Geochemistry of Tributyltin in Coastal Waters: An Experiment in a Merl Mesocosm

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GEOCHEMISTRY OF TRIBUTYL Tin
IN COASTAL WATERS:
AN EXPERIMENT IN A MERL MESOCOSM

BY
DAVID ADELMAN

A THESIS SUBMITTED IN PARTIAL FULFILLMENT
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Abstract

Tributyltin is effective at controlling fouling organisms on vessels' hulls when used as an additive in marine paints. Its effectiveness is due to its extreme toxicity to fouling organisms after it is released from the paint. Unfortunately, it is also toxic to some nontarget organisms at concentrations approaching one part per trillion. After leaching from the hull of a vessel, the rates of degradation and transport from the water column have important implications for estuarine ecosystems, but the processes involved in these removal mechanisms are poorly understood. Using radiolabeled tributyltin, in a 13 m³ mesocosm, the processes of degradation and transport were monitored. The total removal rate was found to be 6 to 20% per day. The primary mechanisms consisted of: biodegradation (40%); transport to the sediment (35%); and transport to the atmosphere (25%). Biodegradation proceeded primarily through the process of debutylation to dibutyltin, which in turn degraded to monobutyltin. In addition to this degradation sequence, the data suggested that a degradation pathway from tributyltin directly to monobutyltin may have been important. Transport to the sediment was accelerated by adsorption to suspended particles. The distribution coefficients between suspended particles and water (in units of \([\mu g/kg]/[\mu g/L]\)) were found to be; \((60 \pm 30)(10)^3\) for tributyltin; \((30 \pm 20)(10)^3\) for dibutyltin; and \((2.9 \pm 0.5)(10)^3\) for monobutyltin. Tributyltin also accumulated in the surface layer of the water column, implying that transport to the atmosphere may have been an important removal process. A concentration as high as of \(1.7 \pm 0.3 \mu g/L\) was found. This value was 3 times the concentration measured in the underlying water column.
Acknowledgement

Special thanks to Dr. K.R. Hinga, whose day to day guidance was invaluable in the design, execution and compilation of this research. I also wish to thank Dr. M.E.Q. Pilson, my major professor, for his support, encouragement and guidance in the execution and compilation of this work. I wish to thank professors J.G. Quinn and S.M. Barnett for their contributions to this research as members of my thesis committee. This work was supported by funding from the U.S. Environmental Protection Agency, under Cooperative Agreement CR-814287-02 and earlier contracts.
Concern for tributyltin (TBT) in the environment results from its extreme toxicity to many marine organisms. Although the validity of data used to justify restrictions on the use of TBT in marine paints has been questioned (Salazar and Champ, 1988), it is generally felt that restrictions are warranted. Problems with the data include abnormally high degrees of uncertainty concerning toxic concentrations and a lack of explanations for cause and effects. One rather large gap in the knowledge concerning TBT in the marine environment is a lack of information on the rates of degradation and transport from the water column. Several studies have been undertaken to quantify the individual process rates. This thesis was undertaken to determine if the findings for the individual process are valid when occurring simultaneously in the marine environment.

The Marine Ecosystems Research Laboratory (MERL) at the University of Rhode Island is well suited for such an investigation. The mesocosms at MERL simulate the naturally varying environment in Narragansett Bay, and are a good predictive tool in the study of both organic and metallic pollutants. However, the largest benefit gained through the use of MERL was not the facilities but the people who work there.

In particular the experience of Senior Marine Scientist Dr. K.R. Hinga was invaluable. Through years of experience with radio-labeled compounds, the design, validation and testing protocols of this study were expedited. Without such experience, this investigation could easily have taken an extra year to complete; because of the environmental significance of TBT, and pending restrictions on its use, a year's delay was considered undesirable. For similar reasons, a preliminary report of the early results from this experiment was presented at, and published in the proceedings of, Oceans 87: the "International Organotin Symposium" (Hinga, Adelman and Pilson, 1987).

This thesis presents the results of the study and one interpretation of the data. Other interpretations could, no doubt, fit the results. Further research designed to investigate the
validity of this interpretation is needed. In particular, a wide range of organotins should be tested, preferably using gas chromatography with flame photometric detection. Hopefully, the restriction of TBT's use in marine paints will not result in a lack of research funds for the needed studies.

Two notes on the presentation of this thesis are called for. The first point concerns the introduction to this study. It is longer than is usual for a masters thesis and covers areas of information not directly related to the research itself. It presents a broad overview of the current knowledge (and opinions) concerning TBT. However, it is by no means an exhaustive review of TBT's chemistry and uses. The information was included to give a broader view of TBT's significance.

It should also be noted that this thesis is primarily concerned with the first 50 days of the 274 day experiment. Data from the full experiment were used in this thesis, when necessary, but they have not been fully analyzed. It was included (as Appendix-F) primarily to provide access for future use.
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I. Introduction

A. History

The French oyster farmers of Arcachon Bay became concerned with the health of their Pacific oysters (*Crassostrea gigas*) in 1976. Since their introduction in 1968, the oysters had been growing well, but now the oyster farmers saw anomalies consisting of shell thickening and slow growth rates. The following year there were no spat falls in many areas of the bay. The occurrence of these problems correlated with proximity to boat moorings and marinas. Alzieu et al. (1980) suggested that the problems might be caused by the recent increased use of the highly effective antifouling paints containing tributyltin (TBT). Analysis of the affected oysters showed a significant elevation in tin concentration. However, no environmental sampling for TBT itself had been performed, so the speculation was unproven (Champ and Lowenstein, 1987).

Attempts to introduce *C. gigas* to England in 1968 had failed from the start. The problem of shell thickening had originally been attributed to high concentrations of fine particles in the water. The speculation of Alzieu and co-workers prompted Waldock and Thain (1983) to investigate the problem further. They found that low concentrations of TBT could cause shell thickening in *C. gigas* and that the areas where their introduction had failed were heavily contaminated with TBT. Shortly thereafter it was found that low concentrations of TBT caused high mortality rates for larvae of the common mussel *Mytilus edulis*, (Beaumont and Budd, 1984). These findings confirmed earlier speculations that organotin compounds could be devastating to certain marine organisms (Zuckerman et al., 1978).

Subsequent research by Alzieu et al. (1982) confirmed the link between TBT and the devastation caused to the French oyster industry. The French government moved quickly to regulate the use of marine paints containing TBT. On January 19, 1982, before the study was published, the French government banned the use of organotin compounds in antifouling paints on vessels under 25 meters in length (Alzieu et al., 1986). The British government proposed restrictions in Feb. 1985. The Paint Makers Association of Britain, which mounted strong opposition, was successful in delaying legislation to ban paints using TBT on the grounds that
the case against TBT was unproven. Instead, water quality standards were instituted. Scientific evidence continued to mount. The use of water quality criteria was found to be ineffective. A ban on the retail sale of antifouling paints containing TBT was finally passed by Parliament in April 1987 (Side, 1987).

In the United States, several states including Virginia, Maryland, Oregon and Washington have restricted the use of paints containing TBT (Champ and Pugh, 1987; Wolniakowski et al., 1987). In addition to legislation passed or pending in several states, the Federal government is also working to restrict the use of TBT paints nation wide. The Environmental Protection Agency initiated a Special Review of all paints containing TBT in January 1986 (50 FR 778). Because of procedural requirements, all the data are not due into the EPA until 1990 (EPA, 1987). Because of this delay in action, a bill entitled "Organotin Antifouling Paint Control Act of 1987" was introduced in the U.S. House of Representatives as H.R. 2210. The bill will restrict the use of TBT on boats smaller than 25 m and set limits on allowable release rates of less than 5.0 µg cm⁻² d⁻¹. Regulating release rates is important because vessels over 25 m in length will still be permitted to use paints containing TBT. The bill is pending as of September 1988 (H.R. 100-400, 1987).

B. Uses of Organotins

Organometaloids are a class of compound characterized by a carbon to metal bond. Although most metals are capable of forming organometaloids, the commercially important compounds are formed from mercury, tin and lead. They are used extensively as pesticides (organomercury and organotin), gasoline additives (methyl and ethyllead) as well as catalysts and stabilizers for plastics (organotin). World wide production of all organometaloids in 1986 was several hundred thousand tonnes of which organotins represented 35,000 tonnes (7% of all tin consumption). Triorganotin compounds used as biocides totaled 8,000 tonnes, of which tributyltin for use in antifouling paints represented 3,000 tonnes.

Various organotin compounds have been in commercial use since the mid 1920's. Their first use was as a mothproofing agent. In the 1930's they were found to be a good stabilizer of chlorinated benzenes and diphenyls used in electrical transformers and
capacitors. They were also found to be useful as a stabilizer of polyvinyl chloride (PVC). Long exposure of PVC to heat or ultraviolet light causes it to discolor or become cloudy. When clear plastic is required, tributyltin is used as a stabilizer to prevent these problems. It is used extensively in the plastic packaging for food and consumer products.

The extreme biocidal properties of triorganotins were discovered in the 1950's. Since that time, many fungicidal applications have been found in agriculture and industry. Major uses of triorganotins include the preservation of wood used for exterior siding and decking as well as fungicidal applications in the home. They are also used extensively in the textile industry to prevent the growth of odor-causing fungi. Tributyltin was first used as a molluscicide in the 1960's. It was found to be an effective control agent for those freshwater snails which act as the intermediate hosts of the worms causing the disease Schistosomiasis in humans. The use of TBT as an antifouling agent in paint soon followed (Champ, 1986).

C. Antifouling Paints

The nature of TBT incorporation into paint matrixes has undergone refinements since its first introduction. The early formulations used TBT in free association. Later improvements include the development of ablative paints and the binding of TBT into the matrix of the paint as a copolymer. In the early free association paints, TBT was simply blended into standard paint formulations. The paint matrix was permeable to water which slowly percolated through the paint leaching out the TBT. After about two years the paint surface became clogged, preventing the further leaching of TBT. The hull of the craft then required scraping before new paint could be applied. This system of protection had two disadvantages. First, high concentrations of TBT were required to maintain adequate release rates as the surface of the paint clogged. This caused unnecessarily high release rates during the early life of the paint. The second disadvantage of this system was that even after the effective life of the paint, the scrapings had high concentrations of TBT. This caused problems with safe disposal of the scrapings.
These problems were reduced with the introduction of free association ablative, also called self polishing, formulations. As with the earlier formulations, the TBT was simply blended into the paint. Rather than controlling the release rate by leaching through a permeable matrix, however, it was controlled by the rate at which the impermeable outer surface of the paint was worn away. Only freshly exposed surfaces release TBT. A more constant release rate was thereby achieved. This improvement allowed lower concentrations of TBT to be used. It also reduced hull preparation when repainting.

Copolymer ablative paints are the most recent form to be developed. In this system the TBT is chemically bonded to the polymer matrix of the paint. The polymer matrix is slightly acidic and slowly breaks down when exposed to slightly alkaline environments, such as seawater. The release rate of TBT is much lower than that for free association or self polishing paints. These paints have an active life of 5 to 7 years. As with free association ablative paints, surface preparation for repainting is minimal (Anderson et al., 1986).

Antifouling paints containing TBT are more effective and longer lasting than the major alternative, which is copper-based paint. TBT's use is economically advantageous for two reasons. First, because they are more effective than copper-based paints, they will keep a vessel's hull cleaner. A cleaner, smoother hull produces less drag, thereby reducing fuel consumption. A vessel with 5% of its hull fouled consumes 5-10% more fuel. A vessel with 33% of its hull fouled can consume up to 50% more fuel. For large commercial ship's the fuel bill can constitute as much as 50% of operating costs. Therefore, a cleaner hull can translate into large savings for commercial vessels (Ludgate, 1987).

The second advantage of TBT copolymer paint is the longer effective life. In a statistical study involving 16,000 vessels covering the years 1977 to 1987, International Paint (USA) Inc. found that after 24 months, the hulls of 99% of ships using TBT in the copolymer form were less than 10% fouled, compared to only 45% of the ships using copper based paints. The study also found that after 5 years 90% of ships using copolymer TBT paints had less than 10% hull fouling (Schatzberg, 1987). Because of the longer effective life of the paint, time between repainting, which
involves dry docking, can be extended (Ludgate, 1987). However, the advantage gained from the extended service life is not often fully realized in practice. Most commercial ships are dry-docked every two years for inspection and maintenance regardless of the need for repainting. The cost of repainting is often, therefore, the marginal cost of painting and not the added expense of dry-docking, contrary to the claims of International Paint Inc. (EPA, 1987).

In a U.S. Navy study of 43 cargo ships, total overall fuel savings were estimated to be 16%. This would represent a savings of $110 million/yr if repainting with TBT copolymers were implemented fleet wide, based on a 1982 fuel cost of $1.45/gal. (Schatzberg, 1987). Using a more conservative fuel price of $0.80/gal. the savings would be more like $60 million. The U.S. Navy study found that copper-base antifouling paints have an effective life of 14 months in most waters and can be as short as 7 months in tropical waters. The study demonstrated that TBT antifouling paints provided fouling-free hull protection for a full 5 to 7 years. The advantages to the U.S. Navy are as much strategic as fuel efficiency. For the Navy, benefits include increased cruising speed, increased range and reduced running noise (important for sonar avoidance).

For small recreational vessels, which account for 33% of TBT paint used, the economic advantages are greatly reduced. Of the vessels which use TBT paint, 79% are repainted within 2 years. No advantage is gained from extended dry docking intervals. Because TBT copolymer paint costs $160-180/gal., as compared to $100/gal for copper-based paint, repainting is more expensive. In addition, copper-based paint and TBT paint give compairable protection over a two year time interval. Therefore, little advantage is realized from TBT paint use on small boats (EPA, 1987).

D. Environmental Considerations

The effectiveness of TBT antifouling paint is an indication of its toxicity to marine organisms. Unfortunately, because all antifouling paints leach TBT into the water, their effects are not limited to the target organisms which cause hull fouling. A summary of research compiled by Cardwell and Sheldon (1986) lists the toxicity of TBT in over 40 tests of nontarget organisms.
Concentrations causing acute toxicity, expressed as 96-h LC$_{50}$, included: 40 µg/L for the adult shrimp Crangon crangon; 15µg/L for juvenile Sheepshead minnow Cyprinodon variegatus; 5 µg/L for Lobster larvae Homarus americanus; and 0.33 µg/L for the diatom Skeletonema costatum. The most sensitive organisms reported were the larvae of the hard shell clam Mercenaria mercenaria, with a 96-h LC$_{50}$ of 0.006 µg/L.

Chronic effects of TBT are more difficult to quantify. They generally consist of slow growth rates and moribund behavior. Concentrations reported to cause chronic effects include; 1.0 µg/L for the Lobster Homarus americanus; 0.2 µg/L for the Rainbow trout Salmo gairdneri; and 0.05 µg/L for spat of the Pacific Oyster Crassostrea gigas. In addition, reproductive failure in the Dogwelk snail Nucella lapillus was projected to occur at a concentration of 0.002 µg/L (summarized by Rexrode, 1987). It should be noted that while TBT is extremely toxic to many forms of marine life, its toxicity to mammals is quite low. The LD$_{50}$ is reported to be 100-200 mg/kg (Neumann, 1967).

Although TBT is toxic to many forms of marine life at ultralow concentrations, its occurrence in the marine environment is limited. Concentrations of TBT are highest in areas were large numbers of recreational boats are moored. These boats are typically moored in small harbors or marinas for extended periods of time and are used only intermittently. TBT leaching from the many hulls is thereby concentrated in a small area. In addition, harbors and marinas are often sheltered waters and poorly flushed, which allows the concentration of TBT to build up. TBT causes problems where boats are kept near areas of important marine populations. Major problems can also result because of the timing of boating activities. For example, spring is when freshly painted boats are often returned to the water, and also a time of major fish spawnings (Stang et al., 1986; Seligman et al., 1987; Thain et al., 1987).

Environmental concentrations of TBT as high as 2.3 µg/L have been reported for lakes in Canada (Maguire et al., 1986a). In the United States, the highest concentration measured was in Port Annapolis, Maryland, at 1.0 µg/L (Hall et. al., 1987). A comprehensive study of 95 locations in California likely to be
affected found that: 10 locations had no detectable concentrations of TBT; 73 locations had concentrations of less than 0.2 µg/L; 10 locations had concentrations between 0.2 and 0.4 µg/L; and two locations had concentrations between 0.4 and 0.6 µg/L (Stallard et al., 1987). A baseline assessment of butyltin concentrations in selected U.S. harbor systems conducted by the Naval Ocean Systems Center included 12 locations in Narragansett Bay sampled on a single day. Nine locations had no detectable concentrations of TBT while three locations had concentrations ranging from 0.009 to 0.13 µg/L. The highest of these values was found at Castle Hill Coast Guard Station, Newport (Grovhoug et al., 1987).

E. Detection Methods

Determination of tributyltin in environmental samples is difficult because of the low concentrations involved. In order to determine the concentration of TBT it is necessary to extract, concentrate, separate and detect the butyltins. The butyltin molecule is made up of an organic and a metallic component; for this reason, separation and detection can utilize either characteristic. Methods relying on detection of the tin atom include Atomic Absorption (AA) spectrometry (Hodge et al., 1979; De Doncker et al., 1986; Valkirs et al., 1986a) or tin specific detectors using Gas Chromatography (GC) (Maguire and Huneault, 1981; Matthias et al., 1986; Unger et al., 1986). Attempts have also been made to use neutron activation analysis and spectrophotometric determination, but interferences and the very low concentrations of TBT in environmental samples make these methods unpractical (Bowen, 1972; Omar and Bowen, 1982). Detection of TBT can also take advantage of the organic component of the molecule by using an organic sensitive detector with GC separation (Dooley, 1986; Junk and Richard, 1987).

Before either GC or AA can be used, it is necessary to convert the butyltins into volatile components so that TBT can be separated from other organotins. When using AA detection a conversion is usually carried out in the water sample itself. The water sample is placed in a modified gas washing bottle, acidified, and reacted with NaBH₄. The resulting butyltin hydrides, such as Bu₃SnH, Bu₂SnH₂ and BuSnH₃, are purged from the sample with an inert gas flow and trapped in a glass U-tube which is immersed in liquid
nitrogen. The trap is then removed from the nitrogen bath and slowly warmed. The organotin species are released sequentially according to their boiling points as they distill from the trap. The gas stream carries the butyltin hydrides to a modified quartz burner in the AA for detection (e.g. Valkirs et al., 1986a).

