCO BAY ESTUARY

Season	Usage on San Pablo And North San Francisco Bays (T/day)	Total Usage (T/day)
75-76	17.7	37.5
74-75	16.5	35.7
73-74	16.9	37.0
72-73	16.2	36.2
71-72	16.7	36.9
70-71	17.4	38.1
69-70	17.1	33.4
68-69	14.0	23.3
67-68	13.8	22.7
66-67	10.8	18.9
65-66	8.2	14.6
64-65	7.4	13.7
63-64	6.9	12.5
62-63	6.1	11.7
61-62	5.7	11.3
60-61	5.6	11.0
59-60	5.8	11.0
58-59	5.4	10.3
57-58	4.6	9.2
56-57	4.0	8.6
55-56	3.7	8.1
54-55	3.6	7.3
53-54	3.3	7.0
52-53	3.6	4.8
51-52	1.4	2.5
50-51	1.4	2.4
49-50	0.6	1.6

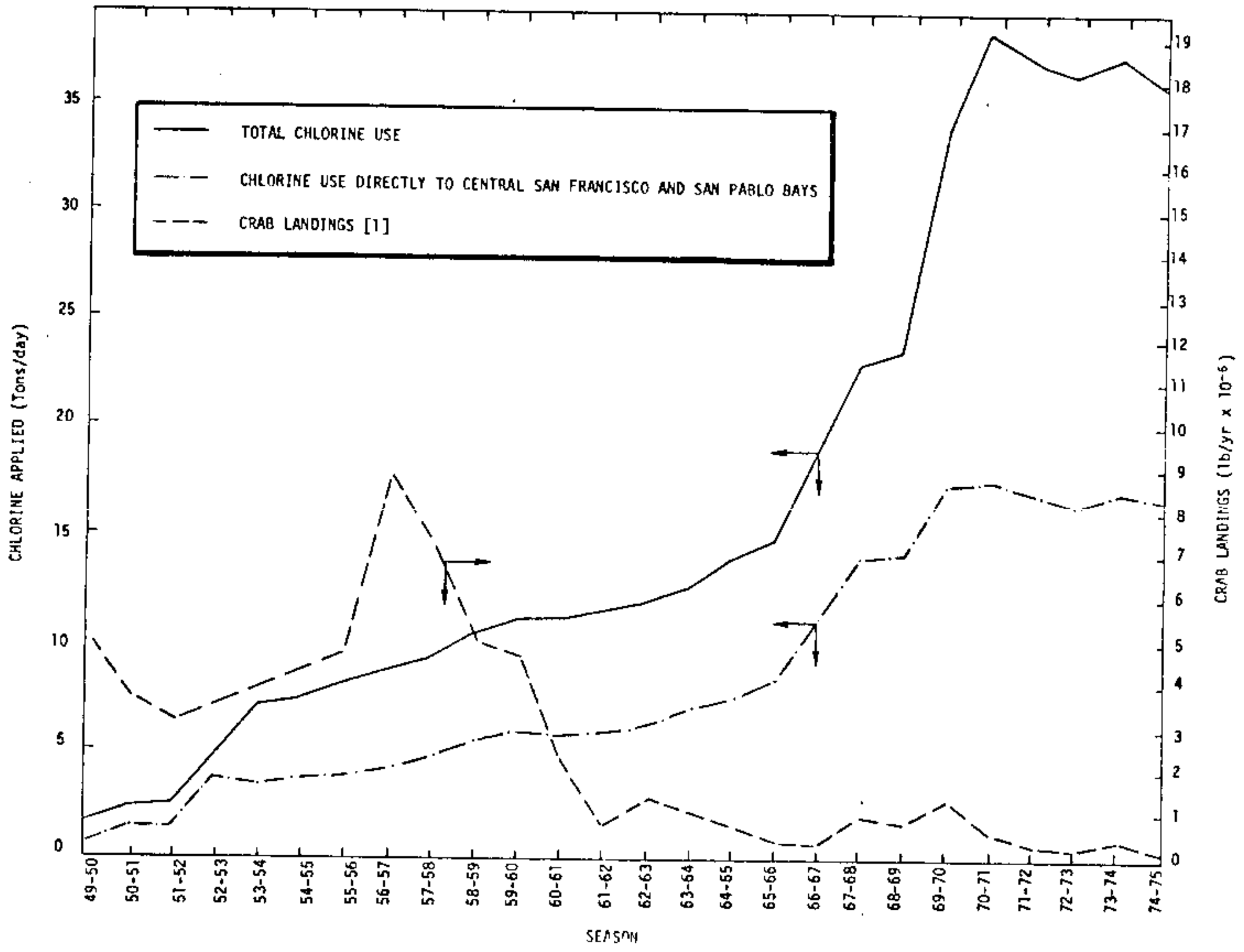


FIGURE 3. CRAB LANDINGS AND CHLORINE USAGE IN THE SAN FRANCISCO BAY AREA

TABLE VI

PERCENTAGE INCREASE IN CHLORINE USE OVER SIX YEAR PERIODS PRECEDING CRAB POPULATION DECLINE

Period (Seasons)	Lag (years)	Directly to Bay %	Total %
56/57 - 62/63	0	1.50	1.40
55/56 - 61/62	1	1.50	1.40
54/55 - 60/61	2	1.60	1.50
53/54 - 59/60	3	1.80	1.60
52/53 - 58/59	4	1.50	2.10
51/52 - 57/58	5	3.30	3.70
50/51 - 56/57	6	2.90	3.60

V. CONCLUSION

Significant increases in chlorine usage by municipal wastewater treatment plants in the San Francisco Bay Area directly proceeded failure of the regional dungeness crab population.

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APPENDIX

APPENDIX

SURVEY FACILITY CASE HISTORIES

This section contains the chlorination histories reported by the wastewater treatment plant personnel contacted. This data, with interpolation where necessary, was used in computing the chlorination activity totals. East Bay Municipal Utilities District, Ora Loma Sanitary District and the three San Francisco plants were the only facilities able to provide complete records for the study period. These five dischargers; however, comprised approximately 45% of the total flow surveyed within the jurisdiction of the Region 2 Water Quality Board. It is probable that any errors of interpolation in individual cases largely cancel each other out, thus preserving the validity of the results.

