The organotin contaminants in food: Sources and methods for detection: A systematic review and meta-analysis

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ABSTRACT

Organotin compounds in low doses have toxic effects. These components may contaminate food. The aim of this systematic review was to determine the type and level of organotin in food that are mainly contaminated with these compounds, as well as common detection methods. The research studies with keywords Organotin, Tributyltin, TBT, Food, Detection, Contamination, and Pollution were searched in PubMed, Scopus, Science Direct, and Google Scholar databases, regardless of publication time. Two author independently investigated the publications. A number of 123 studies were obtained and only 9 articles were finally selected according to exclusion and inclusion criteria. Studies were selected which organotin components were detected in the food matrix. The important data were extracted. Meta-analysis was calculated for the amount of TBT in seafood. The most important of these compounds are TBT, TPT, Dibutyltin (DBT) and di-n-octyltin (DOT). Surveys were conducted on three continents, Europe, America and Asia. Contaminated foods reportedly included seafood and edible oils, according to studies. TBT was investigated more than other tin components in food. The overall average estimate for TBT in seafood was estimated at 182.33 ng/g that This amount was more than maximum limit. Therefore, it is necessary to take measures to treat the wastewater so that these harmful compounds do not reach the water of sea.

Introduction

Organotin has a variety of uses, including stabilizers in plastics, antifoam in paints, wood preservative and pesticide (Chen et al. 2019, He et al. 2020). 40% of organotin compounds are used in plastics (Liu & Jiang, 2002). These components are recognized as organometallic components (Zhu et al. 2013). The organotin have been identified in human blood and liver samples (Forsyth & Casey, 2003). European food safety authority (EFSA) set a tolerable daily intake (TDI) of 0.25 µg/kg for the four organotin. These four compounds include Tributyltin(TBT), Triphenyltin (TPHT), DBT and di-n-octyltin (DOT) (Chung et al. 2020). The origin of these compounds is mainly due to human activities (Sousa et al. 2009, Zabaljauregui et al. 2007). TBT and TPHT have the most toxic effects on the endocrine glands and are banned in many countries (Yang et al. 2010). EU and US regulations prohibit the use of TBT and TPHT in food contact materials (He et al. 2020). TPHT is used as a fungicide in agriculture (Forsyth & Casey, 2003). Dibutyltins (DBTs) and TBT are neurotoxic and damage the bile duct (Amodio-Cocchieri et al. 2000). Tributyltin has more toxic effects (Amodio-Cocchieri et al. 2000, Vacchini et al. 2020). Tributyltins have toxic effects on aquatic populations even at very low doses (Amodio-Cocchieri et al. 2000). This kind of organotin has genotoxic effects (Santos et al. 2009). They lead to deformity of crab limb and death of mussel larvae (Forsyth & Casey, 2003). TBT oxide also induce mutations and have teratogenic (Amodio-Cocchieri et al. 2000). TBT has been identified in humans as an immunotoxic compound, and the tolerance daily intake was calculated 0.25 mg/kg body weight/ day based on immunological studies (Forsyth & Casey, 2003). The tolerance daily intake is 250 ng/kg per body weight for total four organotin TBT, TPT, DOT, and DBT (Rantakokko et al. 2006). These compounds are also persistence in the environment (Sousa et al. 2009). These compounds are also found in house dust. According to studies conducted in house dust, monobutyltin (MBT), another
organotin is found more than other compounds in house dust (Kannan et al. 2010). This dust may be swallowed by children, so they will be more exposed to tin compounds than adults (Kannan et al. 2010). Furthermore, in the blood test of working women of reproductive age, MBT level was higher than other organotin (Program 2016). This type of organotin is used as a dose reference (Sousa et al. 2017).

The organotin in food are absorbed by cells in the gastrointestinal tract and enter the bloodstream (Sant Anna et al. 2012). In order to know that the contaminants, toxins and chemicals in food are in the tolerance daily intake (TDI) range, it is necessary to know the level of these substances in food. Therefore, the objective of this systematic review was to determine the amount and type organotin component in food.

**Methods**

This systematic review was written on PRISMA checklist. Two authors performed all stage including inclusion and exclusion criteria, and data extraction to prevent bias.