The same basic chemical reaction can be used when employing GC separation. Rather than purging the butyltin hydrides from the water sample with a gas stream, however, they are often extracted using an organic solvent. Following extraction with the solvent and separation with GC, the butyltin hydrides can be detected in one of two ways. A flame ionization detector can be used to detect carbon bonds or a flame photometric detector can be used to detect the presence of tin (IV) species (e.g. Dooley, 1986; Matthias et al., 1986).

A different method of extraction and reaction to volatile compounds has also been used to prepare for GC separation of components. A Grignard reagent can be used to convert the butyltins into any desired alkyltin for analysis. The butyltins must first be extracted from the water sample using an organic solvent because Grignard reagents react with water. After extraction, which is generally reported to be 90-95% efficient for TBT, the ionic butyltins are converted to purely covalent species using an appropriate Grignard reagent. It has been found that pentylbutyltins, formed from the Grignard pentylmagnesium bromide, are volatile enough to be separated and detected using GC but are not so volatile as to be lost when the organic solvent is vacuum evaporated to concentrate the extractant (e.g. Maguire and Huneault, 1981).

Analyses of natural water samples using either analytical method are not without problems. It has been observed that diesel fuel (0.04%) in seawater can inhibit butyltin conversion to volatile tin hydrides (Valkirs et al., 1986a). Therefore, the use of hydridization techniques in areas of hydrocarbon pollution (often areas of interest) may result in apparent concentrations of TBT which are lower than the actual concentrations. High concentrations of sulfides have been found to interfere with analysis by forming butylsulfultins (Valkirs et al., 1985). It has also been found that butyltins on particles cannot be fully reacted to hydride forms without first extracting with an organic solvent (Valkirs et al., 1987a).
Analysis using hydride generation with extraction of products into methylene chloride may also be subject to error. Increasing the sample volume by 8-10 fold was found to result in a 5-6 fold decrease of detection limits (Matthias et al., 1986). Possible explanations given for this include larger surface area of the separatory funnel (which is unlikely) or possible problems with shaking larger funnels with an automatic mechanical shaker. Problems have also been reported with flame photometric detectors in general. It has been reported that flame photometric detectors can be poisoned by injections of more than 100 ng of TBT. Recovery from this condition is slow (Maguire and Huneault, 1981).

An inter-laboratory comparison of two hydride generation methods indicated that problems can be encountered with some samples for unknown reasons (Valkirs et al., 1987b). Samples from 14 locations were analyzed by 3 laboratories. Neither method gave consistently higher concentrations or higher relative standard deviations. In most cases, the concentrations for the two methods were within 20% of each other. Three samples, however, indicated problems with the analysis. In two samples, the method using extraction with methylene chloride gave concentrations of only half that found using purge and detection using AA. In addition, the relative standard deviation of these two samples was over 25% as compared to only 15% for the entire set of samples. In another sample, the method using simultaneous hydridization/extraction gave a value over 4 times that of the method using gas purging. Both methods had a relative standard deviation of almost 40% for this sample. No obvious anomalies (oil pollution, excessive particulates, algal blooms, etc.) were noted in these samples.

The analysis of samples using extraction with an organic solvent followed by pentylation was found to give more consistent results. Recoveries of butyltins have been reported to be in the range of 96 ± 8% (Maguire and Huneault, 1981). These results were reported for fresh water samples spiked with 10 ppm of butyltins, which are several orders of magnitude higher than environmental concentrations. However, in a study of speciation and octanol-water partitioning it was found that TBT forms chemical species with all major seawater anions, and that anion concentration can significantly influence the partition coefficient (Laughlin et al., 1986). Results relying on extraction of butyltins from seawater at
ultralow concentrations, 3 to 5 orders of magnitude lower than those used by Maguire and Huneault, could well be affected.

Several techniques have been investigated for the extraction of butyltin from sediment. Investigations have used natural sediments, with unknown concentrations of TBT, or samples spiked with known quantities of TBT. Refluxing spiked samples for 2 h with a complexing agent-solvent system consisting of tropolone in benzene, resulted in the following recoveries; TBT = 63-108%, DBT = 97-180%, and MBT = 55-103% (Maguire, 1984). A method yielding more consistent results consists of mixing the sample with precipitated silica and sodium sulfate (drying agents), followed by 48 h of soxlet extraction using hexane (Rice et al., 1987). Recovery for samples spiked with TBT were reported to be 86-102%. The extraction of 5 subsamples of a homogenized 'natural' sediment resulted in a coefficient of variation (Sd./Mean) of 5%.

An international interlaboratory comparison of methods found that extraction of TBT from dried sediment using an acidified methanol reflux resulted in the lowest coefficient of variation (0.7%). The highest recoveries used an acidified hexane reflux. However, the coefficient of variation using this method was over 30%. A summary of results from this study (Stephenson et al., 1987) is presented in Table No. 1.

Table No. 1. Comparison of methods for extraction of TBT from sediment. From Stephenson et al. (1987).

<table>
<thead>
<tr>
<th>Extraction Chemicals</th>
<th>No. of Replicates</th>
<th>Concentration PPB</th>
<th>Coefficient of Variation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. 5% HCl - Methanol</td>
<td>3</td>
<td>262 ± 2</td>
<td>0.7%</td>
</tr>
<tr>
<td>2. 12 N HCl - Hexane</td>
<td>5</td>
<td>527 ± 167</td>
<td>31.7%</td>
</tr>
<tr>
<td>3. 6 N HCl - Meth. Chloride</td>
<td>3</td>
<td>434 ± 59</td>
<td>13.6%</td>
</tr>
<tr>
<td>4. Hexane</td>
<td>3</td>
<td>220 ± 29</td>
<td>13.2%</td>
</tr>
</tbody>
</table>

**F. Geochemistry**

Organotin compounds are molecules with the form $R_nSnX_{4-n}$, where $R$ can be any organic radical such as methyl, ethyl, butyl, etc, or combinations of such radicals. The X represents any anion such as $Cl^-$ or $OH^-$, and $n$ can be any number between 1 and 4. In addition, aquatic organotins of the form $[R_nSn(OH_2)_m]^{(4-n)+}$ can form and are highly solvated ($n+m = 5$ or 6) and nonvolatile. They also show
strong lipophilic properties (Zingaro, 1979). In seawater, TBT speciation products have been identified using NMR spectrometry of chloroform extracts. It was found that, in an equilibrium mixture, the distribution of species was pH sensitive and was composed primarily of TBTCl, TBTOH, TBTOH$_2^+$ and tributyltin carbonato species. In the environment TBT is primarily in the chloride form (60%) but the distribution is a function of chlorinaty, alkalinity and pH (Laughlin et al., 1986).

Most metals can be methylated in the environment, both chemically and biologically. Methylation is the only proven environmental reaction and occurs only to metals in their highest oxidation state. Mono- through fully saturated methylmetallic compounds have been found in the environment.

When released into the environment, organometallic compounds usually degrade quickly; having half-lifes measured in days or weeks rather than years. If released to the atmosphere, during production or manufacturing, short wavelength radiation in the atmosphere can break the carbon-metal bond. If released to soil, as pesticides or disposed of with refuse in land fills, they are immobilized by the soil and degraded before being transported to aquatic environments. In fact, only aquatic environments are adversely affected by organometaloids to any large extent and then only when they are introduced directly (Craig, 1986).

Degradation of organotins in the environment is thought to proceed by the successive loss of butyl groups. Tetrabutyltin degrades sequentially to tributyltin, dibutyltin, monobutyltin and eventually to ionic Sn (IV). The reaction series is represented:

\[ \text{Sn} \xrightarrow{K_1} \text{R}_3\text{SnX} \xrightarrow{K_2} \text{R}_2\text{SnX} \xrightarrow{K_3} \text{RSnX}_3 \xrightarrow{K_4} \text{SnX}_4 \]

The reaction rates are thought to have the order: \( K_1 > K_2 > K_3 \sim K_4 \) (Zuckerman et al., 1978).

Biodegradation is thought to be important in TBT degradation. Two organisms have been identified as being capable of degrading TBT: Pseudomonas strain 224 (Jackson et al., 1982); and an axenic culture of Skeletonema costatum (Lee et al., 1987). However, preliminary, unpublished data by Lee et al. indicated slow or no degradation of TBT by cultures of dinoflagellates or chrysophytes.
In addition to the major degradation products, intermediates have been identified. Seligman et al. (1986a), using water from San Diego Bay, and Lee et al. (1987), using water from the Skidway river (Georgia) reported that dibutyltin (DBT) was found in environmental samples spiked with radiolabeled TBT. There was evidence that (α-hydroxybutyl)dibutyltin formed, which in turn degraded to dibutyltin. Because CO₂ production did not correlate with DBT production, Lee et al. speculated that butene was also formed, and lost to the atmosphere. This hypothesis was based on earlier work by Kimmell et al. (1977), who found that extracts of rat liver could degrade TBT with the production of (α-hydroxybutyl)dibutyltin in conjunction with butene and butenol.

Olson and Brinkman (1986) reported that laboratory incubation of natural water samples from Chesapeake Bay had resulted in biological synthesis of tetrabutyltin (TTBT) from added TBT. TTBT occurrence was sporadic, usually occurring in only one of the duplicate samples. Its presence was confirmed by gas chromatography-mass spectrometry. Its production was thought to have possibly resulted from a biological redistribution reaction of TBT. In addition, two unidentified products were also found; one had a GC retention time between tri- and dibutyltin and a second had a retention time between di- and monobutyltin (MBT).

Biomethylation of butyltins can take place in the environment, resulting in low concentrations of tributylmethyltin and dibutyldimethyltin (Maguire and Tkacz, 1985; Maguire, 1984). Biomethylation of tin has been demonstrated in laboratory studies (Hallas et al., 1982; Guard and Cobet, 1981). Studies have also found tetra-, tri-, di-, and mono- species of methyl stannanes and methyltins as well as tetramethyltin in the environment (Jackson et al., 1982; Tugrul et al., 1983; Matthias et al., 1986; Weber et al., 1986).

Several laboratory studies using environmental samples have determined degradation rates for TBT. Maguire and Tkacz (1985), using water and sediment from Toronto Harbor, reported a photochemical degradation half-life for TBT of over 3 months and a biological degradation half-life of about 4 months. Olson and Brinkman (1986), using water from Chesapeake Bay, reported a degradation half-life of 6 days in water collected in September, while water collected in February showed no signs of degradation.
Thain et al. (1987) reported the half-life of TBT to be 6 days in fresh water and an extrapolated half-life in salt water of 60 days for TBT and 90 days for DBT. Seligman et al. (1986a), using biocidal concentrations of TBT (744 µg/L), estimated the degradation half-life to be 145 days. Using environmental concentrations (0.5 to 2.0 µg/L) resulted in half-lives of 6 to 9 days for water collected in July. The combined results of all these studies indicate that degradation of TBT is a function of photosynthetic activity in the water because degradation was faster when phytoplankton concentrations were high. However, none of these studies reported chlorophyll concentrations. Lee et al. (1987), using water from the Elizabeth and James Rivers (Norfolk, Va.), reported that the degradation rate was dependent on the concentration of chlorophyll in the water. Half-lives of 4 and 9 days were found for water containing 12 and 3 µg/L of chlorophyll.

While degradation kinetics are clearly important, TBT's variation with time and distribution in the water column are also important. Two temporal variations have been identified. In the short term, the concentration of TBT has been found to be highest at low tide and decreases as incoming uncontaminated water dilutes the existing water mass (Seligman et al., 1986B). The concentration of TBT has also been found to have a yearly cycle. High concentrations are found in the spring, when freshly painted boats are put in the water. The concentration is lowest in winter after boats have been removed (Huggett et al., 1986; Seligman et al., 1986b; Hall et al., 1987). This finding is of importance to the ecology of the marine environment because the spring maximum in concentration of TBT coincides with the maximum for the organisms which spend only their juvenile life in planktonic form; collectively known as the meroplankton.

The distribution of TBT in the water column is important because of its implications regarding transport mechanisms. In addition to TBT dissolved in bulk water, the concentration in other phases is of interest. Such phases are the surface microlayer, particles in the water column and the sediment. Butyltin concentrations in the surface microlayer (SML) were found to be higher than in the water column (WC) at over 30 locations throughout southern England. The concentration ratio, SML/WC, was found to be as high as 27 at one fresh water site, but averaged 9 ± 10 for
samples taken at fresh and salt water locations throughout the year, using screen and glass plate samplers (Cleary and Stebbing, 1987). This was expected because of the surface active nature of the molecule (Gucinski, 1986). Interestingly, the study by Cleary and Stebbing found that the concentration ratio was higher in open waters then in sheltered areas. The SML/WC for TBT at fresh water sites in Canada have been reported to be as high as 6,000. In this study, TBT was occasionally found in the SML without being detected in the underlying water column (Maguire, 1986b). Not all investigators, however, have found consistently elevated concentrations in the SML. In a study of butyltins in Chesapeake Bay the monthly monitoring of 8 locations found no trend in concentration ratios. Low or nondetectable concentrations of TBT in the SML were often accompanied by high concentrations in the underlying water column (Hall et al., 1987). In this study, using a Teflon sheet sampler, the mean enrichment factor was 4 ± 9. The concentration of TBT in the SML was as high as 1.17 µg/L but averaged 0.54-0.31 µg/L.

The variability of reported concentrations in the SML may be the result of several factors. The most apparent reason is the variability of wind and wave action. These forces work to mix the SML down into the water column. A less apparent reason for the variability is the difference in sampling equipment used. It has been found, for example, that using a glass plate can give concentrations that are up to 10 times higher than those obtained by using a screen sampler. Part of the difficulty in comparing the results is caused by uncertainty in the SML thickness. The various sampling techniques also contribute to the problem because it is not known how much subsurface water is collected with the SML. The dilution factor is therefore often unknown (Gucinski, 1986; Cleary and Stebbing, 1987).

The elevated concentration of TBT in the SML is significant for two reasons. Biologically, it could result in high mortality of the plankton community collectively known as the neuston. The bacterial population of the neuston can have a population density 100 to 1,000 times higher than the water just a few centimeters below the SML. These bacteria provide food for relatively high concentrations of protozoa, including tintinnids, radiolarians and foraminiferans. Some large zooplankton, including certain species of copepods and decapod crustaceans, are also members of the
neuston. The floating eggs of some species of fish, which reside temporarily in the SML, could also be affected.

Geochemically, the SML is an area of unique chemistry and energy forms. The SML has high concentrations of many organic molecules such as long-chain fatty acids, alcohols and esters (Maguire, 1986a). Most of the organic compounds in the SML have only been given operational classifications. It is also an area of high O$_2$ concentration and fluctuating salinity as well as being the only area of the ocean subject to high fluxes of infrared and ultraviolet radiation. This raises the possibility of photochemical reactions taking place. Investigations of the SML indicate that it is a temporary sink for organotins in the water column (Gucinski, 1986). Maguire et al. (1983) found that TBT dissolved in fresh water neither volatilized nor lost butyl groups over a period of at least 2 months in the dark at 20 °C, even though 20% of the water had evaporated.

Another mechanism for removal of TBT from the water column is adsorption to particles and transport to the sediment. The distribution of TBT between water and suspended particles can be expressed as the distribution coefficient K$_d$, in units of (µg/kg)/(µg/L). The percent of TBT in the water column found on suspended particles has been reported to vary from 11 to 17%, with corresponding K$_d$'s of (1.5-1.8)(10$^3$) in January and (8.0-8.5)(10$^3$) in July (Valkirs et al., 1986b). Another study by Valkirs, et al. (1987a) found higher fractions on particles, ranging from 13 to 29% giving K$_d$'s of (4.6-39)(10$^3$). Harris and Cleary (1987) reported K$_d$'s of (0.9-2.2)(10$^3$) for marine particles.

Because of the relatively high concentrations of TBT found on particles, transport to the sediment is expected to be of importance (Lee et al., 1987). Accumulation of butyltins in the sediment has been investigated in several environmental and laboratory studies. Concentrations of TBT as high as 0.55 µg/g of sediment have been reported for a marina in San Diego Bay, Calif. (Stang and Seligman, 1986). This value is far higher than any other reported and thought to have possibly been caused by paint chips in the sediment. In a comprehensive study of over 70 sediment samples throughout California, the highest reported
concentration was 0.22 µg/g of sediment (Stallard et al., 1987).
In another environmental study carried out in Pearl Harbor, Hawaii,
using a recirculating dome system enclosing approximately 0.8 m² of
sediment and 100 L of water, Kd values and in situ adsorption and
desorption rates were measured for TBT and DBT. Values of Kd were
found to vary from (6.2-55)(10)³ for TBT and (2.1-26)(10)³ for DBT.
Fluxes of the butyltins, expressed in units of ng cm⁻² d⁻¹, were
found to be: TBT adsorption = 0.57, desorption = 0.00; DBT
adsorption = 0.55, desorption = 0.16; MBT results were statistic­
ally ambiguous (Stang and Seligman, 1987).
A factor found to be of major importance to sorption behavior
was the type of sediment studied. In laboratory sorption experi­
ments conducted by Dooley and Homer (1983), sediment samples were
suspended in seawater and spiked with butyltins, the sorption rate
and equilibrium concentrations on particles were found to decrease
in the order: Silty Clay > Bentonite > Silty Sand > Sand. Equi­
librium concentrations of the three butyltin species, using sandy
sediment and 0.2 to 1.4 mg butyltin/L, were calculated to result in
the following amounts on particles; TBT = 98.0% (Kd = 480);
DBT = 93.8 (Kd = 148); MBT = 98.6% (Kd = 640). Desorption using
the equilibrated sediments and clean seawater were found to be
measurable but slight. However, this experiment used concentra­
tions of butyltin (0.2-1.4 mg/L) and sediment (10g/100g water)
which could not be considered representative of environmental
conditions. In comparison, Randell and Weber (1986) conducted a
series of experiments using iron oxide particles (10 to 1000 mg/L)
coated with fulvic acid in artificial seawater. They found the
following percents of butyltins on particles; TBT = 72-100%,
Kd = (1.5-4.2)(10)³; DBT = 0-56%, Kd = (0.0-1.3)(10)³; and
MBT = 57-95%, Kd = 1,000(10)³.
In a study of Chesapeake Bay sediments of different composi­
tion, the Kd of TBT was found to vary from 0.1(10)³ to 8.2(10)³
(Unger et al., 1987). One sediment type used in this study was
similar to that found in Narraganset Bay. The sediment, from
Carter Creek, consisted of 47.7% clay, 2.9% organic carbon and was
equilibrated with water having a salinity of 24 °/oo. The Kd was
found to be $1.3(10)^3$. In contrast to the study by Dooley and Homer (1983), however, all sediments in this study were found to have equal coefficients of adsorption and desorption. The influence of salinity on sorption was also addressed in the study by Unger et al. It was found that increasing salinity from 0 to 35 $°/oo$ doubled the $K_d$. In contrast, the study by Randell and Weber (1986) reported an inverse relationship between $K_d$ and salinity.

