



Contamination of Tributyltin Compounds on Shellfish Uses Tolerable Average Residue Levels on Pulau Pramuka Kepulauan Seribu

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Authors' contributions

This work was carried out in collaboration among all authors. Author SL designed the study, performed the study, wrote the protocol and wrote the first draft of the manuscript. Authors Zahidah, AS and HH managed the analyses of the study and managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Aims: This research is aimed to know the contamination of tributyltin (TBT) compounds on shellfish uses TARL. To determine the level of contamination in seafood, shellfish use the weight of an average person.

Place and Duration of Study: Ecology Laboratory and Central Laboratory, Padjadjaran University from January until April 2019.

Methodology: The research was conducted purposive sampling consisted of 3 different location with a total 98 shell. Tributyltin analysis is to determine the level of tributyltin contamination in shellfish which can affect an individual in consuming tributyltin-contaminated shells. The results were statistically analyzed TARL carried out and analyzed descriptively.

Results: 8 species of shellfish have been identified, *Tellina virgata*, *Perna viridis*, *Anadara granosa*, *Anadara antiquata*, *Fragum unedo*, *Fimbria fimbriata*, *Gafrarium tumidum* and *Tridacna squamosa*. The number of bivalves found was different at each station. The total number of shells

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identified in all 98 individuals from the three stations, *Anadara granosa* species is the dominant species found, this is because *Anadara granosa* shells can live in different types of habitats and their existence tends to dominate the open, coastal and estuary waters. The results of tributyltin analysis on shellfish showed TBT contamination in shellfish, each research location found different concentrations of tributyltin. Muara Angke pier was found to be the highest TBT concentration in meat in *Perna viridis* at $0.170 \pm 0.0192 \text{ ng.g}^{-1}$. From available TBT analysis data, the estimated daily TBT intake to the average person who likes seafood on Pramuka Island with a bodyweight of 60 kg is $0.54\text{-}10.2 \text{ ng TBT.person}^{-1}.\text{day}^{-1}$ through shellfish consumption. Although this value is still far from the threshold of a tolerable value of $15 \text{ g.person}^{-1}.\text{day}^{-1}$. The estimated daily intake of TBT through seafood products in Indonesia is among the lowest compared to developed countries, such as Japan ($3000\text{-}100000 \text{ g.person}^{-1}.\text{day}^{-1}$), Canada ($<610\text{-}15000 \text{ g.person}^{-1}.\text{day}^{-1}$), USA ($4000\text{-}45000 \text{ g.person}^{-1}.\text{day}^{-1}$), Finland ($970\text{-}9700 \text{ g.person}^{-1}.\text{day}^{-1}$) and some Asian countries such as Thailand ($228\text{-}45714$ through shellfish consumption and the Philippines ($2361\text{-}68312$ through shellfish). Several things that affect daily consumption per capita in determining TART are based on average values. This implies, that some populations consume more seafood products (fishermen, people who have a preference for consuming fish) than the average so that there is a greater risk of seafood that has been contaminated with tributyltin. Then TART is based on the average person weighing 60 kg. A person with a lighter weight will receive relatively more compound tributyltin per kg of body weight.

Conclusion: Based on the results of the research is the contamination of tributyltin compounds in shellfish seafood by 0.170 ng.g^{-1} with an estimated daily intake of TBT to the average person who likes seafood on Pramuka Island with a bodyweight of 60 kg is $0.54\text{-}10.2 \text{ ng TBT.person}^{-1}.\text{day}^{-1}$ through shellfish consumption.

Keywords: Shellfish; contamination; tributyltin; TART.

1. INTRODUCTION

Shipping activities at the port is one of the causes of marine pollution. Ships use the tributyltin (TBT) compound as an antifouling agent on the hull [1]. Tributyltin is usually used as an antifouling paint material on the hull which aims to prevent damage caused by attaching biota namely barnacles, algae and molluscs that live on the underside of the ship [2]. Although TBT can prevent or slow down damage to the hull, the effect of the use of TBT is very dangerous to the condition of the waters, especially on the biota that is included, including non-target biota in these waters [3]. One of the marine biota affected by pollution is molluscs.

TBT is mostly found in seafood. In the EU Scientific Cooperation Project (SCOOP) entitled "Assessment of dietary exposure to organotin compounds of the population of EU member states", data on the concentration of organotin compounds, including TBT were collected from 8 European countries (Belgium, France, Greece, Germany, Italy, Norway, The Netherlands, and Denmark) covering the period 1995 and 2002 [4,5]. Germany delivered the great majority of data (approximately 86%). The calculated concentrations of TBT in seafood other than fish (including molluscs, crustaceans, echinoderms,

and cephalopods) were in general higher (14 and $60 \mu\text{g/kg}$ wet weight (WW), median and mean, respectively) than the concentration in fish (5 and $17 \mu\text{g/kg}$ WW, median and mean, respectively). Based on fully aggregated data for both fish and seafood other than fish, the calculated concentration median and mean of TBT were 7.0 and $28 \mu\text{g/kg}$ WW, respectively [6].