**Search strategy**

The articles in English language were searched on 16 June 2021. There was no time limitation. The chosen databases were PubMed, Science direct, Google scholar, and Scopus. The keywords for searching were set: (Organotin OR Tributyltin OR TBT) AND (Food) AND (Detection) AND (Contamination OR Pollution). A total number of 123 articles were identified from databases. At first, the title and abstract of the manuscript were reviewed. Manuscript that did not meet the inclusion criteria were excluded from the study. The full text were assessed with two author (P.S and Z.N).Then, the full text of the selected Manuscript was carefully studied and the data was extracted according to the protocol.

**Table 1**

| Analysis method/ unit of detection | Type of food /sample size | Type of organotin | Country | Authors / Year |
|-----------------------------------|---------------------------|-------------------|---------|----------------|
| GC-PFPD µg/g                      | 0.008 to 0.135            | Bivalves          | Japan   | Inoue/2006     |
|                                   |                           | Tributyltin(TBT)  |         | (Inoue et al. 2006) |
| GC-PFPD µg/Kg                     |                           | Farm fish – 66    | Italy   | Amrdio-Cocchieri/2000 |
|                                   |                           | TBT               |         | (Amodio-Cocchieri et al. 2000) |
| GC-PFPD ngSn/g                    | 602.3 ± 14.5              | an edible gastropod | Chile  | Mattos/2017 |
|                                   | 368.7 ± 5.5               | TBT               |         | (Mattos et al. 2017) |
|                                   | 203.7 ± 4.0               | DBT               |         | (Gui-bin &Qun-fang 2000) |
| GC-MS µg/g                        | 13.86 ± 0.31              | lard samples      | Chine   | Gui-bin/2000   |
|                                   | 1700 ± 40.0               | sample size – Not |         | (Gui-bin &Qun-fang 2000) |
|                                   | 225.06 ± 7.5              | mentioned         |         |                |
|                                   |                           | TBT               |         |                |
|                                   |                           | MBT               |         |                |
| GC-ICP/MS µg/Kg                   | 0.32                      | Fish              | Hong    | Chung/2020     |
|                                   | 0.04                      | TBT               |         | (Chung et al. 2020) |
|                                   | 0.01                      | DBT               |         |                |
|                                   | 0.19                      | DBT               |         |                |
|                                   | 0.31                      | DOT               |         |                |
|                                   | 15                        | TPhT              |         |                |
| GC-MS ngSn/g                      | 24.2 ± 1.3                | commercial oysters | Chine   | Chen/2019     |
|                                   | 46.0 ± 0.7                | N = 20 for six seafood markets |         | (Chen et al. 2019) |
|                                   | 68.1 ± 20.1               | MBT, DBT, TBT, MPhT , |         |                |
|                                   | 589 ± 25.6                | DPhT              |         |                |
|                                   | 992 ± 18.9                | TPHT              |         | TMM, DMT, TBT, MPhT , |
|                                   | 747 ± 7.3                 |                   |         |                |
| HPLC-MS/MS µg/kg                  |                           | edible vegetable oil | China   | Liu/2016(2016) |
|                                   |                           | N = 20 for each oil size |         | (Liu et al. 2016) |
|                                   |                           | TBT               |         |                |
|                                   |                           | DBT               |         |                |
|                                   |                           | DOT               |         |                |
|                                   |                           | TPhT              |         |                |
| GC-PFPD µg/g                      |                           | DBT was measured in all oil samples except sesame and rape oil. | Spain  | B o r g h i/2002 |
|                                   |                           | All organotin compounds were ND in all fish species except two: |         |                |
|                                   |                           | 1)TPT in Mediterranean codling 3.5 ± 0.8 |         |                |
|                                   |                           | 2)DBT in Gunther grenadier 4.9 ± 4.3 |         |                |
|                                   |                           | MBT, DBT, TBT, DPhT and TPHT |         |                |
|                                   |                           | Common mora Mediterranean codling |         |                |
|                                   |                           | Gunther grenadier |         |                |
|                                   |                           | Riso smooth-head Spiderfish |         |                |
|                                   |                           | N = 3 for each of species |         |                |
|                                   |                           | fish and shellfish |         |                |
|                                   |                           | N = 5 |         |                |
|                                   |                           | DPhT              |         |                |
|                                   |                           | DBT, TBT, DPhT, TPT |         |                |
|                                   |                           | Japan             |         | (Tsunoda 1993) |