In addition to the importance of sediment sorption behavior, degradation of TBT in sediment is also of interest when considering its geochemistry. The measured half-life of TBT in sediment was 120 to 160 days (Maguire and Tkacz, 1985; Stang and Seligman, 1986). Degradation is primarily by sequential debutylation. However, Maguire (1984), using sediment from Ontario Harbor, found that methylation of butyltins had taken place. In this study, tributylmethyldtin was found at 4-8% of TBT concentrations while dibutylmethyldimethyltin was found at 19-124% of DBT concentrations.

In summary, the geochemistry of TBT in the marine environment is not straightforward and many studies contradict each other. TBT appears to break down primarily by biodegradation to DBT. Reports of the degradative half-life range from 4 days to 4 months in natural waters and longer in sediments. The major pathway out of the water column to the sediment is probably through adsorption to particles. The binding strength of adsorption is between weak and very strong. This wide variety of results makes TBT an interesting compound for study.

G. Marine Ecosystems Research Laboratory

This study was conducted at the Marine Ecosystems Research Laboratory (MERL) located at the Graduate School of Oceanography, University of Rhode Island. The facility consists of 14 land based experimental tanks and a building which houses 6 laboratories. Each fiberglass tank, called a mesocosm, is 1.8 m in diameter and 5.5 m deep. Each mesocosm may contain a 0.3 m deep sediment tray which is loaded with sediment collected intact with its existing biological community from one of several locations in adjacent Narraganset Bay (Hunt and Smith, 1983). Tidal mixing is simulated by mixing each tank with a vertical plunger for two hours, four times per day.
The mesocosms can be run in either of two modes of operation. Often, the tanks are run in a flow-through mode. This mode is useful in the study of biological effects of pollutants (Donaghay, 1984). Flow through operation consists of adding water to the mesocosm during mixing cycles so that the residence time for water in the mesocosms matches the residence time in the bay, about 30 days (Kremer and Nixon, 1978; Pilson, 1985). From analysis of six years of operating data, it has been shown that the tanks are representative of the bay for nutrient cycling and chlorophyll within the range of natural variation (Pilson, 1985).

The mesocosms can also be run in batch mode without water additions. In batch operation the mesocosms have been shown to reproduce the functioning of the bay for at least 6 months at a time (Pilson, 1985). Batch mode has been used to investigate the geochemistry of many trace metals and organic pollutants (Santschi, 1982; Donaghay, 1984).

A MERL mesocosm in batch mode is well suited to a study of the geochemistry of tributyltin. The use of an enclosed system with an actively coupled water column and benthos was required to trace the transport pathways, reservoirs and degradation of this compound. In this way a link could be made between laboratory experiments and field measurements summarized above. Indeed, TBT is an excellent compound for study in a MERL mesocosm because of the many independent studies which have been performed on it. Laboratory experiments and extensive field measurements are required to validate the findings of mesocosm experiments (Oviatt et al., 1984). Conversely, mesocosm experiments are useful in validating laboratory and field measurements.

H. Experimental Considerations

Detection methods and sample handling procedures were a primary consideration in the design of this study. Radiolabeled TBT was chosen for several reasons. Although extraction procedures for TBT in seawater have been found to be generally reproducible by other investigators, little was known of extraction and detection procedures for the breakdown products; DBT and MBT. The use of radiolabeled TBT would permit determination not only of extracted fractions, but also compounds not extracted by standard techniques.
This was thought to be of importance for particulate matter and sediment, where verification of extraction techniques is difficult. In addition, the use of $^{14}$C-TBT provides an independent check of degradation through the formation of detectable $^{14}$CO$_2$. 
II. Experimental

At 20:30 h, July 13, 1987, 480 µCi of Tri-n[1-14C]butyltin chloride in 10 ml of MeOH (21 mCi/mmol; Amersham Corporation, made available through the courtesy of D. Rudnick and U.S. EPA Gulf Breeze Laboratory) was added to a MERL mesocosm. It was introduced to the mesocosm by submerging the 20 ml scintillation vial in which it was contained to a depth of about 0.5 m and removing the cap. Daily samples were taken at 08:30 h, during a mixing cycle. The mesocosm was spiked in the evening to allow 3 sets of samples to be taken before sunrise the following day. This avoided any possibility of the initial measurements being affected by photochemical reactions.

A. Procedures

The processing of samples for the detection of radiolabeled TBT required numerous samples to be extracted, concentrated, separated, and the activity measured. Each of these steps, with the exception of measurement, may cause losses of TBT or other products, which would affect the apparent concentration of each butyltin species in the samples. The procedures for TBT were found to be reproducible, with a low standard deviation, prior to the start of the experiment. The losses in determination of di- and monobutyltin were not then known. Following is an outline of the processing of a sample for radiolabeled butyltins in the water column. Modifications to this procedure will be discussed under each of the other four factors monitored. These included total activity, total particulate activity, particulate extractible activity and radiolabeled carbon dioxide.

1. Extraction

Whole seawater samples of 500 ml were acidified in a separatory funnel to a pH of about 2 with 2 ml of 6 N HCl. Hexane (15 ml) was added. The funnel was capped and vigorously shaken at 15 min intervals for one hour. After settling, the aqueous phase was removed to a 500-ml Erlenmeyer flask. The hexane phase was transferred to a 50-ml pear-shaped flask, and the water sample returned to the separatory funnel. The Erlenmeyer flask was rinsed with 9 ml of hexane which was transferred to the separatory funnel and
used for the second extraction. After the second extraction, the hexane was removed and combined with the first extract. The separatory funnel was rinsed with 5 ml of additional hexane which was added to the hexane phase.

2. Concentration

The combined hexane extract was concentrated using vacuum evaporation. A centrifugal pump was used to lower the pressure to 7.5 cm of Hg while the sample was immersed in a water bath maintained at 25 °C. The volume was reduced from about 29 ml to about 0.1 ml. In some cases the final volume was reached by blowing a gentle stream of N₂ gas over the hexane phase at one atmosphere pressure.

3. Component Separation

Following concentration of the extracted phase, the components were separated using thin layer chromatography (TLC). In this procedure the differences in polarity of the components were utilized to effect the separation. The system consists of two phases; 1) a stationary phase of silica gel impregnated glass fiber filter paper (Gelman ITLC media) and; 2) a mobile phase of organic solvent. For the separation of butyltins, a mobile phase consisting primarily of isopropyl ether worked well. While TBT was mobilized at the solvent front, and MBT was not mobilized with any solvent, a 1% acetic acid addition placed the DBT half way between these positions. Adding less acid left the DBT closer to the origin, while more acid moved it closer to the solvent front. A central location was desirable to minimize the overlap of components.

Each 20 x 20 cm sheet of TLC paper was marked with a pencil so that 3 extracted samples could be separated at one time. The sheet was marked with an origin 1-2 cm from the lower edge of the paper and a location for the final solvent front 10 cm above the origin. This field was then marked along the origin to produce six areas for individual separations. In addition to separating 3 samples, three positions on the sheet were used for separation of a nonradiolabeled mixture of tri-, di- and monobutyltin (Aldrich Chemical Co.). These nonlabeled separations were used to locate the areas occupied by the butyltins in the samples.
The samples were transferred from the pear flask to the paper using the capillary action of Pasteur pipets. A nonradiolabeled standard mixture of TBT, DBT and MBT was placed on the origin at both sides of the TLC paper and at one of the central locations along the origin. (The extreme outside areas of the paper were not used for samples because the paper edges often had nonlinear solvent flow). The samples were then placed at the origin in intermediate positions. The paper was placed vertically in a vapor saturated TLC chamber with its lower edge (below the origin) immersed in a 100 ml reservoir of the solvent. When the solvent front reached the premarked location the separation was stopped by removing the paper from the TLC chamber.

After separation, the TLC paper was allowed to dry for several minutes. The paper was cut into 6 strips perpendicular to the origin. Nonlabeled standards of the butyltins were visualized using sprays containing pyrocatechol violet in ethanol and dithizone in chloroform (both from Kodak Chemical Co.), as described by Kimmell et al. (1977). The corresponding sections of the paper containing the extracted samples were cut out, placed in separate 7-ml scintillation vials and counted.

4. Counting

The determination of activity was performed by liquid scintillation counting (LSC) with a Beckman Model LS-3801. Samples were placed in 7 or 20 ml glass scintillation vials, depending on the size of sample. The vials were then filled with liquid scintillation 'cocktail' (Aquasol II, New England Nuclear). Samples were counted for one hour or until the error in counting represented by two standard deviations was less than one percent of the activity, which ever came first.

B. Processing Efficiencies

In order to quantify losses or fractionation (caused by the differences in chemical/physical properties of the three butyltins), the extraction and separation procedures were carried out using samples with known concentrations of TBT, DBT and MBT. Recovery efficiencies were then calculated for each component. Radiolabeled tri-, di- and monobutyltin were recovered from a clean-up of the source material used for this study. The TBT, as
received from Gulf Breeze Lab, was found to be 93% pure. The purity was increased to 97% using preparative thin layer chromatography. The impurities in the TBT solution ran at the same location in TLC separation as DBT and MBT standards in a variety of solvent systems. Therefore, a by-product of this purification was the recovery of a quantity of radiolabeled DBT and MBT. They were recovered from the TLC paper by extracting the appropriate areas with methylene chloride and used as reference solutions. The DBT solution (designated DBT reference solution No. 1) was later purified using vacuum evaporation followed by preparative TLC. The appropriate area was extracted with methanol and designated DBT reference solution No. 2. The MBT solution was found to be too impure for use (17 ± 3%). Efforts to purify it were unsuccessful because of MBT's volatility.

Aliquots of each reference solution were analyzed using TLC. Over the course of 185 days, reference solution DBT No. 1 was analyzed 5 times. No trend in change of compositions was apparent. The compositions of the reference solutions are reported in Table No. 2.

Table No. 2. Composition of the radiolabeled reference solutions used in verification of processing efficiencies.

<table>
<thead>
<tr>
<th>Reference Solution</th>
<th>No. of Replicates</th>
<th>Component</th>
<th>Concentration</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>TBT</td>
<td>4</td>
<td>TBT</td>
<td>97.1 ± 0.2</td>
<td>2.8 ± 0.2</td>
</tr>
<tr>
<td>DBT No. 1.</td>
<td>5</td>
<td>DBT</td>
<td>40 ± 5</td>
<td>60 ± 5</td>
</tr>
<tr>
<td>DBT No. 2.</td>
<td>3</td>
<td>MBT</td>
<td>14 ± 2</td>
<td>79 ± 1</td>
</tr>
</tbody>
</table>

With three solutions of known composition (maximum standard deviation of 5%) it should be possible to perform extraction and concentration procedures, then to calculate the recovery efficiency for all three butyltin compounds using simultaneous equations. However, it was found that the low concentration of MBT in each solution, combined with the rapid propagation of uncertainty when solving three equations, resulting in an uncertainty of over 100% for MBT. This problem will be addressed below.

Solving of the efficiency equations for TBT and DBT was simplified by considering the concentration of MBT in the TBT and DBT No. 1 solutions to be zero. This was a reasonable assumption because the concentration of MBT in these two solutions was within
the margin of error of these two compounds. When reference
solution DBT No. 2 was used, the behavior of MBT was determined
independently. A nonradiolabeled solution of MBT was processed and
concentrations determined using Atomic Absorption (AA)
spectrometry. The simultaneous equations used were in the form:

\[
\begin{align*}
\text{Solution} & \quad \text{Equation} \\
\text{TBT} & \quad X = (0.97 \pm 0.00) \text{TBT} + (0.03 \pm 0.00) \text{DBT} \\
\text{DBT} - 1 & \quad Y = (0.40 \pm 0.05) \text{TBT} + (0.60 \pm 0.05) \text{DBT} \\
\text{DBT} - 2 & \quad Z = (0.14 \pm 0.02) \text{TBT} + (0.79 \pm 0.01) \text{DBT} + (0.07 \pm 0.02) \text{MBT}
\end{align*}
\]

In this form the values of X, Y and Z represent the total activity
of each solution after processing. The equations can then be
solved for the values TBT and DBT, which represent the fraction of
each compound recovered.

1. Extraction Efficiency

The determination of the extraction efficiency for TBT required
the use of two reference solutions; TBT and DBT No. 1. A quantity
of the TBT reference solution was added to 10 g of hexane (15 ml).
Two aliquots of 0.2 g were weighed and counted. The remainder was
taken through the extraction procedure as outlined above. After
the second extraction and rinsing of the separatory funnel the
hexane phases were combined and weighed. Three 1.5 g aliquots were
weighed and counted. Three 10-g aliquots of the aqueous phase were
also weighed and counted. This procedure was carried out in trip­
llicate. The mean value of fraction of activity in the organic phase
(0.94 ± 0.02 from Table No. C-3) was used in the above equations as
the value "X". The procedure was repeated using DBT reference
solution No. 1, giving a value for "Y" of 0.70 ± 0.05 (from Table
No. C-4). The equations were then solved for TBT and DBT in the
organic phase as follows:

For TBT:

\[
\begin{align*}
(0.94 \pm 0.02) & = (0.97 \pm 0.00) \text{TBT} + (0.03 \pm 0.00) \text{DBT} \\
(0.70 \pm 0.05) & = (0.40 \pm 0.05) \text{TBT} + (0.60 \pm 0.05) \text{DBT} \\
(18.1 \pm 0.4) & = (19.0 \pm 0.1) \text{TBT}
\end{align*}
\]

\[
\text{TBT} = 0.95 \pm 0.03
\]
For DBT:

\[
(0.94 \pm 0.02) = (0.97 \pm 0.00)\text{TBT} + (0.03 \pm 0.00)\text{DBT}
\]
\[
(0.70 \pm 0.05) = (0.40 \pm 0.05)\text{TBT} + (0.60 \pm 0.05)\text{DBT}
\]
\[
-(0.76 \pm 0.12) = -(1.43 \pm 0.12)\text{DBT}
\]

\[
\text{DBT} = 0.53 \pm 0.18
\]

Using the fraction of activity transferred to the water yielded nonextractable values of \(0.5 \pm 0.5\)% for TBT and \(50 \pm 10\)% for DBT. In one set of replicates all glassware was rinsed 3 times with 2 ml of MeOH. It was found that 0.3% of the TBT and 1.2% of the DBT were on the glass.

Because of the large standard deviation for DBT, a quantity of reference solution No. 2 (butyltins in MeOH) was added to seawater and extracted as outlined above in extraction procedures. Because MBT represented 7 \(\pm\) 2% of that solution, solving the equation for DBT No. 2 required the determination of extraction efficiency for MBT.

Using non-radiolabeled MBT and analysis using Atomic Absorption spectrometry (AA) the quantity retained in the seawater was determined. The MBT (Aldrich Chemical Co.) was used without further purification. Concentrations higher than those found in experimental samples were used because of the high detection limit involved. Three different concentrations were used for replicates to insure that results were not concentration dependent. The following procedure was carried out using solutions containing 275; 575; and 960 mg MBT/L. The MBT was weighed and dissolved in 2 liters of seawater. Three 500 ml aliquots were weighed, acidified with 2 ml of 6 N HCl and placed in 500-ml separatory funnels. Hexane (15 ml) was added to two of the separatory funnels. The third funnel was used as a blank. After the second 9 ml hexane extraction, 100 ml of the aqueous phase was removed from each funnel. Three 10-g aliquots from each were taken and analyzed by AA using external standards.

Standards were remade for each day’s analysis because of the possibility of MBT loss to glassware. The standards were made by dissolving 300 mg of MBT in 1.00 L of deionized water. Dilution of this solution to a range of concentrations was used to make a standard curve. The use of MBT in deionized water as a standard for MBT in seawater was validated by analyzing samples of MBT in
deionized water and seawater using tin chloride for the standard curve. The concentration of MBT in the organic phase could not be measured because of its low solubility in hexane and high AA detection limits for tin. The results from the analysis of MBT in the aqueous phase indicate that a maximum of $11 \pm 3\%$ was extracted using hexane. This value was then used in the equation for DBT reference solution No. 2 to redetermine the distribution of DBT between the organic and aqueous phases. The results for extraction efficiencies are presented in Table No. 3.

Table No. 3. Component distributions calculated for TBT, DBT and MBT resulting from extraction of acidified seawater using hexane. Analysis by liquid scintillation counting and atomic absorption spectrometry. Values in percent.