Shellfish is a colloquial and fisheries term for exoskeleton-bearing aquatic invertebrates used as food, including various species of molluscs, crustaceans and echinoderms. Shellfish aquaculture is a rapidly growing enterprise that helps provide a stable source of shellfish while not harming wild populations. Ecologically, not only are shellfish vital in marine food chains, but some filter-feeding forms can purify a great amount of water and thus are crucial to the stability of marine systems. And it was shellfish that were the "canaries of the sea" and alerted people to the danger of the disposal of human sewage and other pollution as outbreaks of hepatitis and typhoid fever was traced to consumption of shellfish from contaminated waters [7]. Seafood consumption is considered to pose a health risk if it exceeds the maximum tolerable daily intake (TDI = Tolerable Daily Intake), the TDI for TBT has been set at $0.25 \mu\text{g}$ (kg x day weight) for TBT. The maximum

TDI for TBT is 15 µg per day with a bodyweight of 60 kg.

2. MATERIALS AND METHODS

2.1 Time and Place of Research

The research was conducted from January 2019 until April 2019, and have three stations. The station I is located on the Muara Angke pier, Jakarta, Station II is located on the Pramuka Island pier and Station III is located in the mangrove and seagrass area of Pramuka Island, the Thousand Islands National Park. Determination of research stations using a purposive sampling method, namely determining the criteria for each station taken based on the concentration of tributyltin pollutants by vessel activity. And tributyltin level was analysis in ecology laboratory and central laboratory, analysis section 3, Padjadjaran University.

2.2 Materials and Tools

The tools used in this research are Ekman grab, quadrant transect, ziplock, GPS, cool box, dissecting kit, and HPLC. The material used was 98 shellfish samples, 4% formalin and chemicals to analyze tributyltin.

2.3 Procedures

2.3.1 Prepare shellfish samples

Catch shellfish for sample using Ekman grab method, total from three stations is 98 shellfish.

2.3.2 Tributyltin analysis

Analysis of TBT concentrations in shellfish samples by using the HPLC (High-Performance Liquid Chromatography) method [8]. The tributyltin analysis procedure consists of three parts as follows:

A. Tributyltin standard preparation procedure

The standard solution of tributyltin is taken 5 mL into a 100 mL measuring flask (intermediate solution 1), then dissolved with methanol up to 100 mL in volume. Then the intermediate solution 1 pipette 25 mL and put in a 100 mL measuring flask, then dissolved with methanol to a volume of 100 mL (intermediate solution 2). Intermediate solution 2 pipetted into 7 50 mL

measuring flasks with a volume of 0 ml; 0.25 mL; 0.5 mL; 1mL; 1.5 mL; 6.1 mL and 12.2 mL and mix with methanol until the volume is 100 mL. Standard series solutions are pipetted as much as 1 mL of each concentration and put into a 1.5 mL microtube. The seven standard series were then centrifuged for 10 minutes at a speed of 10000 rpm The supernatant was piped and filtered with a syringe filter so that a standard solution free of particles was put into an HPLC vial bottle. Concentration of 7 tributyltin standard series tested in HPLC 0; 0.0204; 0.0408; 0.081; 0.4974 and 0.9947 ng.mL⁻¹.

B. Sample extraction (dry) procedure

Samples of shellfish are mashed using a mortar (viscous), then put into a 10 mL measuring flask and methanol added to the boundary markers. Samples of meat that have been dissolved in methanol then added 1 ml of dichloromethane and 1 mL of ascorbic acid. Then the solution is homogenized with a sonicator. Pipette the sample that has been homogeneous as much as 1 mL and put in a 1.5 mL microtube, then centrifuged for 10 minutes at a speed of 10000 rpm. The supernatant is pipetted and filtered with a syringe filter put into an HPLC vial bottle. The extraction results are then analyzed using the HPLC tool.

2.3.3 TARL (Tolerable Average Residue Level)

TARL analysis in seafood products aims to determine the average tolerance in consuming shellfish with an average body weight of 60 kg. The TARL calculation is based on the following equation:

$$\text{TARL} = (\text{TDI} \times 60 \text{ kg body weight}) / \text{average daily consumption of seafood}$$

The TDI value, in this case, is for TBT set at 0.25 µg.kg⁻¹ body weight per day, this is based on the TBT toxicity data from observations that have an LC₅₀ (Lethal concentration) effect on *Mytilus edules* of 2.5 ppb [5].

2.4 Data Analysis

Data obtained from observations of Tributyltin analysis and TARL were analyzed descriptively by comparing parameters with the literature so that it can be said that the shellfish studied has tributyltin compound levels that are fit for human consumption.