| Type of organotin Country Authors / Year |
|-----------------------------------------|-----------------------------|
| Tributyltin(TM) Japan Inoue/2006         |
| TBT Japan Tsunoda/1993                   |
| TBT Italy Amrodio-Cocchieri/2000         |
| TBT Chile Mattos/2017                    |
| DBT Chile Mattos et al. 2017             |
| TPhT Chine Gui-bin/2000                  |
| TBT Hong Chung/2020                     |
| TBT Kong Chung et al. 2020              |
| TBT Chine Chen/2019                     |
| TBT China Liu/2016                      |
| DBT Spain B o r g h i/2002               |
| MBT, DPhT, TBT, MPhT Japan Tsunoda/1993 |
Inclusion and exclusion criteria

The two reviewers (P.S and M.J) searched the keywords in databases independently. Invitro and animal study, environmental sample, review and chapter of book, non-English article, biomonitoring, and application of organotin were excluded. Inclusion criteria for this systematic review included original articles that measured organotin levels by valid methods of measuring. All publication that was according to inclusion criteria was assessed. The sample size of a significant number of studies was only one, so it was excluded from this systematic review.

Data extraction

The name of the first author, time of study, country, type of food and organotin, amount and sample size, method of measuring in samples were extracted in the Table 1. The data extracted by two reviews (P.S and M.J) indecently. In all steps, disagreement were consulted with third author. If the full text of the articles was not available, the authors of the article were emailed.

Estimation of the mean of tributyltin in seafood

Among the organotin, tributyltins was reported more than others, so this compound was selected for meta-analysis. The levels of tributyltins in seafood were converted to ng/g units. For this estimation, studies were selected that had mean, standard deviation and number of samples. The total mean was estimated with comprehensive meta-analysis software.

Results

The search processes

123 articles were achieved by searching in PubMed, Scopus, Science

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Fig. 1. The diagram of study.
direct, and Google Scholar database. 31 articles were excluded from the study due to duplicating. The title and abstracts of the remaining articles were carefully studied and 45 articles were excluded because of using review and chapter of book, animal studies, other sample including environmental sample, application and non-English article. Then, the full texts of 47 papers were taken. The quality of the studies was also assessed by two persons. At end step, 9 articles were chosen. The PRISMA checklist was conducted for this systematic review. Fig. 1 shows PRISMA diagram of database searches.

The descriptive results of screened manuscript

Of all screened manuscript, 9 were selected for the systematic review. The type of food and type of organotin identified and the method used to identify organotin are shown in Table 1. Important results of each study are listed in the discussion section.

Estimation of the mean of tributyltin in seafood

Four of the studies in the table had the mean, standard division and sample of size for TBT in seafood. Therefore, 4 publications were selected in the present study for meta-analysis. The overall mean was estimated with the random model. This was estimated at 182.33 ± 84.62 ng/g.

Discussion

In this study, tin compounds were observed in seafood and liquid oils. The extracted data shows that most of the samples examined are foods of marine origin. According to previous studies, tin compounds are a global threat to marine ecosystems (Hu et al. 2006). TBT, DBT and MBT foods of marine origin. According to previous studies, tin compounds are estimated with the random model. This was estimated at 182.33 ng/g.

The analysis methods included 8 gas chromatographs and one liquid chromatography (Table 1). According to previous studies, gas chromatography is commonly used (Inoue et al. 2006). In both studies, organotin compounds in food were investigated. Exposure to tin compounds is not limited to food. Occupational exposure also occurs (Hoppe 2002, Ichihara et al. 2018). Therefore, determining the amount of these compounds in the blood of individuals and biomonitoring is recommended for future studies.

Conclusion

In this systematic review, the type of organotin compounds in food was identified. Most research has been found in countries bordering the seas. Seafood showed higher levels of these compounds. so, we need more assess of seafood. According to the polished manuscript, TBT was reported more frequently than other organotin but the amount of phenyltins were higher than other compounds. The analytical methods were GC-FPD. According to the estimate of the overall mean TBT, the amount of this type of organotin was higher than the allowable level announced by the European authorities. Due to the fact that water can be contaminated with these compounds through municipal waste, it is necessary to treat them well. Most of the reported samples in food are seafood, so sea pollution should be given more importance. One of the limitations of this systematic review study was limited to Europe, south of America and Asia and did not cover other continents. Due to the fact that these compounds are also used in pesticide, so extensive studies such as seafood have not been performed in plant products.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

Amadio-Cocchieri, R., Cirillo, T., Amorena, M., Cavaliere, M., Luciano, A., & Del Prete, U. (2000). Alkyltins in farmed fish and shellfish. International journal of food sciences and nutrition, 51, 147-151.