<table>
<thead>
<tr>
<th>Reference Solution</th>
<th>Analysis</th>
<th>Organic Efficiency</th>
<th>Aqueous Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>TBT</td>
<td>LSC Back Extraction</td>
<td>95 $\pm$ 3</td>
<td>0.5 $\pm$ 0.5</td>
</tr>
<tr>
<td>DBT No. 1</td>
<td>LSC Back Extraction</td>
<td>50 $\pm$ 20</td>
<td>50 $\pm$ 10</td>
</tr>
<tr>
<td>DBT No. 2</td>
<td>LSC Extraction</td>
<td>60 $\pm$ 35</td>
<td>45 $\pm$ 35</td>
</tr>
<tr>
<td>MBT</td>
<td>AA Extraction</td>
<td>-</td>
<td>89 $\pm$ 3</td>
</tr>
</tbody>
</table>

2. Concentration Efficiency

The loss of butyltin during vacuum evaporation of the hexane phase was determined using the reference solutions designated TBT and DBT No. 1. A quantity of each reference solution was added to hexane in a 50-ml pear-shaped flask and weighed. Three 0.5 g aliquots of each were weighed and counted. The remainder was vacuum evaporated at a pressure of 7.5 cm Hg while being maintained at a temperature of 25°C. The resulting 0.1 ml sample was transferred to a scintillation vial and counted. This procedure was carried out 3 times. The fractions of recoverable tri- and dibutyltin were determined by solving the simultaneous equations.

In addition to this analysis, the quantity of activity adhering to the glassware was determined for each replicate. For 2 of the replicates the pear shaped flask was rinsed 3 times with 1 ml of hexane. The hexane from these washes was transferred using the same pipet used to transfer the sample. The three rinses were combined in a scintillation vial and counted. The rinsing procedure was repeated 3 more times for a total of 12 rinses for each of the flasks. For the third replicate, 3 washes of 1 ml MeOH
was used. Evaporative losses were calculated as that activity not accounted for in the hexane sample or washed from the glass. The results are presented in Table No. 4.

Table No. 4. Calculated distribution of components following vacuum evaporation of hexane spiked with reference solutions. Values in percent.

<table>
<thead>
<tr>
<th>Component</th>
<th>Recovered</th>
<th>On Glass</th>
<th>Evaporated</th>
</tr>
</thead>
<tbody>
<tr>
<td>TBT</td>
<td>88 ± 6</td>
<td>10 ± 4</td>
<td>2 ± 7</td>
</tr>
<tr>
<td>DBT</td>
<td>33 ± 3</td>
<td>20 ± 20</td>
<td>50 ± 20</td>
</tr>
</tbody>
</table>

The quantity of butyltin measured on glass, and the resulting quantity calculated by difference as evaporative losses, were functions of the solvent used to recover the compound adhering to the glass. In two replicates, hexane was used to recover components on glass. The third replicate used methanol for recovery from glass. This did not affect the values for TBT which is soluble in both solvents. The change in solvent did affect the values for DBT which, evidently, was more soluble in MeOH. This resulted in the standard deviation of DBT on glass being calculated from two low values and one high value. A high degree of variation resulted (20 ± 20)%. However, analytical results had a coefficient of uncertainty of 10% rather than 100% because only the percent recovered in vacuum evaporation was used in calculating sample composition.

The recovery of MBT from vacuum evaporation was not determined. The boiling point of MBT is 93 °C at 10 mm Hg, as compared to 143 °C for DBT. With a recovery for DBT of only 33 ± 3%, combined with an extraction efficiency for MBT of only 11%, the recovery of MBT would be negligible.

3. Counting Efficiency

All results of liquid scintillation counting were corrected for counting efficiency and natural background radiation using appropriate sample types, volumes and blanks. A discussion of LSC theory and corrections used in this research are presented in Appendix-F.
C. Factors Monitored

In order to determine the distribution and composition of radiolabeled compounds in the water column, a series of seven representative samples was taken at each sampling time. The samples were initially taken every 6 hours. After two days the schedule was reduced to daily. After two weeks samples were taken every other day. After one month samples were taken twice per week. Sediment samples were taken just before weekly tank cleaning.

1. Total Activity

The most important factor in determining a mass balance in a radiolabeled multicomponent system is to know the total amount of activity present at any time. Total activity was determined using 3 samples by pipeting 10 ml samples directly from the tank into 20 ml scintillation vials. Each vial was precharged with 4 drops of phenethylamine (PEA) and weighed. The PEA was added, starting on day 8, to complex the CO₂. It was used to prevent the loss of any radiolabeled CO₂ which had formed. The only processing required on returning to the lab was reweighing the vials and addition of scintillation cocktail.

2. Extractable Activity

Solvent extraction was used to determine the activity attributable to organotin compounds in unfiltered seawater. The sample for this procedure was taken in a preweighed 500-ml polyethylene bottle. The processing consisted of extraction, concentration and separation, as described earlier. After extraction the combined hexane phases were weighed. The total activity of the hexane phase was determined by weighing and counting two 1.5 g aliquotes. The remainder of the hexane phase was concentrated by rotary evaporation. Thin layer chromatography was used to separate the components.

In addition to the analysis of the hexane phase, a sample of the water phase after extraction was counted. A 100 ml portion of the aqueous phase was degassed of any remaining CO₂ by mixing with a magnetic stirrer in an open 500-ml beaker for 5 min. A 10 ml aliquot was weighed and counted.
3. Total Particulate Activity

A measurement of the total activity on suspended particles was required for the calculation of partition coefficients of the components. There was little agreement in the literature as to the effectiveness of any extraction processes for suspended particulate matter or sediment. It was therefore anticipated that there might be processing losses and a nonextractable fraction remaining on the particles.

Total particulate activity was determined by vacuum filtration of a one liter sample of water through a Watman GF/C glass fiber filter. Two filter papers were used in series. The top filter paper was used for measuring the total activity. The second filter paper was used as a blank to correct for the fraction of activity attributable to adsorption of dissolved radiolabeled components by the fibers of the filter paper. After filtration of the sample each filter paper was placed in a 7 ml scintillation vial and counted.

4. Particulate Extractable

The components contributing to the activity on the particles were analyzed by vacuum filtering another one liter sample through two filter papers. The filter paper and blank were extracted with 15 ml of hexane for 24 h. A 1.5 g aliquot was weighed and counted. The remaining hexane was concentrated by rotary evaporation and separated into its components by TLC. Each fraction was then counted.

5. Carbon Dioxide

The concentration of radiolabeled CO₂ was monitored as an indication of the degradation of butyl-groups. Combining this with data on total extractable activity a mass balance could be calculated and compared with the total water column activity. Samples for determination of CO₂ activity were taken in a 500-ml glass sampling bomb. This was necessary to prevent the loss of CO₂ before processing. The sample was placed in a glass wash bottle, acidified with 2 ml of 6 N HCl to a pH of < 2. N₂ gas was then bubbled through the sample for one hour. The exiting gas stream was passed through an activated carbon filter to remove any entrained butyltin. It was bubbled through 4.5 ml of the CO₂
complexing agent, phenethylamine (PEA), held in a 20-ml scintillation vial equipped with a stopper and glass stripping column. After collection of the CO₂ several drops of DI water were added to the PEA to help dissolve the complex in the scintillation cocktail. The stripping column was rinsed with 0.75 ml of PEA and 10 ml of scintillation cocktail which was added to the scintillation vial. The solvent was then counted.

6. Other Particulate Samples

Two other samples were processed in order to characterize the particulate matter in the water column. These samples were not taken for determination of radioactivity, but for correlation of environmental conditions in the mesocosm.

a. Total Particulate Weight

Total particulate weight was measured to determine the distribution coefficient of butyltins between seawater and suspended particles and to correlate experimental results to estuarine conditions. A 500 ml sample of water was collected in a polyethylene bottle. It was weighed and vacuum filtered through a pre-dried and weighed 0.45 µm Nuclepore filter. Following filtration the filter papers were stored in a freezer under vacuum conditions so as to preserve the material and dry the filter paper. The filter papers were then reweighed to determine total particulate weight. Each filter paper was weighed 3 times using a Cahn balance which has a precision of 1 µg.

b. Chlorophyll

The photosynthetic ability and general health of the phytoplankton was determined by fluorescence analysis. It was thought that part of the degradation of TBT in the water column could be carried out by phytoplankton. By the measurement of chlorophyll and phaeophytin, it should be possible to correlate the conditions found in the mesocosm with the open bay and possibly other environments of interest.

Chlorophyll was determined by filtration of two replicate 10 ml water samples through Watman GF/F glass fiber filters. A 50 ml sample was collected for this purpose in a 100-ml polyethylene bottle. One ml of saturated MgCO₃ was added as a wetting agent.
The 10 ml samples were pipeted to a syringe and pressure filtered. Samples were stored frozen in aluminum foil. Preparation for analysis consisted of grinding a filter paper in 5 ml of buffered 90% acetone. The tissue grinder consisted of a glass tube and a motor driven Teflon pestle. The resulting slurry was transferred to a 15 ml polystyrene centrifuge tube. Two rinses of buffered acetone were used to quantitatively transfer the sample. Total volume was brought up to 10.0 ml. The sample was capped and allowed to stand in the dark for 20 min before further processing. After centrifuging and decanting, the liquid was analyzed using a Turner Design's Model 10 Series Field Fluorometer. The sample was then acidified with two drops of 10% HCl and the combined phaeophytin and reacted chlorophyll determined (Kelly, 1986).

7. Surface Microlayer

The surface microlayer was sampled 8 times over the first 6 weeks of the experiment. The calculated thickness of water layer sampled was 440 ± 20 µm (Gearing and Gearing, 1982). A 40 X 40 cm stainless steel screen suspended in a frame was dipped under the surface of the water. It was raised in a horizontal orientation through the surface layer, allowed to drain off excess water, and was then tilted. When the upper edge of the screen lost its water film, the remaining water was collected in a 100-ml beaker. This procedure was repeated three times. Two 10 ml samples were pipeted directly into 20 ml scintillation vials, weighed and counted.

8. Sediment

Sediment samples were taken throughout the course of the experiment. Four replicate samples were taken weekly prior to tank cleaning. Samples were collected in a 2.5 cm I.D. stainless steel core barrel which was attached to the end of an aluminum pole (Frithsen, 1986). The corer was lowered to the bottom. It was then pushed firmly into the sediment, with the intention of recovering the floc-like material at the interface and about 10 cm of sediment. To prevent loss of sample on retrieval of the core, a rubber stopper was placed in the lower end of the core barrel before it was removed from the water. The outside of the core barrel was washed free of any sediment, removed from the water and
a stopper placed in the top end. Samples were kept upright until frozen. They were then extruded from the core barrel, wrapped in aluminum foil and restored in a freezer until analyzed.

Several sediment extraction techniques were evaluated over the course of this experiment. Low extraction efficiencies and poor component separation in TLC resulted in a lack of useful data. In this study, the data used to represent sediment activity were therefore limited to the sum of extracted activity plus activity collected when the samples were combusted to determine extraction efficiencies. A discussion of extraction procedures can be found in Appendix-D.

D. Calculations

1. Activity Balance

Two types of activity balances were calculated in this study; a water column balance; and a mesocosm balance. In order to calculate an activity balance for the water column, it was necessary to add the measured quantity of $^{14}$CO$_2$ to the calculated butyltin components. This combined component activity was designated $\Sigma$ and compared to total measured activity in the water column. For calculation of the mesocosm activity balance, it was also necessary to account for activity loss resulting from the natural exchange of CO$_2$ with the atmosphere. For the mesocosm activity balance a loss rate for CO$_2$ of 1%/d was used (Hinga, 1984).

2. Component Concentrations

It was possible to calculate the concentration of TBT, DBT and MBT in the water column by setting up a system of equations based on; the efficiencies of extraction and vacuum evaporation; and on the fraction of TBT making up the extracted phase. It was assumed for these calculations that all radiolabeled organic species in the water were tri-, di- and monobutyltin. It was also assumed that all CO$_2$ was removed in sample processing. The equations used were:

$$T = (A - 0.11M)F/0.95$$
$$D = (A - 0.11M)(1 - F)/0.55$$
$$M = [R - (0.05T + 0.45D)]/0.89$$
In these equations the numbers represent extraction efficiencies and:

\[ T \text{ = Concentration of TBT in the water.} \]
\[ D \text{ = Concentration of DBT in the water.} \]
\[ M \text{ = Concentration of MBT in the water.} \]
\[ A \text{ = Activity extracted from the water.} \]
\[ R \text{ = Activity remaining in the aqueous phase after extraction.} \]
\[ F \text{ = Fraction of activity in the organic phase attributed to TBT} \]
\[ \text{after correction for losses in vacuum evaporation. It can be expressed as:} \]

\[
F = \frac{\text{TBT}/0.88}{\text{TBT}/0.88 + \text{DBT}/0.33}
\]

The calculated activity distributions resulting from the use of the simultaneous equations had several points which were obviously in error. The values for TBT (days 4.5 and 5.5) were considerably below the otherwise smooth trend. The values for MBT (days 0.1 - 5.5) indicated negative concentrations were present. These values were redetermined using graphical interpolation between the origin and the trend after day 6. Values for DBT were then recalculated by maintaining the same total activity as calculated by the simultaneous equations.

After determining the distribution of activity between TBT, DBT and MBT the concentrations were converted from an activity basis to a molar basis. The conversion consisted of two parts. For TBT the activity of the compound was known. Activity of TBT used to spike the mesocosm was 60 mCi/g. The molecular weight of TBT is 325 g/mol. The conversion factors: 1.0 Ci = 2.22(10)^{12} dpm; and 1.0 L of seawater equals 1.02 kg, resulted in a conversion factor for TBT of 1.0 dpm/g = 0.024 nmol/L. For conversion of DBT and MBT a correction for specific activity was required.

The need for activity correction factors resulted from the makeup of the radiolabeled TBT used in this experiment. It was labeled with one atom of \(^{14}\text{C}\) in one of the three butyl groups. A molecule of TBT is degraded to DBT by the removal of one butyl group. There is a 1/3 chance that the lost butyl group contains the \(^{14}\text{C}\) atom. There is a 2/3 chance that the radiolabeled butyl group is retained. This results in only 2/3 of the produced DBT being detected with liquid scintillation counting. Activity
resulting from DBT was multiplied by a factor of 1.5 to correct for the lost $^{14}$C. Similarly, there is only a $\frac{1}{3}$ chance that MBT retains the $^{14}$C atom. This required that the measured activity of MBT be multiplied by a factor of 3 to account for nondetected MBT. This resulted in conversion factors of: 1.0 dpm/g = 0.036 nmol/L for DBT; and 1.0 dpm/g = 0.072 nmol/L for MBT.
III. Results

Data resulting from this work are presented in tabular form in Appendix-A. Procedures for calculation of data are presented in Appendix-B.

A. Water Column Activity

The direct measurement of activity in the water column, which will be referred to as direct activity, was initially $77 \pm 5 \text{ dpm/g}$ over the first 7 h (5 samples). Activity extracted from the water over 7 h was $78 \pm 2 \text{ dpm/g}$. Although the two values were essentially equal, the latter value had a lower standard deviation and so was used as initial activity in the water column for computational purposes.

Both direct and extractable activities decreased and essentially stabilized by day 50. Extractable activity decreased more rapidly than direct activity, as can be seen from Fig. 1. As extractable activity decreased, nonextractable activity and CO$_2$ (not shown) increased.

![Figure 1. Activity measured in the water column directly as well as extractable and nonextractable components (but not including CO$_2$).](image-url)
Although not part of this thesis, direct activity for the full 278 days is presented in Fig. 2. Also presented in this figure is the sum of the components (extractable, nonextractable and CO₂) minus directly measured activity. Figure 2 is included in order to highlight a measurement problem not noticeable in the first 50 days data. As can be seen in this figure, measurements had a tendency to oscillate. The minimum values in direct activity correlated with maximum values of component minus direct activity as emphasized by the vertical lines. Because the oscillations were present in all radio-measurements, it was thought to have resulted from a problem in liquid scintillation counting.

Figure 2. Comparison of activity measured directly from the mesocosm to the sum of extracted, nonextracted and CO₂ minus direct activity.

The frequency of the oscillation was about 35 days. The amplitude of the oscillation varied with sample type. While it was about 11 dpm/g for direct activity it was about 4.5 dpm/g for non-extractable; 3.0 dpm/g for CO₂; and 0.5 dpm/g for the extractable fraction (Figs. E1-4). The lower line in Figure 2 represents the sum of the components minus direct activity, which should have equalled zero. They added up to zero at midcycle but to as much as 8.5 dpm/g at the cycle ends. The amplitudes, however, were
variable and the cause of the error was unknown. This, in combination with the variability of the amplitude, made correction of the raw data impractical. The fluctuations in extractable and nonextractable activities, the only values used for calculating butyltin concentrations, were small. Graphical corrections made to these concentrations were thought to be adequate to remove much of the uncertainty.

The activities attributed to TBT, DBT and MBT by the system of equations, and as corrected by graphical interpolation, are presented in Figs. 3-6 and Table No. A-3. The activity attributed to TBT is presented as a log plot in Fig. 3. The decrease of TBT in the water column over the course of the 49 day period could not be described by a single first order rate equation. However, it could be considered as first order by assuming two sequential first order processes with an initial rate constant of 20%/d (half-life of 3.5 d) which changed to 6.5%/d (half-life of 11 d) between days 15 and 20.

![Figure 3. Activity of TBT in the water column.](image)

The activity calculated for DBT formed a less consistent pattern than the values calculated for TBT in the first 10 days (Fig. 4). Activity ranged from a minimum of 5.2 dpm/g on day 0.5 to a maximum on day 5.5 of 36 dpm/g. For days 10-40 the activity decreased nearly linearly, from 10 to 1.2 dpm/g, at a rate of 0.29 dpm g⁻¹ d⁻¹. From the data corrected for negative concentra-
tions of MBT in Fig. 5, it can be seen that the concentration of DBT reached a maximum on day $10 \pm 2$ of $10.5 \pm 0.5$ dpm/g. Because the data from days 0.0-3.5 was inconsistent with data from days 4.5-7.5, it was not clear which of two possible paths was taken to reach the maximum concentration. Considering the consistency of the data from day 10 through day 40 it was felt that the data after day 4, and the lower dotted line in Fig. 5, was more representative of the full data set. The reason for change in trend at day 4 was not known.

![Figure 4](image1.png)

Figure 4. Activity of DBT in the water column before corrections.

![Figure 5](image2.png)

Figure 5. Activity of DBT in the water after corrections were made to MBT concentrations.
The calculated values for MBT in the first 6 days ranged from a minimum of -12.5 to a maximum of -1.4 dpm/g (Fig. 6). The reason for these negative concentrations was not known. On day 7.5 the activity jumped from negative values to 7.6 dpm/g. The trend after this point was linear. In order to correct for the negative concentrations, the line from day 10 to 40 was extended back to the origin to obtain concentrations of MBT for the first 6 days (dotted line).