APPENDIX

CITY OF SAN FRANCISCO

CONTACT: DR. A.E. Bagot

Chlorine Usage (lb/yr) x 10^{-6}

Season	Richmond Sunset Plant (1)	North Point Plant (2)	Southeast Plant (3)
75-76	0.90	3.10	2.43
74-75	0.99	2.32	2.43
73-74	1.02	2.90	2.64
72-73	0.89	2.26	3.03
71-72	0.86	2.16	3.81
70-71	0.85	2.09	4.14
69-70	0.77	2.29	2.82
68-69	0.64	2.13	1.62
67-68	0.61	2.26	1.60
66-67	0.61	2.17	1.19
65-66	0.54	1.77	--
64-65	0.54	1.74	--
63-64	0.54	1.68	--
62-63	0.47	1.56	0.08
61-62	0.52	1.48	0.15
60-61	0.40	1.48	0.07
59-60	0.52	1.31	0.06
58-59	0.61	1.28	0.18
57-58	0.55	1.20	0.05
56-57	0.52	1.21	0.13
55-56	0.48	1.23	0.17
54-55	0.45	1.09	0.40
53-54	0.41	1.16	0.44
52-53	0.45	1.54	0.14
51-52		1.00	
50-51		0.98	
49-50		0.95	

CITY OF SAN MATEO (4)

CONTACT: Mr. Al Esch

Began dry weather chlorination: 1953-54.

Began year-round chlorination in 1964-65 at one ton/day for 8 mgd.

Present: 2,500 lb/day for 10.5 mgd.

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CITY OF PALO ALTO (5)

CONTACT: Mr. Chuck Williamson

Began chlorination in mid-1950's at same dosage as present

Flow mid-1950's through 1970: 12-13 mgd.

Applied 1,800 lb/day, 1971 to present for 28 mgd.

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CITY OF SUNNYVALE (6)

CONTACT: Ms. Helen Farnham

1971 through 1974-75: 800 lb/day for dry weather flows of 15.5 mgd and 1,200 lb/day for wet weather flows.

1975-76: 400 lb/day for dry weather flows of 15.5 mgd and 700 lb/day for wet weather flows.

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SAN JOSE — SANTA CLARA SANITARY DISTRICT (7)

CONTACT: Mr. Baker

1955-56 began prechlorination at 4-5 ppm for 30 mgd

1969-70 began postchlorination for total dosage of 15 ppm for 75-80 mgd

Present: 15 ppm for 90 mgd.