Authority EFSA. (2004). Opinion of the Scientific Panel on contaminants in the food chain [CONTAM] to assess the health risks to consumers associated with exposure to organotin in foodstuff. EFSA Journal, 2, 102.
Choi, M., Moon, H.-B., & Choi, H.-G. (2012). Intake and potential health risk of butyltin compounds from seafood consumption in Korea. Archives of environmental contamination and toxicology, 62(2), 333–340.

S.W.C. Chang J.S.Y. Lau J.P.K. Lau Occurrence of organotin compounds in seafood from Hong Kong market 154 2020 111 10.1016/j.marpolbul.2020.111116.

Forsty, D. S., Weber, D., & Cleroux, C. (1992). Determination of butyltin, cyclohexyltin and phenyltin compounds in beers and wines. Food additives and contaminants, 9(2), 161–169.

Forsty, D. S., & Casey, V. (2003). Butyltin compounds in retail mollusc products. Food additives and contaminants, 20(5), 445–452.

Guerin, T., Sirot, V., Volatier, J.-L., & Leblanc, J.-C. (2007). Organotin levels in seafood and its implications for health risk in high-seafood consumers. Science of The Total Environment, 388(1-3), 66–77.

Gui-bin, J., & Qin-fang, Z. (2000). Direct Grignard pentylation of organotin-contaminated lard samples followed by capillary gas chromatography with flame photometric detection. Journal of Chromatography A, 886(1-2), 197–205.

Hu, H., Tian, M., Hu, L., & Yang, L. (2020). Ultra sensitive determination of organotin compounds in plastic food packaging and edible oils by sheetless capillary electrophoresis-electrospray ionization-mass spectrometry. The Analyst, 145(6), 2286–2296.

Hoppe HW (2002): Butyl tin compounds (mono-, di-, tri- and tetrabutyltin) [Biomonitoring Methods, 2010]. The MAK-Collection for Occupational Health and Safety: Annual Thresholds and Classifications for the Workplace, 121-143.

Hu, J., Zhou, H., Wan, Y., Gao, J., An, W., An, L., & Jin, X. (2006). Trophic magnification of triphenyltin in a marine food web of Bohai Bay, North China: Comparison to tributyltin. Environmental science & technology, 40, 3142–3147.

Ichihara, G., Isida, M., Fujie, T., Kaji, T., & Kim, Y. (2018). 165 Urinary trimethyltin reflects blood trimethyltin in scrap recycling workers. BMJ Publishing Group Ltd.

Ikonomou, M. G., Fernandez, M. P., He, T., & Cullen, D. (2002). Gas chromatography-high-resolution mass spectrometry method for the simultaneous determination of nine organotin compounds in water, sediment and tissue. Journal of Chromatography A, 975(2), 319–333.

Inoue, S., Abe, S.-I., Oshima, Y., Kai, N., & Honjo, T. (2006). Tributyltin contamination of bivalves in coastal areas around northern Kyushu, Japan. Environmetal toxicology, 21(3), 244–249.

K. Kannan S. Tanabe H. Iwata R. Tatukawa 90 3 1995 279 290.

Kannan, K., Takahashi, S., Fujiwara, N., Mizukawa, H., & Tanabe, S. (2010). Organotin compounds, including butyltin and octyltins, in house dust from Albany, New York, USA. Archives of environmental contamination and toxicology, 58(4), 901–907.

Liu, J.-Y., & Jiang, G.-B. (2002). Survey on the presence of butyltin compounds in Chinese alcoholic beverages, determined by using headspace solid-phase microextraction coupled with gas chromatography–flame photometric detection. Journal of agricultural and food chemistry, 50(23), 6683–6687.

Liu, Y., Ma, Y., Wan, Y., Guo, L., & Wan, X. (2016). Fast and effective low-temperature freezing extraction technique to determine organotin compounds in edible vegetable oil. Journal of Separation Science, 39(12), 2380–2387.