Figure 6. Activity of MBT in the water as calculated.

On day 0.8, when the first analysis for radiolabeled CO₂ was made, the water column contained 0.1 dpm/g of water. Production reaching a value of 19.2 dpm/g by day 41.5, which included a loss of 1%/d to the atmosphere. The three low values on days 7.5-9.5 were the result of sample processing problems. The value of 11 dpm/g on day 4.5 was also suspected of being erroneous (Fig. 7).
Component activity, the sum of TBT, DBT, MBT and CO₂ (designated $\Sigma$), accounted for between 70 and 100% of directly measured activity (Fig. 8). Over the course of 41.5 days the value $\Sigma$/Measured was $0.90 \pm 0.10$. This difference may have resulted from the measurement problem discussed earlier.
The concentration of tri-, di- and monobutyltin in units of nmol/L are presented in Figure 9. The initial concentration of TBT in the mesocosm averaged 1.86 ± 0.05 nmol/L over the first 7 h. The concentration decreased at a rate of 20%/d decreasing to a rate of 6.5%/d. The concentration of DBT increased from an initial value of 0.05 nmol/L to a maximum of 0.38 nmol/L on day 10. This represented an average rate of increase of 0.03 nmol L⁻¹ d⁻¹. The concentration then decreased linearly from days 10-40 at a rate of 0.012 nmol L⁻¹ d⁻¹. The concentration of MBT increased from zero at time zero to a concentration of 0.5 nmol/L on day 10. This represented an average rate of increase of 0.05 nmol L⁻¹ d⁻¹. The concentration of MBT then increased linearly from day 10 at a rate of 0.012 nmol L⁻¹ d⁻¹.

![Figure 9](image_url)

Figure 9. Concentrations of TBT, DBT and MBT in the water column. Values in nmol/L.
B. Mesocosm Activity Balance

Activity retained in the water column through day 50 was $31.4 \pm 0.5$ dpm/g, or 40% of activity added to the mesocosm. Activity in the sediment increased to 32% of activity added to the mesocosm by day 15 and remained at that level for the remaining 35 days (Fig. 10). It should be noted that the apparent increase of activity on day 49 (dotted line) resulted from one of 4 sediment samples which indicated over 95% of initial activity was then in the sediment. This value was known to be incorrect. A better estimate for day 49 could be made by disregarding the one high sample and only considering the 3 other samples taken on day 49. This resulted in $20 \pm 20$ dpm/g being in the sediment on that day, which was consistent with the findings on the other 5 sediment sampling days (plotted on day 50 for clarity).

![Figure 10](image)

Figure 10. Activity measured in the sediment.

As can be seen in Figure 11, total activity accounted for over 50 days was 70 to 75% of activity initially added to the mesocosm. In addition to activity retained in the water column and transported to the sediment, two other sinks were determined to have accounted for slight losses. Based on a rate of 1%/d, loss of $^{14}$CO$_2$ to the atmosphere was 3.2% of initial activity. When the mesocosm was drained on day 284 the walls accounted for 0.9% of initial activity.
Figure 11. Activity in the water column, sediment and lost to the atmosphere as CO₂. Values in percent of initial activity.

C. Activity on Particles

On day 0.1, total activity on particles was 3.42 dpm/g of water, which represented 4.6% of activity in the mesocosm (Fig. 12). On day 0.3, the value had decreased to 3.20 dpm/g (3.9% of activity in the water). Filtration problems on days 0.5 through 1.5 invalidated the results from those samples. After correcting the filtration problems, particulate activity on days 2.5 and 3.5 remained at 4.0% of activity in the water. The percent of activity on particles decreased thereafter, with fluctuations, reaching a minimum by day 45.5 of 0.2%. The fluctuations correlated with days of cleaning the mesocosm walls (days 7, 15, 22, 29, 36). Cleaning the mesocosm walls consisted of scrubbing with abrasive pads and allowing the removed material, which became temporarily suspended in the water, to sink to the sediment. In this way the compounds under study, as well as nutrients in the algae, were retained and recycled within the system.
The concentration of particles in the water column was 680 µg/L on day 1.5 (first day of measurement) and increased slightly through the course of the experiment at a rate of 30 µg L⁻¹ d⁻¹. Chlorophyll concentration was 16 µg/L on day 1.5 and decreased through the course of the experiment at the rate of 0.4 µg L⁻¹ d⁻¹ (Fig. 13).
Because the percent of activity on particles and chlorophyll concentrations were both decreased simultaneously, percent of activity on particles was plotted against chlorophyll concentration in order to determine there relationship (Fig. 14). A geometric mean regression indicated a relationship with an equation:

\[
(\text{Percent on Particles}) = 0.21(\mu g/L \text{ of Chlorophyll}) - 0.86
\]

Figure 14. Percent of activity on particles versus chlorophyll concentration in the water column.

The activity extracted from particles was initially 100% TBT. It decreased to less than 30% TBT by day 18.5 (Table A-8). These fractions were used in conjunction with concentrations of TBT and DBT in the water to calculate distribution coefficients, in units of [µg/kg]/[µg/L]. The \( K_d \) for TBT ranged from 12,200 to 87,700 with a mean value of \((60 \pm 30)(10)^3\). For DBT the values ranged from 4,000 to 40,400 with a mean of \((30 \pm 20)(10)^3\). The \( K_d \) for MBT was calculated from the data of days 40-52 (4 points) using total activity on particles and total activity in the water. They ranged from 2,500 to 3,600 with a mean of \((2.9 \pm 0.5)(10)^3\) (Tables A9-11).

D. Surface Layer Activity

Activity in the surface layer of the water was \(220 \pm 40 \text{ dpm/g} \) on day 0.8, or 3 times the activity in the water column at that time. The activity decreased to a value equal to that in the water column by day 10 and continued to decrease through day 28.5 when the concentration was 24 dpm/g or 75% of the activity found in the water column (Table A-12).
IV. Discussion

A. Water Column Activity

Results of this experiment indicated that the process of TBT degradation proceeded primarily by successive debutylation through DBT to MBT, as suggested by Zuckerman et al. (1982). Evidence for this reaction sequence came from qualitative changes in butyltin concentrations. The concentration of DBT and MBT increased as the concentration of TBT decreased. Then, after the concentration of TBT had dropped below 10% of its initial value, the concentration of DBT decreased at a rate equal to the increase in MBT concentration. However, this may not have been the only reaction sequence to have taken place. The data also suggested that some TBT may have degraded directly to MBT without formation of the intermediate compound - DBT. Evidence for this reaction pathway consisted of a rapid increase in the concentration of MBT and an imbalance in production of degradation products.

The decrease of TBT in the water column could be represented as first order by assuming two sequential first order processes with an initial rate constant of 20%/day (half-life = 3.5 days), and a rate change occurring between days 15 and 20 to 6.5%/day (half-life = 11 days). These results were comparable to biological degradation half-lives of: 6 days reported by Olson and Brinkman (1986); 9 days reported by Seligman et al. (1986a); and 4-9 days reported by Lee et al. (1987), who also reported a correlation of half-life to chlorophyll concentrations.

Using water from the Elizabeth and James Rivers (Norfolk, Virginia), Lee et al. found that incubated samples of water having chlorophyll concentrations of 12 and 3 µg/L were able to degrade TBT at rates of 17 and 8%/day respectively. The change in removal rate in the mesocosm, from 20 to 6.5%/day, corresponded to a change in chlorophyll concentrations from 15 ± 6 to 8 ± 4 µg/L. This indicated that the removal rate of TBT was, at least to some extent, a function of biological activity in the water, as was found by Lee et al.

The degradation rate of TBT can be calculated as a function of the production rates of its degradation products. The degradation rate would be equal to the production rate of DBT and that fraction of MBT not derived from DBT degradation. The concentrations of DBT
and MBT were poorly defined in the first week of the experiment even after corrections were made for negative concentrations of MBT. Analysis of the degradation process was also hampered by the unexplained oscillation in activity measurements. The first cycle of the oscillation (the first 35 days), was the time of most interest in this experiment. It was the only cycle in which the sum of the components added up to less than the activity measured directly from the water column. This may have indicated that another component, such as butene, was produced during this time, as suggested by Lee et al. (1987). Any such component, if present, would have been volatilized in sample processing. This would have resulted in the components summing to less than the direct measurement, as was the case. The imbalance in activity could also have resulted from the measurement fluctuations. However, the cause of the fluctuations was not known. Efforts to correct for its effect were unsuccessful because the validity of correcting values in the first cycle, in which component activities summed to less than the direct activity measurement was not known. A more reliable indication that a highly volatile degradation product other than CO₂ may have been formed came from analysis of CO₂ production.

The concentration of DBT reached a maximum of about 10 dpm/g on day 10. The concentration of detectable CO₂ resulting from DBT production would be 5 dpm/g. The concentration of MBT on day 10 was 7.5 dpm/g indicating that 15.0 dpm/g of CO₂ would be detected from MBT production. The measured concentration of CO₂ was about 7 dpm/g or one third of the calculated value. The extreme imbalance did not persist through the experiment, as will be discussed below.

By day 10 the concentration of TBT had decreased by over 90%. A maximum degradation rate for DBT after day 10 could be calculated by assuming production and transport of DBT to be negligible. Based on these assumptions, the maximum degradation rate of DBT was not more than 0.012 nmol L⁻¹ d⁻¹. This represented a half-life for DBT of approximately 16 days.

In contrast, Thain et al. (1987) found the half-life for DBT to be 90 days. This figure was found for natural seawater incubated at 5 °C which had a half-life for TBT of 60 days. The water in the mesocosm was 18-22 °C. The lower rates found by Thain et al. could have been caused by the low temperature or by a lack of biota in
the water. Although Thain's rates were lower than the rates found in the mesocosm, the degradation rates for TBT and DBT were found to be of the same order of magnitude. The data from Thain et al. also indicated that degradation of DBT may have been consistent with linear reaction kinetics. The results from the mesocosm experiment support these findings.

The production rate of MBT in the first 10 days was 0.05 nmol L\(^{-1}\) d\(^{-1}\), which was 4 to 5 times higher than the rate after day 10. This implied that there may have been a degradation pathway from TBT directly to MBT. Taken in conjunction with the imbalance of CO\(_2\) in the first 10 days, this reaction sequence may have liberated a volatile degradation product other than CO\(_2\). After day 10, the production rate of MBT was equal to the degradation rate of DBT. No indication of MBT removal from the water column was evident. The equal rates indicated that all DBT in the water column was degraded to MBT rather than being transported from the water column. This was supported by analysis of CO\(_2\) production.

Production of CO\(_2\) through day 41.5, assuming a loss to the atmosphere at the rate of 1%/d, was 19.2 dpm/g. Production of MBT over this time was 11.7 dpm/g indicating that 23.4 dpm/g of CO\(_2\) should have been detected from MBT production. Only 80% of expected CO\(_2\) was accounted for. If butene had been produced as an intermediate in the degradation of butyl groups to CO\(_2\) some loss to the atmosphere would have been expected. The unaccounted for CO\(_2\) was 4.2 dpm/g, or about 5% of activity added to the mesocosm. At any rate, the large deficit in measured CO\(_2\) on day 10 had been greatly reduced by day 40. The production of a volatile degradation product other than CO\(_2\) could have been responsible for these results.

B. Mesocosm Activity Balance

About 40% of the activity added to the mesocosm was retained in the water column. Activity not retained in the water column could have been transported to any of 3 places: the sediment; atmosphere; or the mesocosm walls. The measured accumulation of activity in the sediment indicated that transport to the sediment was a major
pathway. Because of analytical problems, only total activity in the sediment was determined. Total activity accumulation in the sediment represented about 30% of activity added to the mesocosm. Although this represented a major removal mechanism, transport to the sediment accounted for only half of activity removed from the water column.

Transport of activity to the atmosphere was thought to be a major removal process because combined activity accounted for in the water column and sediment over the course of 49 days was only $74 \pm 9\%$ of activity added to the mesocosm. The loss of activity was not gradual. At the end of 7 days, when the first sediment samples were taken, the total activity accounted for was only $70 \pm 20\%$. These figures indicated that a 25 to 30% loss of activity was confined to the first week of the experiment. The unaccounted for activity, which was presumed to have been lost to the atmosphere, was not unusual for studies involving the degradation of TBT. Typical mass balances reported in the literature include; 86-102% by Rice et al. (1987); 73% by Seligman et al. (1986a); 65-85% by Olson and Brinkman (1986); and 55 ± 26 to 180 ± 100% by Maguire and Tkacz (1985).

It has been reported that TBT dissolved in fresh water did not volatilized over a period of 2 months (Maquire et al., 1983). However, the samples for that experiment were kept in a closed cabinet, in the dark, which was only opened for removal of samples. It is unlikely that those conditions would be useful in providing data for the evaluation of a mesocosm experiment opened to the sun and air. Solar radiation, air turbulence and mesocosm mixing could greatly affect transport to the atmosphere. In addition, the loss may not have resulted from the volatilization of TBT. Biomethylation of tin, methytlins and butyltins have been reported in the literature (Matthias et al., 1986; Weber et al., 1986; Maguire and Tkacz, 1985; Maquire, 1984; Tugrul et al., 1983; Jackson et al., 1982). Biomethylation of TBT would have resulted in tributylmethyltin which would have been far more volatile than TBT itself.

The formation of tributylmethyltin, if produced, might also have been responsible for the calculation of negative concentrations of MBT in the first few days of the experiment. The equations for calculating the distribution of activity between TBT, DBT and MBT were based on the assumption that they were the only
butyltins present. Tributylmethyltin would have been partitioned into the organic phase. This would have resulted in concentrations of TBT and DBT being over estimated because they were predominantly extracted into the organic phase. In order for activity to balance, a reduction in MBT concentration would result. The presence of butene would have a similar affect.

It was therefore quite possible that tributylmethyltin and/or butene was formed. Based on a loss of 25% of activity to the atmosphere, 3.2% was in the form of CO₂ as calculated from a rate of 1%/d. From concentrations of MBT and CO₂, 5.4% may have been lost as butene. The remaining 16.4% could have been lost in the form of tributylmethyltin.

It was possible that some of the activity thought to have been lost to the atmosphere was actually adsorbed to the mesocosm walls. Evidence that the mesocosm walls acted as a sink for butyltins, at least over the course of several weeks, can be found from activity on suspended particles. Activity on particles, as a percent of activity in the water column, generally doubled on sampling days following wall cleaning. The first wall cleaning was on day 7. The activity on suspended particles increased from 0.83 dpm/g on day 6.5 to 1.52 dpm/g on day 7.5. This indicated that 0.69 dpm/g (0.9% of initial activity) was removed from the walls. However, no direct measurements were made until the mesocosm was drained. It was thought that, at most, several percent of the TBT may have been adsorbed onto the mesocosm walls in the first few days. The mesocosm walls could not have been a permanent sink for butyltins because when the mesocosm was drained, on day 284, the walls accounted for only 0.3% of activity added to the mesocosm. In contrast, sediment from day 189 showed that 35 ± 10% of activity added to the mesocosm was still in the sediment.

C. Particulate Concentrations

The activity on particles in the water column over the first 5 days was predominantly TBT. Over this time 4.1 ± 0.3% of activity was on suspended particles. This value was less than that reported from other investigations. Valkirs et al. reported environmental samples were found containing; 11-17% (1986); and 13-29% (1987a).
The low fraction of TBT found on particles in the mesocosm may have been due to the low concentration of particles in the water (≤ 2.6 mg/L). The distribution coefficient, which correct for particulate concentration, were at the high end of values reported for TBT and DBT. The distribution coefficients in the mesocosm were found to be \((60 \pm 30) \times 10^3\) for TBT and \((30 \pm 20) \times 10^3\) for DBT. Reported values for TBT were; \((4.6 \text{ to } 39) \times 10^3\) by Valkirs et al. (1987a) and \((6.5 \text{ to } 55) \times 10^3\) by Stang and Seligman (1987). For DBT Stang and Seligman reported a value in the range of \((2.1 \text{ to } 26) \times 10^3\). These values were in reasonable agreement with the mesocosm results. On the other hand, the \(K_d\) of MBT was \((2.9 \pm 0.5) \times 10^3\) in the mesocosm as compared to a value of \(1,000 \times 10^3\) reported by Randell and Weber (1986), using iron oxide particles and artificial seawater.

The distribution coefficient corrects for the affect of particulate concentration but it was found to be a function of particle type (Dooley and Homer, 1983). While the total concentration of particulate matter in the water column remained relatively constant, the chlorophyll concentration decreased by an order of magnitude. The correlation of percent of activity on particles to chlorophyll concentration indicated that the \(K_d\) may have been particle specific. The change in chlorophyll concentration and a shift in butyltin composition from TBT to MBT occurred simultaneously. The effect of these simultaneous changes on the determination of the distribution coefficients was unknown.

However, the dependance of \(K_d\) on particle type may account for the large discrepancy in values for MBT found in this study as compared to the values reported by Randell and Weber.

D. Butyltin in Surface Samples

The maximum concentration in the SML of the mesocosm was \(1.7 \pm 0.3 \, \mu g \text{TBT/L}\) on day one. This concentration was comparable to maximum environmental concentrations of \(1.2 \, \mu g \text{TBT/L}\) reported by Hall et al. (1987) and \(2.7 \, \mu g \text{TBT/L}\) reported by Cleary and Stebbing (1987). The enrichment factor was also found to be representative of environmental values. On the first day of the experiment, activity in surface samples was found to be higher than that in the underlying water column by a factor of \(3.0 \pm 0.1\). The enrichment
factor decreased with time, reaching a value of \(0.75 \pm 0.01\) by day 29. Presumably the enrichment factor decreased because of the change in composition of butyltins in the water column. These enrichment factors were comparable to reported values of \(4 \pm 9\) by Hall et al. and \(9 \pm 10\) by Cleary and Stebbing.