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CITY OF HAYWARD (8)

CONTACT: Mr. Fred Cebalt

1970-71: 1,000 lb/day dry weather flows and 2,000 lb/day wet weather flows

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ORA LOMA SANITARY DISTRICT (9)

CONTACT: Mr. Larry Freitas

Season	Usage (lb/day)
75-76	1000
74-75	1170
73-74	1000
72-73	1200
71-72	1150
70-71	1500
69-70	1500
68-69	1500

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EAST BAY MUNICIPAL UTILITIES DISTRICT (10)

CONTACT: Mr. Cariss

Season	Usage (lb/yr) x 10^{-3}
75-76	6935
74-75	6360
73-74	6120
72-73	6382
71-72	6902
70-71	7560
69-70	7679
68-69	5706
67-68	5225
66-67	3119
65-66	2046
64-65	1548
63-64	1826
62-63	1526
61-62	1300
60-61	1382
59-60	1546
58-59	1176
57-58	1042
56-57	682
55-56	540
54-55	796
53-54	568

CITY OF RICHMOND (11)

CONTACT: Mr. Rick Alexander

1958-59 through 1967-68: 1300 lb/day

1968-69 through present: 650 lb/day

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SAN PABLO SANITARY DISTRICT (12)

CONTACT: Mr. Buzz Vandershoot

1966-67 to present: 14 ppm for 7 mgd flow.

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TOWN OF PINOLE (13)

CONTACT: Mr. Dave Pinoto

1970-71 to present: 100 lb/day

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CENTRAL CONTRA COSTA SANITARY DISTRICT (14)

CONTACT: Mr. Packwood

1956-57: 250 lb/day for prechlorination

1969-70: postchlorination began, 4000 lb/day total.

1970-71: 3600 lb/day

1974-75: 3161 lb/day

1975-76: 3170 lb/day

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VALLEJO SANITATION AND FLOOD CONTROL DISTRICT (15)

CONTACT: Mr. Nealy

1964-76: 230 T/yr

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NAPA SANITATION DISTRICT (16)

CONTACT: Mr. Bernie Erskine

1970-71: began chlorinating

1972-73: 525 lb/day for dry weather flows and 3180 lb/day for wet weather flows.

1975-76: 90 lb/day

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SONOMA VALLEY COUNTY SANITATION DISTRICT (17)

CONTACT: Mr. Bennett and Mr. Wayne Goodrich

1953-54: began chlorinating

1960 to present: 12 ppm for 2 mgd

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CITY OF PETALUMA (18)

CONTACT: Mr. Fred Schoeneweis

1955-56: 300 lb/day

1975-76: 400 lb/day

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SANITARY DISTRICT NO. 6 OF MARIN COUNTY, IGNACIO (19)

CONTACT: Mr. Marvin Miller

1965: 80 lb/day

1969: 150 lb/day

Present: 6.6 ppm for 1.1 mgd flow

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SANITARY DISTRICT NO. 6 OF MARIN COUNTY, NOVATO (20)

CONTACT: Mr. Marvin Miller

1965: 175 lb/day

1969: 250 lb/day

Present: 10.2 ppm for 2.5 mgd flow.

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LAS GALLINAS VALLEY SANITARY DISTRICT, SAN RAFAEL (21)

CONTACT: Mr. Walter Sadler

1970: 125 lb/day

1976: 250 lb/day

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SAN RAFAEL SANITARY DISTRICT (22)

CONTACT: Mr. Ross Tidwell

1964: began chlorination of 1 mgd

1972-75: 127,000 lb/yr for 3 mgd flow

SANITARY DISTRICT NO. 1 OF MARIN COUNTY (23)

CONTACT: Mr. John Anderson

1956: Began chlorination
1966: 5 T/month
1976: 6.5 T/month

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CITY OF MILL VALLEY (24)

CONTACT: Mr. Ralph Dilcher

1951: began chlorination of primary effluent at 500 lb/day
1959: began chlorination of secondary effluent at 30 lb/day
1965: to present – 200 lb/day

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SAUSALITO – MARIN CITY SANITARY DISTRICT (25)

CONTACT: Mr. Davener

1956: 200 lb/day
1976: 300 lb/day

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CITY OF SACRAMENTO

CONTACT: Mr. Tom Ikesaki

1954-59: 51 T/yr

1960-70: 61 T/yr

1971 to present — 60 T/yr April through July, 275 T/yr August through October, 52 T/yr November through March.

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CITY OF STOCKTON

CONTACT: Mr. Lin Norton

Season	Chlorine Usage (T/yr)
75-76	838
74-75	480
73-74	731
72-73	541
71-72	284
70-71	252
69-70	464
68-69	429
67-68	630
66-67	584

Plant began operation in 1928.

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