Mattos, Y., Stotz, W. B., Romero, M. S., Bravo, M., Fillmann, G., & Castro, L. B. (2017). Butyltin contamination in Northern Chilean coast: Is there a potential risk for consumers? The Science of the total environment, 595, 209–217.

Morbrito, B., Massanit, P., & Quevauviller, P. (2000). Derivatization methods for the determination of organotin compounds in environmental samples. TrAC Trends in Analytical Chemistry, 19, 113–119.

Mozughi, N., Lepes, G., Bravo, M., Dachraoui, M., & Potin-Gautier, M. (2005). Organotin speciation in Bizerte lagoon (Tunisia). Science Of The Total Environment, 349(1-3), 211–222.

Program NT (2016): NTP research report on organotin and total tin levels in Danish women of reproductive age.

Rantalakko, P., Kuningas, T., Saastamoine, K., & Vartiainen, T. (2006). Dietary intake of organotin compounds in Finland: A market-basket study. Food additives and contaminants, 23(8), 749–756.

Sant-Anna, B. S., Santos, D. M. D., Sandron, D. C., Souza, S. C.D., de Marchi, M. R. R., Zara, F. J., & Turra, A. (2012). Hermits crabs as biocenodons of recent tributyltin (TBT) contamination. Ecological Indicators, 14(1), 184–188.

Santos, M. M., Enes, P., Reis-Henriques, M. A., Kuballa, J., Castro, L. F. C., & Vieira, M. N. (2009). Organotin levels in seafood from Portuguese markets and the risk for consumers. Chemosphere, 75(5), 661–666.

Souza, A., Ikemoto, T., Takahashi, S., Barroso, C., & Tanabe, S. (2009). Distribution of synthetic organotin and total tin levels in Mytilus galloprovincialis along the Portuguese coast. Marine pollution bulletin, 58(8), 1130–1136.

Souza, A. C. A., Coelho, S. D., Pastorinho, R. M., Taborda-Barata, L., Nogueira, A. J. A., Isobe, T., … Tanabe, S. (2017). Levels of TBT and other selected organotin compounds in duplicate diet samples. Science of The Total Environment, 579, 19–23.

SUSNODA, MASASHI (1993). Simultaneous determination of organotin compounds in water, sediment and tissue. Journal of Chromatography B, 615(2), 340–349.

Zabaljauregui, M., Delgado, A., Usobiaga, A., Zuloaga, O., de Diego, A., & Santos, M. M., Enes, P., Reis-Henriques, M. A., Kuballa, J., Castro, L. F. C., & Vieira, M. N. (2009). Organotin levels in seafood from Portuguese markets and the risk for consumers. Chemosphere, 75(5), 661–666.

Takahashi, S., Tanabe, S., & Matsumoto, K. (1988). Determination of organotin compounds by anodic stripping voltammetry. Analyst, 113(7), 659–662.

Tohoku Journal of experimental medicine, 169(2), 167–178.

Vacchina, V., Epova, E. N., Beraill, S., Medina, B., Donard, O. F. X., & Seby, F. (2020). Tin and mercury and their speciation (organotin compounds and methylmercury) in worldwide red wine samples determined by ICP-MS and GC-ICP-MS. Food additives & contaminants. Part B, Surveillance, 13(2), 88-98.

Yang, G., Xu, J., Xu, L., Chen, G., & Fu, P. (2010). Analysis of ultratrace triorganotin compounds in aquatic organisms by using capillary electrophoresis-inductively coupled plasma mass spectrometry. Talanta, 80(5), 1913–1918.

Zabala-Arguelles, M., Delgado, A., Usobiaga, A., Zuloaga, O., de Diego, A., & Madariaga, J. M. (2007). Fast method for routine simultaneous analysis of methylmercury and butyltins in seafood. Journal of Chromatography A, 1148(1), 78–85.

Zhou, Q.-F., Jiang, G.-B., & Liu, J.-Y. (2002). Organotin pollution in China. TheScientificWorldJournal, 2, 655–659.

Zhu, S., Hu, F., Yang, T., Gan, N., Pan, D., Cao, Y., & Wu, D. (2013). Synthesis and characterization of a molecularly imprinted polymer for the determination of trace tributyltin in seawater and seafood by liquid chromatography-tandem mass spectrometry. Journal of Chromatography B: Analytical Technologies in the Biomedical and Life Sciences, 921–922, 21–26.