E. Conclusions

The use of a marine mesocosm for this experimental work resulted in a significant increase in our knowledge of the geochemistry of TBT, compared with that gained from measuring and monitoring in the environment. While the experimental results were not discordant with expectations from previous experiments, the greater control over the variables to be measured, as well as the use of \(^{14}\text{C}\)-labelled TBT, resulted in new evidence about several aspects of TBT's behavior in nature. These include the degradation rate of TBT as well as the transport rate from the water column.

It has generally been assumed that TBT degrades through a process of sequential debutylation, through DBT to MBT. Based on this assumption, the degradation rate of TBT has usually been calculated as the production rate of DBT. The results of this investigation indicate that TBT may degrade directly to MBT. If this reaction sequence does take place, models based on sequential debutylation would lead to an underestimation of the total degradation rate of TBT.

The removal rate of TBT is also a function of the transport rates from the water column. This experiment found two indications that the transport rates may be higher than was previously appriciated. First, the distribution coefficients found in this investigation were higher than those previously reported. A larger distribution coefficient would result in a larger fraction of TBT adsorbed to particles. Transport to the sediment would therefore be enhanced. In addition, the rate at which TBT would be transferred from the sediment back to the water column would also be lower than anticipated. The combined results of a higher distribution coefficient would therefore be a lower estimation of the concentration of TBT in the water column.

Another reason TBT's residence time in the water column may be less than previously thought is the possible transport of TBT to the atmosphere. This process, although only indicated by the result of
an activity imbalance in the mesocosm, was of the same order of magnitude as transport to the sediment. Ignoring this transport pathway could also lead to calculation of a higher concentration of TBT in the water column. In addition, if it were assumed that all TBT not in the water column had been transported to the sediment, this would cause an over estimation of the concentration of TBT in the sediment.

In summary, the results of this investigation indicate that the persistence of TBT in the coastal marine environment may be less than previously thought. Further study of these processes are warranted.
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Appendix - A

Data Tabulation of Results
Table A-1. Total activity measured in the water, extractable and nonextractable activity as well as fraction of extracted activity attributed to TBT.

<table>
<thead>
<tr>
<th>Day</th>
<th>Direct (dpm/g of Water)</th>
<th>Extracted (dpm/g of Water)</th>
<th>Nonextracted (dpm/g of Water)</th>
<th>Frac. TBT</th>
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</thead>
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<tr>
<td>0.1</td>
<td>74 +/- 1</td>
<td>79.7 +/- 2.0</td>
<td>0.6</td>
<td>0.89</td>
</tr>
<tr>
<td>0.3</td>
<td>82 +/- 3</td>
<td>77.2 +/- 0.0</td>
<td>1.1</td>
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<td>0.5</td>
<td>72 +/- 4</td>
<td>71.5 +/- 0.8</td>
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<td>0.96</td>
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<tr>
<td>0.8</td>
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<tr>
<td>1.0</td>
<td>69 +/- 2</td>
<td>63.8 +/- 0.5</td>
<td>2.3 E</td>
<td>0.85E</td>
</tr>
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<td>69 +/- 2</td>
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<td>32.5 +/- 0.1</td>
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<td>0.64</td>
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<td>27.8 +/- 0.2</td>
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<td>25.0 +/- 0.4 E</td>
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<td>14.0</td>
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*Note: E = Estimated.*
Table A-2. Component activity distribution as calculated from the model equations and as corrected by graphical interpolation for values of TBT and MBT. Values in dpm/g of water.

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<td>DBT</td>
<td>MBT</td>
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<td>9.3</td>
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<td>9.1</td>
</tr>
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</tr>
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<td>2.3</td>
<td>8.4</td>
<td>8.0</td>
</tr>
<tr>
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<td>2.4</td>
<td>6.8</td>
<td>8.5</td>
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<td>1.5</td>
<td>7.1</td>
<td>8.4</td>
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<td>8.0</td>
<td>9.0</td>
</tr>
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<td>0.9</td>
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<td>9.4</td>
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<td>13.1</td>
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<td>11.8</td>
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<td>34.5</td>
<td>0.5</td>
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<td>10.6</td>
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<td>0.2</td>
<td>1.2</td>
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</tr>
<tr>
<td>41.5</td>
<td>0.3</td>
<td>2.5</td>
<td>11.7</td>
</tr>
</tbody>
</table>

*Note: Bold type indicates numbers which were corrected.*
Table A-3. Comparison of calculated activity of butyltins plus measured values of $^{14}$CO$_2$ to total water column measured activity.

<table>
<thead>
<tr>
<th>Day</th>
<th>Butyltins</th>
<th>CO$_2$</th>
<th>$\Sigma$</th>
<th>Measured</th>
<th>$\Sigma$/Measured Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>79.8</td>
<td>-</td>
<td>79.8</td>
<td>74 +/- 1</td>
<td>1.08</td>
</tr>
<tr>
<td>0.3</td>
<td>78.0</td>
<td>-</td>
<td>78.0</td>
<td>82 +/- 3</td>
<td>0.95</td>
</tr>
<tr>
<td>0.5</td>
<td>78.3</td>
<td>-</td>
<td>78.3</td>
<td>72 +/- 4</td>
<td>1.09</td>
</tr>
<tr>
<td>0.8</td>
<td>70.2</td>
<td>0.1</td>
<td>70.3</td>
<td>75 +/- 2</td>
<td>0.94</td>
</tr>
<tr>
<td>1.0</td>
<td>66.0</td>
<td>0.2</td>
<td>66.2</td>
<td>69 +/- 2</td>
<td>0.96</td>
</tr>
<tr>
<td>1.5</td>
<td>59.6</td>
<td>-</td>
<td>59.6</td>
<td>69 +/- 2</td>
<td>0.86</td>
</tr>
<tr>
<td>2.5</td>
<td>50.9</td>
<td>0.8</td>
<td>51.7</td>
<td>59 +/- 2</td>
<td>0.88</td>
</tr>
<tr>
<td>3.5</td>
<td>49.1</td>
<td>0.8</td>
<td>49.9</td>
<td>63 +/- 1</td>
<td>0.79</td>
</tr>
<tr>
<td>4.5</td>
<td>39.6</td>
<td>11.0</td>
<td>50.6</td>
<td>57 +/- 1</td>
<td>0.89</td>
</tr>
<tr>
<td>5.5</td>
<td>37.9</td>
<td>3.1</td>
<td>41.0</td>
<td>56 +/- 1</td>
<td>0.73</td>
</tr>
<tr>
<td>6.5</td>
<td>36.0</td>
<td>3.3</td>
<td>39.3</td>
<td>43 +/- 1</td>
<td>0.91</td>
</tr>
<tr>
<td>7.5</td>
<td>33.7</td>
<td>1.5</td>
<td>35.2</td>
<td>43 +/- 1</td>
<td>0.82</td>
</tr>
<tr>
<td>8.5</td>
<td>31.0</td>
<td>2.0</td>
<td>33.0</td>
<td>41 +/- 2</td>
<td>0.81</td>
</tr>
<tr>
<td>9.5</td>
<td>28.0</td>
<td>3.6</td>
<td>31.5</td>
<td>41 +/- 1</td>
<td>0.77</td>
</tr>
<tr>
<td>10.5</td>
<td>25.2</td>
<td>7.4</td>
<td>32.6</td>
<td>39 +/- 1</td>
<td>0.84</td>
</tr>
<tr>
<td>11.5</td>
<td>25.0</td>
<td>8.3</td>
<td>33.3</td>
<td>39 +/- 1</td>
<td>0.85</td>
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<tr>
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<td>23.2</td>
<td>8.5</td>
<td>31.7</td>
<td>37 +/- 1</td>
<td>0.86</td>
</tr>
<tr>
<td>14.5</td>
<td>22.0</td>
<td>9.2</td>
<td>31.2</td>
<td>36 +/- 1</td>
<td>0.87</td>
</tr>
<tr>
<td>16.5</td>
<td>18.7</td>
<td>10.4</td>
<td>29.1</td>
<td>35 +/- 0</td>
<td>0.83</td>
</tr>
<tr>
<td>18.5</td>
<td>17.7</td>
<td>11.7</td>
<td>29.4</td>
<td>34 +/- 1</td>
<td>0.86</td>
</tr>
<tr>
<td>20.5</td>
<td>17.0</td>
<td>12.0</td>
<td>29.0</td>
<td>33 +/- 1</td>
<td>0.88</td>
</tr>
<tr>
<td>22.5</td>
<td>18.4</td>
<td>13.1</td>
<td>31.5</td>
<td>35 +/- 1</td>
<td>0.90</td>
</tr>
<tr>
<td>24.5</td>
<td>16.4</td>
<td>14.2</td>
<td>30.6</td>
<td>34 +/- 1</td>
<td>0.90</td>
</tr>
<tr>
<td>27.5</td>
<td>16.7</td>
<td>15.8</td>
<td>32.5</td>
<td>30 +/- 1</td>
<td>1.08</td>
</tr>
<tr>
<td>28.5</td>
<td>16.3</td>
<td>16.2</td>
<td>32.5</td>
<td>32.0 +/- 0.3</td>
<td>1.02</td>
</tr>
<tr>
<td>30.5</td>
<td>17.0</td>
<td>17.0</td>
<td>34.0</td>
<td>34.3 +/- 0.5</td>
<td>0.99</td>
</tr>
<tr>
<td>34.5</td>
<td>14.5</td>
<td>17.0</td>
<td>31.5</td>
<td>26.1 +/- 0.4</td>
<td>1.21</td>
</tr>
<tr>
<td>38.5</td>
<td>13.0</td>
<td>18.8</td>
<td>31.8</td>
<td>31.4 +/- 0.3</td>
<td>1.01</td>
</tr>
<tr>
<td>41.5</td>
<td>14.5</td>
<td>19.2</td>
<td>33.7</td>
<td>30.8 +/- 0.5</td>
<td>1.09</td>
</tr>
</tbody>
</table>

Mean: 0.90 +/- 0.10

Note: $\Sigma = \text{Butyltins} + \text{CO}_2$

Measured = Direct count of unprocessed samples.
Table A-4. Concentration of tri-, di- and monobutyltin. Values in nmol/L.

<table>
<thead>
<tr>
<th>TBT</th>
<th>DBT</th>
<th>MBT</th>
</tr>
</thead>
<tbody>
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<td>0.3</td>
<td>1.82</td>
<td>0.09</td>
</tr>
<tr>
<td>0.5</td>
<td>1.89</td>
<td>-0.04</td>
</tr>
<tr>
<td>0.8</td>
<td>1.73</td>
<td>0.19</td>
</tr>
<tr>
<td>1.0</td>
<td>1.61</td>
<td>0.05</td>
</tr>
<tr>
<td>1.5</td>
<td>1.39</td>
<td>0.21</td>
</tr>
<tr>
<td>2.5</td>
<td>1.08</td>
<td>0.44</td>
</tr>
<tr>
<td>3.5</td>
<td>0.99</td>
<td>0.25</td>
</tr>
<tr>
<td>4.5</td>
<td>0.86</td>
<td>0.33</td>
</tr>
<tr>
<td>5.5</td>
<td>0.74</td>
<td>0.15</td>
</tr>
<tr>
<td>6.5</td>
<td>0.66</td>
<td>0.20</td>
</tr>
<tr>
<td>7.5</td>
<td>0.51</td>
<td>0.25</td>
</tr>
<tr>
<td>8.5</td>
<td>0.42</td>
<td>0.34</td>
</tr>
<tr>
<td>9.5</td>
<td>0.38</td>
<td>0.28</td>
</tr>
<tr>
<td>10.5</td>
<td>0.23</td>
<td>0.44</td>
</tr>
<tr>
<td>11.5</td>
<td>0.19</td>
<td>0.39</td>
</tr>
<tr>
<td>12.5</td>
<td>0.21</td>
<td>0.39</td>
</tr>
<tr>
<td>13.5</td>
<td>0.11</td>
<td>0.35</td>
</tr>
<tr>
<td>14.5</td>
<td>0.11</td>
<td>0.35</td>
</tr>
<tr>
<td>16.5</td>
<td>0.06</td>
<td>0.30</td>
</tr>
<tr>
<td>18.5</td>
<td>0.06</td>
<td>0.24</td>
</tr>
<tr>
<td>20.5</td>
<td>0.04</td>
<td>0.26</td>
</tr>
<tr>
<td>22.5</td>
<td>0.03</td>
<td>0.29</td>
</tr>
<tr>
<td>24.5</td>
<td>0.02</td>
<td>0.22</td>
</tr>
<tr>
<td>27.5</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>28.5</td>
<td>0.02</td>
<td>0.19</td>
</tr>
<tr>
<td>30.5</td>
<td>0.01</td>
<td>0.17</td>
</tr>
<tr>
<td>34.5</td>
<td>0.01</td>
<td>0.12</td>
</tr>
<tr>
<td>38.5</td>
<td>0.00</td>
<td>0.04</td>
</tr>
<tr>
<td>41.5</td>
<td>0.00</td>
<td>0.09</td>
</tr>
</tbody>
</table>
Table A-5. Total activity in the sediment. Values in dpm/g of water after conversion from dpm/core.

<table>
<thead>
<tr>
<th>Day Analyzed</th>
<th>7</th>
<th>15</th>
<th>22</th>
<th>29</th>
<th>36</th>
<th>49</th>
</tr>
</thead>
<tbody>
<tr>
<td>120</td>
<td>13.6</td>
<td>26.0</td>
<td>14.2</td>
<td>27.4</td>
<td>18.9</td>
<td>6.4</td>
</tr>
<tr>
<td>128</td>
<td>7.8</td>
<td>38.0</td>
<td></td>
<td></td>
<td>22.9</td>
<td>17.6</td>
</tr>
<tr>
<td>130</td>
<td></td>
<td></td>
<td></td>
<td>24.0</td>
<td>13.5</td>
<td>24.8</td>
</tr>
<tr>
<td>211</td>
<td>15.7</td>
<td></td>
<td>3.0</td>
<td>42.7</td>
<td>76.3</td>
<td></td>
</tr>
<tr>
<td>218</td>
<td></td>
<td>9.8</td>
<td>14.0</td>
<td>19.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>293</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>303</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Mean: 12 ± 4 25 ± 14 14 ± 9 26 ± 13 24 ± 4 35 ± 30 20 ± 20

Note: 
\[ \text{dpm/g of water} = (\text{dpm/Core}) \times (\text{Core}/2.55 \times 10^3 \text{ g of water}) \].

(1) Mean for day 49 calculated without value of 76.3 dpm/g.

Table A-6. Mesocosm activity distribution. Values in percent.

<table>
<thead>
<tr>
<th>Day</th>
<th>Water</th>
<th>Sediment</th>
<th>Atmosphere</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.5</td>
<td>55</td>
<td>15</td>
<td>-</td>
<td>70</td>
</tr>
<tr>
<td>14.5</td>
<td>46</td>
<td>32</td>
<td>-</td>
<td>79</td>
</tr>
<tr>
<td>22.5</td>
<td>44</td>
<td>18</td>
<td>1</td>
<td>63</td>
</tr>
<tr>
<td>28.5</td>
<td>41</td>
<td>33</td>
<td>2</td>
<td>76</td>
</tr>
<tr>
<td>38.5</td>
<td>40</td>
<td>31</td>
<td>2</td>
<td>73</td>
</tr>
<tr>
<td>48.5</td>
<td>40</td>
<td>26</td>
<td>3</td>
<td>69</td>
</tr>
</tbody>
</table>

Note: 
Water = Direct measurement of unprocessed samples.
Sediment = Mean value from sediment analysis.
Atmosphere = Calculated exchange of CO₂ at rate of 1%/d.
Table A-7. Activity on particles in dpm/g of water and as percent of total activity in the water column.

<table>
<thead>
<tr>
<th>Day</th>
<th>dpm/g of Water</th>
<th>Percent on Particles</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Particles</td>
<td>Water</td>
</tr>
<tr>
<td>0.1</td>
<td>3.42</td>
<td>74</td>
</tr>
<tr>
<td>0.3</td>
<td>3.20</td>
<td>82</td>
</tr>
<tr>
<td>0.5</td>
<td>0.66</td>
<td>72</td>
</tr>
<tr>
<td>0.8</td>
<td>0.58</td>
<td>75</td>
</tr>
<tr>
<td>1.0</td>
<td>0.41</td>
<td>69</td>
</tr>
<tr>
<td>1.5</td>
<td>0.14</td>
<td>69</td>
</tr>
<tr>
<td>2.5</td>
<td>2.33</td>
<td>58</td>
</tr>
<tr>
<td>3.5</td>
<td>2.47</td>
<td>62</td>
</tr>
<tr>
<td>4.5</td>
<td>1.87</td>
<td>46</td>
</tr>
<tr>
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<td>1.09</td>
<td>53</td>
</tr>
<tr>
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<td>0.83</td>
<td>40</td>
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<tr>
<td>7.5</td>
<td>1.52</td>
<td>42</td>
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<tr>
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<td>0.89</td>
<td>39</td>
</tr>
<tr>
<td>9.5</td>
<td>0.69</td>
<td>38</td>
</tr>
<tr>
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<tr>
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<tr>
<td>16.5</td>
<td>0.64</td>
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</tr>
<tr>
<td>18.5</td>
<td>0.22</td>
<td>23.2</td>
</tr>
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<td>0.17</td>
<td>21.4</td>
</tr>
<tr>
<td>22.5</td>
<td>0.46</td>
<td>22.3</td>
</tr>
<tr>
<td>24.5</td>
<td>0.15</td>
<td>20.6</td>
</tr>
<tr>
<td>27.5</td>
<td>0.05 E</td>
<td>15.6</td>
</tr>
<tr>
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<td>0.07</td>
<td>17.1</td>
</tr>
<tr>
<td>30.5</td>
<td>0.11</td>
<td>18.8</td>
</tr>
<tr>
<td>34.5</td>
<td>0.10</td>
<td>10.7</td>
</tr>
<tr>
<td>38.5</td>
<td>0.17</td>
<td>14.4</td>
</tr>
<tr>
<td>41.5</td>
<td>0.05</td>
<td>13.6</td>
</tr>
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<td>0.03</td>
<td>12.1</td>
</tr>
<tr>
<td>48.5</td>
<td>0.02</td>
<td>13.2</td>
</tr>
<tr>
<td>52.5</td>
<td>0.05</td>
<td>12.4</td>
</tr>
</tbody>
</table>
Table A-8. Fraction of activity extracted from particles attributed to TBT.