3. RESULTS AND DISCUSSION

3.1 Tributyltin in Shellfish

The results of tributyltin analysis on shellfish showed TBT contamination in shellfish, each research location found different concentrations of tributyltin. Muara Angke pier was found to be the highest TBT concentration in meat in *Perna viridis* at $0.170 \pm 0.0192 \text{ ng.g}^{-1}$. From available TBT analysis data, the estimated daily TBT intake to the average person who likes seafood on Pramuka Island with a bodyweight of 60 kg is $0.54\text{-}10.2 \text{ ng TBT.person}^{-1} \cdot \text{day}^{-1}$ through shellfish consumption. Although this value is still far from the threshold of a tolerable value of $15 \text{ g.person}^{-1} \cdot \text{day}^{-1}$.

The estimated daily intake of TBT through seafood products in Indonesia is among the lowest compared to developed countries, such as Japan ($3000\text{-}100000 \text{ g.person}^{-1} \cdot \text{day}^{-1}$), Canada ($<610\text{-}15000 \text{ g.person}^{-1} \cdot \text{day}^{-1}$), USA ($4000\text{-}45000 \text{ g.person}^{-1} \cdot \text{day}^{-1}$), Finland ($970\text{-}9700 \text{ g.person}^{-1} \cdot \text{day}^{-1}$) and some Asian countries such as Thailand ($228\text{-}45714$ through shellfish consumption) [5] and the Philippines ($2361\text{-}68312$ through shellfish) [9].

Several things that affect daily consumption per capita in determining TARL are based on average values. This implies, that some populations consume more seafood products

(fishermen, people who have a preference for consuming fish) than the average so that there is a greater risk of seafood that has been contaminated with tributyltin [5]. Then TARL is based on the average person weighing 60 kg. A person with a lighter weight will receive relatively more compound tributyltin per kg of body weight.

3.2 Side Effect Contamination Tributyltin on Shellfish

Stacks of tributyltin in the body of a clam might not be detected by ordinary chromatography because the content is very low in quantity. However, given the destructive power of tributyltin, which is long-term, because the danger is not felt directly, such as vomiting, dizziness, or diarrhea so that people who are contaminated with tributyltin due to consuming marine life do not feel the presence of the danger [5]. Therefore, people who are tainted with tributyltin will not immediately stop eating seafood that contains tributyltin. Finally, there was an accumulation of tributyltin that threatened the health of seafood consumers [10,11]. Second, the danger is chronic. That is, the danger arises when the accumulation lasts a long time and has reached the limit can cause diseases such as cancer or damage to reproductive organs [12].

Table 1. Tributyltin concentration in shellfish in each research station

Research location	n	Shell length (mm)	TBT (ng.g ⁻¹)	Stdev
Muara Angke pier				
<i>Perna viridis</i>	15	6,3 - 7,8	0,170	0,0192
<i>Anadara granosa</i>	15	5,5 - 7,0	0,127	0,0226
Pulau Pramuka pier				
<i>Tellina virgata</i>	8	5,0 - 7,0	0,136	0,0036
<i>Anadara antiquata</i>	4	6,5 - 7,5	0,109	0,0009
<i>Fragum unedo</i>	2	5,5 - 6,0	0,009	0,0004
<i>Fimbria fimbriata</i>	5	5,5 - 6,3	0,008	0,0008
<i>Perna viridis</i>	5	6,0 - 7,0	0,113	0,0037
<i>Anadara granosa</i>	11	5,5 - 6,5	0,083	0,03
Mangrove Pulau Pramuka				
<i>Tellina virgata</i>	16	5,0 - 7,0	0,103	0,0034
<i>Anadara antiquata</i>	3	6,0 - 7,0	0,132	0,0098
<i>Fragum unedo</i>	2	5,0 - 6,5	0,01	0
<i>Fimbria fimbriata</i>	2	5,5 - 6,3	0,064	0,0014
<i>Gafrarium tumidum</i>	1	5,0 - 6,2	0,009	0,0006
<i>Anadara granosa</i>	6	5,0 - 6,5	0,009	0,0006

*n, numbers of sample; *highest TBT concentration of the sample

Many studies have been carried out showing that tributyltin is a DNA destroyer and an endocrine disruptor [13]. *In vitro* and using the Single Cell Gel Electrophoresis Assay technique, according to Alzieu that at levels of $10 \mu\text{g.L}^{-1}$, tributyltin is strong enough to damage the DNA of blue shells. This indicates that tributyltin is genotoxic (DNA damaging substances) that can cause cancer [9]. Third, the danger of damage caused by tributyltin is transgenerational. This means, due to pollution that is not only suffered by people who consume contaminated shellfish or marine life but also suffered by their offspring because the negative impact of tributyltin is passed down from generation to generation.

4. CONCLUSION

Based on the results of the research is the contamination of tributyltin compounds in shellfish seafood by 0.170 ng.g^{-1} with an estimated daily intake of TBT to the average person who likes seafood on Pramuka Island with a body weight of 60 kg is $0.54\text{-}10.2 \text{ ng TBT .person}^{-1}.\text{day}^{-1}$ through shellfish consumption.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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