<table>
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<tr>
<th>Day</th>
<th>TBT</th>
<th>DBT</th>
<th>Fraction TBT</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5</td>
<td>1.10E</td>
<td>0.01</td>
<td>0.98</td>
</tr>
<tr>
<td>2.5</td>
<td>1.37</td>
<td>0.00</td>
<td>1.00</td>
</tr>
<tr>
<td>3.5</td>
<td>1.93</td>
<td>0.05</td>
<td>0.94</td>
</tr>
<tr>
<td>4.5</td>
<td>1.59</td>
<td>0.63</td>
<td>0.49</td>
</tr>
<tr>
<td>5.5</td>
<td>0.74</td>
<td>0.00</td>
<td>1.00</td>
</tr>
<tr>
<td>6.5</td>
<td>0.32</td>
<td>0.00</td>
<td>1.00</td>
</tr>
<tr>
<td>7.5</td>
<td>2.66</td>
<td>1.36</td>
<td>0.42</td>
</tr>
<tr>
<td>8.5</td>
<td>0.89</td>
<td>0.08</td>
<td>0.81</td>
</tr>
<tr>
<td>9.5</td>
<td>0.33</td>
<td>0.00</td>
<td>1.00</td>
</tr>
<tr>
<td>11.5</td>
<td>14.54</td>
<td>13.51</td>
<td>0.27</td>
</tr>
<tr>
<td>13.5</td>
<td>0.22</td>
<td>0.01</td>
<td>0.89</td>
</tr>
<tr>
<td>14.5</td>
<td>3.45</td>
<td>1.89</td>
<td>0.41</td>
</tr>
<tr>
<td>16.5</td>
<td>6.51</td>
<td>4.49</td>
<td>0.35</td>
</tr>
<tr>
<td>18.5</td>
<td>2.57</td>
<td>2.51</td>
<td>0.28</td>
</tr>
</tbody>
</table>

\[
\text{Fraction TBT} = \frac{\text{TBT}/0.88}{\text{TBT}/0.88 + \text{DBT}/0.33}
\]

Table A-9. Calculation of distribution coefficient for TBT. Activities in dpm of TBT/g of water. $K_d$ in $(\mu g/Kg)/(\mu g/L)$.

<table>
<thead>
<tr>
<th>Day</th>
<th>Particulate Activity</th>
<th>Water Activity</th>
<th>Part. Wt. mg/g H2O</th>
<th>$K_d \times 10^{-3}$</th>
</tr>
</thead>
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<td>0.78</td>
<td>72.5</td>
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<td>-</td>
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<td>0.83</td>
<td>21.4</td>
<td>0.82</td>
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<td>0.82</td>
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<td>0.12</td>
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</table>

Mean \[ 60 \pm 30 \]

Note: $K_d = (1,020 \text{ g water/L}) (\text{TBT on Part.}/\text{TBT in Water})/ (\text{Part. Wt.} \times 10^{-3})$
Table A-10. Calculation of distribution coefficient for DBT. Activities in dpm of DBT/g of water. $K_d$ in $(\mu g/Kg)/(\mu g/L)$.

<table>
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<tr>
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<th>Particulate Activity</th>
<th>Water Activity</th>
<th>Part. Wt. mg/g H$_2$O</th>
<th>$K_d \times 10^{-3}$</th>
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</thead>
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<td>-</td>
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<td>0.00</td>
<td>7.0</td>
<td>0.82</td>
<td>-</td>
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<td>0.00</td>
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<td>-</td>
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<td>1.08</td>
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<td>0.00</td>
<td>10.8</td>
<td>0.82</td>
<td>-</td>
</tr>
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<td>0.34</td>
<td>6.9</td>
<td>0.70</td>
<td>70.9</td>
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<td>4.0</td>
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<td>0.16</td>
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<td>30 ± 20</td>
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*Mean: See notes of Table A-9.*

Table A-11. Calculation of distribution coefficient for MBT. Particulate and water column activities in units of dpm/g of water. $K_d$ is in $(\mu g/Kg)/(\mu g/L)$.

<table>
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<tr>
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<th>Particulate Activity</th>
<th>Water Activity</th>
<th>Part. Wt. mg/g H$_2$O</th>
<th>$K_d \times 10^{-3}$</th>
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<td>0.72</td>
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<td></td>
<td>2.9 ± 0.5</td>
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Table A-12. The ratio of activity in surface samples to activity in the water column.

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<th>Column dpm/g</th>
<th>Ratio Surf./Column</th>
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<td>75 ± 2</td>
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<tr>
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<td>205 ± 1</td>
<td>69 ± 2</td>
<td>3.0 ± 0.1</td>
</tr>
<tr>
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<td>93 ± 5</td>
<td>63 ± 1</td>
<td>1.5 ± 0.1</td>
</tr>
<tr>
<td>7.5</td>
<td>47 ± 7</td>
<td>43 ± 4</td>
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<td>38 ± 1</td>
<td>39 ± 1</td>
<td>1.0 ± 0.0</td>
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<td>0.89 ± 0.02</td>
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<td>0.75 ± 0.01</td>
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Appendix - B

Data Calculations
Table B-1. Total Water Column Activity.

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<th>dpm/g of Water</th>
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</thead>
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**Note:** Continued on next page.
Table B-1 Continued. Total Water Column Activity.

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Continued. Total Water Column Activity.

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Eff. = 0.89 cpm/dpm
Activity = (cpm - Blank)(Extract)/(Eff. x Aliquot x Water)
Day 52.5 aliquot 2
= (98 - 25)(19.630)/(0.89 x 1.500 x 514)
= 2.09 dpm/g of H₂O
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Note: E = Estimate
Blank = 35 cpm
Activity = (cpm - Blank)/(Eff. x Weight)
Day 52.5

= (143 - 35)/(0.75 x 10.31)

= 14.0 dpm/g of H2O
## Table B-4. Tributyltin Activity in Extracted Phase.

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<th>dpm/g</th>
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Note:  
1) Start Sample Storage  
2) End Sample Storage  
E = Estimate  
Blank = 25 cpm  
Efficiency = 0.87  
Activity = \((cpm - Blank) \times (Hexane)/(Eff. \times Sample \times Water)\)  
Day 41.5  
\[= (930 - 25) \times (21.47)/(0.87 \times 18.47 \times 516)\]  
\[= 2.34 \text{ dpm/g H}_2\text{O}\]
Table B-5. Total Extractable Dibutyltin.

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<th>dpm/g</th>
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E = Estimate

Blank = 25 cpm
Efficiency = 0.88
Activity = (cpm - Blank) x (Hexane)/(Eff. x Sample x Water)
Day 41.5

= (1613 - 25) x (21.47)/(0.88 x 18.47 x 516)
= 4.07 dpm/g H₂O
Table B-6. TBT as fraction of activity in hexane.

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\[ E = \text{Estimate} \]
\[
\text{Fraction TBT} = \frac{\text{TBT}/0.88}{\text{TBT}/0.88 + \text{DBT}/0.33}
\]
Table B-7. $^{14}\text{CO}_2$ In Water and corrected for loss to the atmosphere.

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Note: $E =$ Estimate
Blank = 41 cpm
Efficiency = 0.84
Weight = 503 gm
Activity = (cpm - Blank)/(Eff. x Weight)
Day 52.5
= (8081 - 41)/(0.84 x 503)
= 19.0 dpm/g of H$_2$O
**Table B-8. Total Activity on Particles.**

<table>
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<th>Day</th>
<th>H</th>
<th>Eff.</th>
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<th>dpm/g</th>
<th>cpm</th>
<th>dpm/g</th>
<th>F - B</th>
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<td>126</td>
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**Note:** E = Estimate  
Blank = 25 cpm  
Sample Weight = 1020 g  
Efficiency for Blanks = 0.87  
Activity = (cpm - Blank)/(Eff. X Weight)

**Day 52.5**

Filter Activity = (172 - 25)/(0.85 X 1020)  
= 0.17 dpm/g H₂O  
Blank Activity = (126 - 25)/(0.87 X 1020)  
= 0.12  
Activity on Particles = (F - B)  
= (0.17 - 0.12) = 0.05 dpm/g H₂O
### Table B-9. Blank for Total Activity Extracted from Particles.

<table>
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<th>Day</th>
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<th>g</th>
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Note: See notes of Table B-10.

### Table B-10. Total Activity Extracted from Particles.

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<th>dpm/g</th>
<th>F - B</th>
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Note: E = Estimate, Blank = 25 cpm, Efficiency = 0.89, Sample Wt. = 1020 g
Activity = (cpm - Blank) (Hexane Wt)/(Eff. X Sample Wt X Water Wt)
For Day 18.5
Filter Activity = (47 - 25) (10.10)/(0.89 X 1.50 X 1020)
= 0.16 dpm/g H₂O
Blank Activity = (29 - 25) X (10.36)/(0.88 X 1.51 X 1020)
= 0.03
Sample Activity = (F - B) = 0.13 dpm/g H₂O
Table B-11. Calculation of Blanks for TBT Extracted from Particles.

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<th>Weight g</th>
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Note: See notes of Table B-12.

Table B-12. Calculation of TBT Extracted from Particles.

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<th>H</th>
<th>Efficiency</th>
<th>Weight g</th>
<th>dpm/g</th>
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</table>

Note: E = Estimate, Blank = 25 cpm, Water Sample Wt. = 1020 g
Activity = (cpm - Blank) (Hex. Wt.)/(Eff.X Sample Wt.X H2O Wt.)

Day 18.5
Sample Activity = (1951 - 25) X (10.10)/(0.85 X 8.61 X 1020)
= 2.58 dpm/g H2O
Blank Activity = (35 - 25)(10.36)/(0.85 X 8.85 X 1020)
= 0.01 dpm/g H2O
TBT Activity = F-B = (Sample dpm) - (Blank dpm)
= 2.58 - 0.01 = 2.57 dpm/g H2O
Table B-13. Calculation of Blanks for DBT Extracted from Particles.

<table>
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<tr>
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<th>Hexane</th>
<th>dpm/g</th>
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</thead>
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See notes on Table B-14.

Table B-14. Calculation of DBT Extracted from Particles.

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<th>dpm/g</th>
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Note: Blank = 25 cpm, Efficiency = 0.88, Water Sample Wt. = 1020 g
Activity = (cpm - Blank) (Hex. Wt.)/(Eff. X Sample Wt. X Water Wt.)

Day 18.5
Sample Act. = ((1951 - 25) (10.10)/(0.85 X 8.61 X 1020)
= 2.51 dpm/g H_2O

Blank Act. = (28 - 25) X (10.36)/(0.85 X 8.85 X 1020)
= 0.00 dpm/g H_2O

DBT activity = (F - B) = (Sample Activity) - (Blank Activity)
= 2.51 - 0.00 = 2.51 dpm/g H_2O
### Table B-15. TBT as fraction of activity extracted from particles.

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<td>1.00</td>
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</tr>
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\[ F = \frac{\text{TBT}}{0.88} \]
\[ F = \frac{\text{TBT}}{0.88} + \frac{\text{DBT}}{0.33} \]

### Table B-16. Suspended Particulate Concentration.

<table>
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<th>Particulates mg/L</th>
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**Note:** Particulate Weight = 1.02(After - Before)/(Sample Wt.)

For Day 52.5

Particulate Wt. = 1.02(14.766 - 14.040)/(0.508)

= 1.46 mg/L
### Table B-17. Chlorophyll and Paeophytin Concentrations. Values in µg/L.

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<td>30</td>
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</tr>
<tr>
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Table B-18. Activity in Surface Samples.

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<th>S. Dev.</th>
<th>Error Percent</th>
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Note: Blank = 35 cpm  
Efficiency: Based on Sample Weight.  
0.79 for 10 g  
0.73 for 11 g  
0.70 for 12 g  
Activity = (cpm - Blank)/(Eff. X Wt.)  
Day 28.5 = (230 - 35)/(0.73 X 11.127)  
= 27.4 dpm/g of H₂O
Appendix - C

Process Efficiency Data
Table C-1. Calculation of reference solution composition from TLC of solutions with replicates.

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Continued on next page.
Table C-1. Calculation of reference solution composition.

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Note: Blank = 25 cpm  
Efficiency = 0.88  
Activity = (cpm - Blank)/(Eff.)  
Percent = (Activity of Component)/(Total Activity of Solution)

Table C-2. Distribution of extracted component. Values in percent.

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<th>Aqueous</th>
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Table C-3. Activity distribution when seawater spiked with TBT reference solution is extracted with hexane.

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<td></td>
<td>13.2</td>
<td></td>
</tr>
<tr>
<td>213</td>
<td>9,270 0.200 53,100</td>
<td>10.10 534 ± 3</td>
</tr>
<tr>
<td></td>
<td>9,240 0.201 52,700</td>
<td></td>
</tr>
<tr>
<td>Organic</td>
<td>42,400 1.50 32,500</td>
<td>15.78 513 ± 1</td>
</tr>
<tr>
<td></td>
<td>42,500 1.50 32,500</td>
<td></td>
</tr>
<tr>
<td></td>
<td>32,500</td>
<td>32,500</td>
</tr>
<tr>
<td>Aqueous</td>
<td>153 5.10 30.8</td>
<td>515 16 ± 0</td>
</tr>
<tr>
<td></td>
<td>155 5.10 31.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>31.1</td>
<td></td>
</tr>
<tr>
<td>Glass</td>
<td>91 0.198</td>
<td>2.73 1.0</td>
</tr>
<tr>
<td></td>
<td>48 0.200</td>
<td>2.87 0.4</td>
</tr>
<tr>
<td></td>
<td>42 0.204</td>
<td>1.75 0.2</td>
</tr>
</tbody>
</table>

Mean:
- Organic 94 ± 2
- Aqueous 2.4 ± 0.6
- Glass 0.4
- Unknown (By Difference) 3 ± 2

Note:

<table>
<thead>
<tr>
<th>Blank cpm</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Organic Phase</td>
<td>25</td>
</tr>
<tr>
<td>Aqueous Phase</td>
<td>35</td>
</tr>
</tbody>
</table>
Table C-4. Activity distribution when seawater spiked with DBT reference solution No. 1. is extracted with hexane.

<table>
<thead>
<tr>
<th>Date</th>
<th>Phase</th>
<th>Aliquot Counted</th>
<th>Total Phase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>cpm</td>
<td>Wt. g</td>
</tr>
<tr>
<td>78 Spike</td>
<td>233</td>
<td>0.198</td>
<td>1,150</td>
</tr>
<tr>
<td></td>
<td>204</td>
<td>0.197</td>
<td>1,050</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1,100</td>
</tr>
<tr>
<td>Organic</td>
<td>708</td>
<td>1.50</td>
<td>511</td>
</tr>
<tr>
<td></td>
<td>650</td>
<td>1.50</td>
<td>468</td>
</tr>
<tr>
<td></td>
<td>603</td>
<td>1.50</td>
<td>434</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>471</td>
</tr>
<tr>
<td>Aqueous</td>
<td>67</td>
<td>10.59</td>
<td>4.1</td>
</tr>
<tr>
<td></td>
<td>99</td>
<td>10.43</td>
<td>8.2</td>
</tr>
<tr>
<td></td>
<td>73</td>
<td>10.42</td>
<td>4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>5.7</td>
</tr>
<tr>
<td>85 Spike</td>
<td>1,460</td>
<td>0.197</td>
<td>8,170</td>
</tr>
<tr>
<td></td>
<td>1,450</td>
<td>0.198</td>
<td>8,100</td>
</tr>
<tr>
<td></td>
<td>1,510</td>
<td>0.202</td>
<td>8,240</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>8,170</td>
</tr>
<tr>
<td>Organic</td>
<td>3,800</td>
<td>1.50</td>
<td>2,860</td>
</tr>
<tr>
<td></td>
<td>3,820</td>
<td>1.50</td>
<td>2,870</td>
</tr>
<tr>
<td></td>
<td>3,840</td>
<td>1.50</td>
<td>2,880</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2,870</td>
</tr>
<tr>
<td>Aqueous</td>
<td>474</td>
<td>10.32</td>
<td>56.7</td>
</tr>
<tr>
<td></td>
<td>477</td>
<td>10.32</td>
<td>57.1</td>
</tr>
<tr>
<td></td>
<td>473</td>
<td>10.29</td>
<td>56.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>56.8</td>
</tr>
<tr>
<td>213 Spike</td>
<td>755</td>
<td>0.203</td>
<td>4,000</td>
</tr>
<tr>
<td></td>
<td>760</td>
<td>0.206</td>
<td>3,960</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3,980</td>
</tr>
<tr>
<td>Organic</td>
<td>2,460</td>
<td>1.50</td>
<td>1,800</td>
</tr>
<tr>
<td></td>
<td>2,450</td>
<td>1.51</td>
<td>1,790</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1,790</td>
</tr>
<tr>
<td>Aqueous</td>
<td>135</td>
<td>5.10</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>142</td>
<td>5.15</td>
<td>24</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>24</td>
</tr>
<tr>
<td>Glass</td>
<td>53</td>
<td>0.209</td>
<td>154</td>
</tr>
<tr>
<td></td>
<td>47</td>
<td>0.202</td>
<td>125</td>
</tr>
<tr>
<td></td>
<td>38</td>
<td>0.204</td>
<td>73</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.54</td>
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<tr>
<td>Mean Organic</td>
<td>70 ± 5</td>
<td></td>
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<tr>
<td>Aqueous</td>
<td>30 ± 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Glass</td>
<td>1.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: See Note at bottom of Table C-3 for blank and counting efficiency.
Table C-5. Activity distribution when seawater spiked with DBT reference solution No. 2 is extracted with hexane.

<table>
<thead>
<tr>
<th>Sample Phase</th>
<th>Aliquot Counted</th>
<th>Total Phase</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>cpm</td>
<td>Wt. g</td>
<td>dpm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Spike</td>
<td>Aqueous</td>
<td>10.2</td>
<td>16.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>75.3</td>
<td>10.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>77.2</td>
<td>10.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.51</td>
<td>309</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.50</td>
<td>304</td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.61</td>
<td>5.1</td>
</tr>
<tr>
<td></td>
<td>Aqueous</td>
<td>10.36</td>
<td>4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10.29</td>
<td>5.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.50</td>
<td>313</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.50</td>
<td>287</td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.50</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>Aqueous</td>
<td>10.26</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10.21</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10.21</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td>1 Spike</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.50</td>
<td>293</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.50</td>
<td>287</td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>1.50</td>
<td>290</td>
</tr>
<tr>
<td></td>
<td>Aqueous</td>
<td>10.25</td>
<td>5.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10.24</td>
<td>6.0</td>
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<td></td>
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<td></td>
<td>5.7</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>5.7</td>
</tr>
<tr>
<td></td>
<td>Glass</td>
<td>0.50</td>
<td>27</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.50</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Known (By Difference)</td>
<td>100</td>
<td>1.2</td>
</tr>
<tr>
<td></td>
<td>Known (By Difference)</td>
<td>32 ± 3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Known (By Difference)</td>
<td>1 ± 0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Known (By Difference)</td>
<td>4 ± 3</td>
<td></td>
</tr>
<tr>
<td>Blank (cpm)</td>
<td>Spike</td>
<td>25.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>9.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Aqueous</td>
<td>25.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Glass</td>
<td>9.0</td>
<td></td>
</tr>
<tr>
<td>Efficiency (%)</td>
<td>Spike</td>
<td>32</td>
<td>56</td>
</tr>
<tr>
<td></td>
<td>Organic</td>
<td>9.0</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>Aqueous</td>
<td>25.6</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>Glass</td>
<td>9.0</td>
<td>44</td>
</tr>
</tbody>
</table>
| Notes: Because of counting problems, cpm are from channel No.2 only.
Table C-6. Extraction of MBT from seawater using hexane. Atomic absorption standards, results and calculation of MBT retained in seawater.

<table>
<thead>
<tr>
<th>Solution Concentration</th>
<th>mg MBTCl/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>275</td>
<td>575</td>
</tr>
<tr>
<td>STANDARD CURVES</td>
<td></td>
</tr>
<tr>
<td>0.003</td>
<td>19.0</td>
</tr>
<tr>
<td>0.015</td>
<td>37.4</td>
</tr>
<tr>
<td>0.026</td>
<td>75.5</td>
</tr>
<tr>
<td>0.053</td>
<td>151</td>
</tr>
<tr>
<td>0.104</td>
<td>302</td>
</tr>
</tbody>
</table>

DILUTION FACTORS

RESULTS

| Blank                  |            |
|                       |            |
| Replicate No. 1.      |            |
|                       |            |
| Replicate No. 2.      |            |

BEIJ::IIIAIION

\[
\frac{(256 + 248)}{2} = 0.91 \quad \frac{(463 + 523)}{2} = 0.86 \quad \frac{(857 + 860)}{2} = 0.89
\]

Mean = 0.89 ± 0.03

Note: The concentrations listed under RESULTS were calculated by interpolation of absorbence using the appropriate STANDARDS CURVE and multiplying by the DILUTION FACTOR used to bring the sample into the region of the standards curve.
Table C-7. Distribution of butyltins following vacuum evaporation of reference solutions in hexane. All values in percent.

<table>
<thead>
<tr>
<th>Component</th>
<th>Recovered</th>
<th>On Glass</th>
<th>Evaporated</th>
</tr>
</thead>
<tbody>
<tr>
<td>TBT</td>
<td>88 ± 6</td>
<td>10 ± 4</td>
<td>2 ± 7</td>
</tr>
<tr>
<td>DBT</td>
<td>33 ± 3</td>
<td>20 ± 20</td>
<td>50 ± 25</td>
</tr>
</tbody>
</table>

Note: Calculated by solution of simultaneous equations using reference solution compositions (from Table C-1) and values of recovery and loss to glassware of reference solutions (from Table C-8 and C-9).

Table C-8. Activity distribution when hexane spiked with TBT reference solution is vacuum evaporated in preparation of TLC separation.

<table>
<thead>
<tr>
<th>Day</th>
<th>Phase</th>
<th>Aliquot Counted</th>
<th>Total Phase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>cpm</td>
<td>Wt. g</td>
</tr>
<tr>
<td>65</td>
<td>Spike</td>
<td>4,950</td>
<td>0.198</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5,000</td>
<td>0.199</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5,020</td>
<td>0.200</td>
</tr>
<tr>
<td></td>
<td>Recovered</td>
<td>194,000</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Glass</td>
<td>32,300</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1,200</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1,000</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Unknown</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

| 99  | Spike | 16,100         | 0.496       | 37,700 | 14.63 | 537 ± 12 | - |
|     |       | 16,200         | 0.502       | 36,200 | 14.63 | 537     | 89.6 |
|     |       | 16,300         | 0.506       | 36,300 | 14.63 | 537     | 89.6 |
|     | Recovered | 423,000     | -           | 481,000 | 14.63 | 481     | 89.6 |
|     | Glass  | 33,900         | -           | 38,100 | 14.63 | 481     | 89.6 |
|     |       | 1,500          | -           | 1,600  | 14.63 | 481     | 89.6 |
|     |       | 400            | -           | 400    | 14.63 | 481     | 89.6 |
|     | Unknown | -             | -           | 40,100 | 14.63 | 481     | 89.6 |

Note: Continued on next page.
Table C-8 Continued. Activity distribution when hexane spiked with TBT reference solution is vacuum evaporated in preparation of TLC separation.

<table>
<thead>
<tr>
<th>Day</th>
<th>Phase</th>
<th>Aliquot Counted</th>
<th>Total Phase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>cpm</td>
<td>Wt. g</td>
</tr>
<tr>
<td>213</td>
<td>Spike</td>
<td>11,900</td>
<td>0.204</td>
</tr>
<tr>
<td></td>
<td></td>
<td>11,900</td>
<td>0.206</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Recovered</td>
<td>528,000</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Glass</td>
<td>36,600</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4,300</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td>500</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Unknown</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mean Recovered</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Glass</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Unknown</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: Blank = 25 cpm
Efficiency = 0.88
Table C-9. Activity distribution when hexane spiked with DBT reference solution No. 1 is vacuum evaporated in preparation of TLC separation.

<table>
<thead>
<tr>
<th>Spike Phase</th>
<th>Aliquot Counted</th>
<th>Total Phase</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>cpm</td>
<td>Wt. g</td>
</tr>
<tr>
<td>66 Spike</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Spike</td>
<td>357</td>
<td>0.199</td>
</tr>
<tr>
<td></td>
<td>364</td>
<td>0.202</td>
</tr>
<tr>
<td></td>
<td>339</td>
<td>0.199</td>
</tr>
<tr>
<td>Recovered</td>
<td>8,880</td>
<td>-</td>
</tr>
<tr>
<td>Glass</td>
<td>1,200</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>190</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>80</td>
<td>-</td>
</tr>
<tr>
<td>Recovered</td>
<td>2,470</td>
<td>-</td>
</tr>
<tr>
<td>Glass</td>
<td>1,470</td>
<td>0.505</td>
</tr>
<tr>
<td></td>
<td>1,480</td>
<td>0.501</td>
</tr>
<tr>
<td></td>
<td>1,470</td>
<td>0.504</td>
</tr>
<tr>
<td>Recovered</td>
<td>1,450</td>
<td>0.207</td>
</tr>
<tr>
<td>Glass</td>
<td>1,440</td>
<td>0.202</td>
</tr>
<tr>
<td></td>
<td>43,900</td>
<td>-</td>
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<tr>
<td>Unknown</td>
<td>15,100</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>2,500</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>900</td>
<td>-</td>
</tr>
<tr>
<td>Recovered</td>
<td>56</td>
<td>± 4</td>
</tr>
<tr>
<td>Glass</td>
<td>15</td>
<td>± 10</td>
</tr>
<tr>
<td>Unknown</td>
<td>30</td>
<td>± 15</td>
</tr>
</tbody>
</table>

Note: Activity on glass removed; with hexane for days 66 and 99 with MeOH for day 213

Blank = 25 cpm
Efficiency = 0.90
Appendix - D

Sediment Analysis
Analysis of cores started on day 120. Four cores were processed simultaneously. Rather than processing all four cores from one sample day, they were processed sequentially. This processing scheme was used for three reasons.

1. Reduction of systematic errors.
2. Insurance against contamination or other possible errors from invalidating an entire days data.
3. Allowance for improvement of technique to be used for samples from all days.

Several solvent systems, extraction methods and extraction times were used in the processing the 12 sets of sediment samples. The solvent systems used included; hexane, hexane/methanol, hexane/methanol/water and methylene chloride/methanol. Extraction methods included; reflux, soxlet extraction and soxlet extraction with drying agents mixed with the sediment. Time of extraction varied from 2 to 39 h. The systems used are summarized in Table D-1.

Table D-1. Sediment processing methods.

<table>
<thead>
<tr>
<th>Day Analyzed</th>
<th>Sed. Wt. gm</th>
<th>Volume ml</th>
<th>Time Hrs.</th>
<th>Note</th>
</tr>
</thead>
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Seven of the extraction series used reflux of the sediment samples. The top 5 cm of each core, about 25 g, was processed. The samples were transferred while frozen to a 250 ml round bottom flask. To this was added 10 ml each of: deionized water, methanol, and hexane. The samples were acidified with 0.5 ml of 6 N HCl and
refluxed. The first set of 4 cores were refluxed for 2 hours. The second set of 4 cores were refluxed for 7 hours. It was found that the 7 h reflux had lower recoveries than the 2 h extraction. The third set of cores were therefore refluxed for 2 h. The next two extraction sets, of 5 h, used hexane, methanol and acidified water. The sixth set of cores were refluxed for 5 h using hexane and water. In the seventh series of reflux, a complexing agent, tropolone, was included and reflux time was 3 h.

After allowing the samples to cool each was transferred to two 50 ml screw capped teflon centrifuge tubes. The round bottom flasks were rinsed 3 times with a 50:50 mixture of water and methanol which was added to the centrifuge tube. The samples were centrifuged and the hexane phase was transferred to a pear shaped flask. The sediment was resuspended, 5 ml of hexane was added and the sample was vigorously shaken. After recentrifuging the hexane phases were combined.

Replicate 0.5 g aliquots were removed from the hexane phase and counted. Replicate 0.5 g aliquots of the aqueous phase were also counted. The residue sediment was stored frozen in a 20 ml scintillation vial for later analysis by combustion. The remainder of the hexane phase was vacuum evaporated, separated by TLC and counted.

The remaining 5 sets of sediment samples were processed using soxlet extraction. Sediment preparation for the first 3 sets consisted of placing the top 5 cm of the frozen core into the soxlet thimble. Two of the sets used hexane and methanol with an extraction time of 6 h. Drainage of the thimble was poor, which greatly decreased extraction efficiency. The third set of samples used a solvent system of methylene chloride and methanol acidified with 0.5 ml of 6 N HCl. TLC of this system was impossible because the natural water in the sediment deactivated the TLC paper.

The last two sets of samples were analyzed using soxlet extraction with pure hexane. Sample preparation consisted of homogenizing the top 5 cm of the core, removing a 10 g aliquot and mixing this with 4 g silica gel and 35 g of anhydrous sodium sulfate. These were added to dry the sample and improve solvent flow through the samples. These two extraction sets were run for several hours per day. When the system was shut down for the night, the thimbles were drained into the round bottom flask and a 1.00 g aliquot was
weighed and counted. The thimble was then filled with hexane so that the sediment could soake over night. The first set was extracted for a total of 39 h. The second set was extracted for a total of 29 h.

The residue sediment which was left over after extraction (about 12 g) was analyzed for extraction efficiency of radiolabeled compounds. The analysis consisted of taking a 0.5 g sample and combusting at 960 °C in a stream of oxygen. The gas stream was bubbled through 9 ml of PEA in order to complex the generated CO₂. The extraction column was then rinsed with an additional 1.5 ml of PEA and the total counted.

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Total Mean: \((63 \pm 36)(10)^3\)
Table D-8. Sediment extraction. Day 22.

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Table: | Day | Phase | cpm | Wt. g | Wt. g | Extr. | Total | cpm/Core |
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Mean

(35 ± 22) (10)³
Table D-9. Sediment extraction. Day 29.

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Total Mean

\[(68 \pm 35) (10)^3\]

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Total Mean

(100 ± 40) (10)³
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Total Mean: \((58 \pm 21) 
(10)^3\)
**Table D-17. Sediment extraction. Day 189.**

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Total Mean \( (70 \pm 20) \times (10)^3 \)
Appendix - E

Post-Thesis Data
Figure No. E-1. Activity measured directly from the mesocosm as well as sum of the components minus direct.

Figure No. E-2. Activity extracted from the water as well as sum of the components minus direct.
Figure No. E-3. Activity not extractable from the water as well as sum of the components minus direct.

Figure No. E-4. Radiolabeled CO$_2$ measured in the water as well as sum of the components minus direct.
Table E-1. Total water column activity.

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Note: 1. Counting efficiency after day 120 = 0.78.
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Table E-2 Continued. Water column extractable activity.

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Note: Blank = 25 cpm
Efficiency = 0.89
Table E-3. Activity in water after extraction.

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<td>249.5</td>
<td>7,000</td>
<td>16.5</td>
</tr>
<tr>
<td>256.5</td>
<td>5,460</td>
<td>12.8</td>
</tr>
<tr>
<td>263.5</td>
<td>6,340</td>
<td>14.9</td>
</tr>
<tr>
<td>270.5</td>
<td>5,670</td>
<td>13.3</td>
</tr>
<tr>
<td>273.5</td>
<td>6,290</td>
<td>14.8</td>
</tr>
</tbody>
</table>

Note: 1. Corrected for seawater blank run at same time.
2. Gas wash bottle found to be leaking - value is low.

Blank = 41 cpm
Efficiency = 0.84
Sample weight = 503 g
Appendix-F

Liquid Scintillation Counting
In liquid scintillation counting, radioactive decay is converted to light which can be detected by a photomultiplier tube. This conversion is accomplished by mixing the sample to be analyzed in a vial with a scintillation 'cocktail' consisting of a solvent, an emulsifier and a fluor. A beta particle emitted when a $^{14}\text{C}$ molecule decays interacts with some solvent molecules. The energy imparted to the solvent molecules causes them to become excited. The excitation energy is transferred to other solvent molecules causing them to become excited and is eventually passed to a fluor molecule. Unlike the solvent molecule, the excited fluor molecule dissipates the energy, not by passing it on but, by dropping back to the ground state. In the process it emits a photon. All such flashes reaching the photomultiplier tube are counted. The count divided by the time of counting gives an indication of the sample's activity. The count is not equal to the decays per minute because of inefficiencies in energy transfer from source to fluor. This inefficiency is known as quench.

A correction can be made for quench by employing a high activity gamma source ($^{137}\text{Cs}$) placed near the sample temporarily before counting. The gamma rays interact with the solvent molecules giving a characteristic spectrum. The liquid scintillation counter sorts the signals by intensity and stores the results in one of several hundred channels. When components causing quench in the sample lower the counting efficiency, they do so in a nonlinear way. This causes a shift in the channel number having the largest count accumulation. The shift in channel number is designated as the H-Number. By counting standards with known activity and a range of quenches, the relationship of counting efficiency to H-Number can be established for the solvent/sample system being used.

This was done for all sample types used in this study. Most materials were found to have counting efficiencies of about $89 \pm 2\%$ over the range of interest. For these matrices one counting efficiency was used for all H-Numbers. For some matrices a larger variation was found. A straight line fit of the quench curve (8 points) was found to be consistent with the data in light of the errors involved. These errors consist of pipeting errors (about 1.5%) and counting errors (about 2.0%). The exception to a straight line fit was found to be the relationship between H-Number
and counting efficiency for seawater samples. These samples often
turned cloudy, causing variations in H-number determination. A
much better correlation was found between counting efficiency and
sample weight. For samples with low activity it was also necessary
to correct for the natural radiation of the sample and the glass of
the scintillation vial. LSC corrections used are presented in
Table F-1.

Table F-1. Counting efficiencies and blank corrections used for
calculating decays per minute from counts per minute.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Condition</th>
<th>Weight (g)</th>
<th>H Number</th>
<th>Efficiency Percent</th>
<th>Blank cpm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hexane</td>
<td>Seawater</td>
<td>0.5 - 1.7</td>
<td>68 - 86</td>
<td>89 ± 1</td>
<td>25.5 ±</td>
</tr>
<tr>
<td>0.4</td>
<td>Sediment</td>
<td>0.4 - 0.5</td>
<td>165 - 200</td>
<td>79 ± 1</td>
<td>67 ± 1</td>
</tr>
<tr>
<td>Seawater</td>
<td>7 ml Vial</td>
<td>0.49 - 0.53</td>
<td>100 - 105</td>
<td>78 ± 3</td>
<td>35 ± 2</td>
</tr>
<tr>
<td>20 ml Vial</td>
<td>8.5 - 9.5</td>
<td>158 - 160</td>
<td>81 ± 1</td>
<td>35 ± 2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>9.5 - 10.5</td>
<td>153 - 162</td>
<td>79 ± 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>10.5 - 11.5</td>
<td>156 - 164</td>
<td>73 ± 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>11.5 - 12.5</td>
<td>168 - 181</td>
<td>71 ± 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Seawater</td>
<td>Particulate</td>
<td>-</td>
<td>110 - 135</td>
<td>84 ± 2</td>
<td>25 ± 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>-</td>
<td>135 - 165</td>
<td>80 ± 2</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>-</td>
<td>165 - 185</td>
<td>78 ± 2</td>
<td></td>
</tr>
<tr>
<td>Phenethylamine</td>
<td>Seawater</td>
<td>5.0 ml</td>
<td>140 - 160</td>
<td>84 ± 2</td>
<td>41 ± 2</td>
</tr>
<tr>
<td></td>
<td>Sediment</td>
<td>10.0 ml</td>
<td>180 - 210</td>
<td>79 ± 4</td>
<td>62 ± 2</td>
</tr>
</tbody>
</table>