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Daily intake of TBT, Cu, Zn, Cd and As for fishermen in Taiwan

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Abstract

The consumption of contaminated seafood has been reported as an important route of human exposure to metals in Taiwan. We consider the concentrations of TBT, Cu, Zn, Cd, As, and the consumption of oysters of Taiwanese to be the important information related to public health in Taiwan. Therefore, the aim of this study was to evaluate the public health risks associated with TBT, Cu, Zn, Cd and As from shellfish for the general population and fishermen of Taiwan. In general, TBT concentrations in various oysters ranging from 0.32 to 1.51 $\mu\text{g/g}$ dry wt. varied with sampling locations. The highest TBT, Cu, and Zn geometric mean (GM) concentrations in oysters of 1.51, 1180 and 1567 $\mu\text{g/g}$ dry wt. were obtained from the Hsiangshan coastal area. The values of oyster consumption for fishermen were 94.1 and 250 g/day for typically and maximally exposed individuals, respectively. In particular, the highest intake (250 g/day) from fishermen was almost two times greater than that of the general population (139 g/day). The THQ (target hazard quotient) values of Hsiangshan's fishermen are 3.87 and 20.50 for TBT and Cu for maximally exposed individuals are higher than other oyster culture areas. It is interesting that those consuming oysters from Hsiangshan, Lukang, Taishi caused abnormally high THQs of TBT and other metals (100% over 1.0), and TBT was attributed to only 3–21% of the total THQs in different fishermen of Taiwan. Our results suggest that current environmental levels of TBT and other metals are associated with a significant potential threat to human health for fishermen resident in coastal areas of Taiwan. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: TBT; Metals; Intake; Fishermen

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1. Introduction

Seafood is the primary source of protein and an important part of diet for people around the world. People can be exposed to toxic chemicals that accumulate in contaminated seafood which they consumed (Han et al., 1994; Svensson et al., 1995). The average marine fish and shellfish consumption of Taiwan for 1996 was 104 kg/person/year; approximately 4.5-fold greater than the world average (23 kg/person/year) (WWF, 2000). Recently, several investigations have focused attention on human exposure to chemicals resulting from the consumption of contaminated seafood, especially for fishermen and heavy fish consumers. For example, the estimated daily intake values of dioxins for heavy fish consumers is two-fold higher than the general population in Japan (Yoshida et al., 2000). During 1991–1998, the potential carcinogenic (organochlorine pesticides and inorganic As) and non-carcinogenic (Cu, Zn and Cd) risk to the public from ingestion of the oysters was evaluated in Taiwan. It was found that the most current health risk was associated with the chemical contamination of fish and shellfish (Han et al., 1998, 2000). In Taiwan, as industries and sea journeys grew year by year, the continued presence of chemicals in the coastal ecosystem poses an ecotoxicological threat.

Recently, organotins have been widely used as biocidal ingredients in anti-fouling paints applied to ships. There are two main ship-building companies and many ship repairing and remodeling companies as well as many ships passing through the Taiwan Strait. Unfortunately, organotin pollution is widespread in the coastal waters of Taiwan as dibutyltin (DBT), tributyltins (TBT) and triphenyltins (TPT) (Hung et al., 1998, 2001). It is assumed that TBT is the toxic species. The TBT cation leaches from the anti-fouling paint matrix into seawater (Bryan et al., 1986). For example, organotin compound concentrations in the coastal waters of Taiwan ranged from not detectable to 77 ng/l (Chen et al., 1992; Chu, 1995). Imposex has been observed in rock shells (*Thais clavigera*) (range from 48 to 100%) from the different coastal areas of Taiwan (Liu et al., 1997). In particular,

high percentages of imposex of rock shells has been showed at Shiangshan, Lukang and Chiku coastal areas which were increased from 67.1%, 59.3% and 36.7% in summer to 100%, 100% and 80% in winter, respectively (Hung et al., 2001). In other countries, the widespread occurrence of imposex in Japanese gastropods has been observed as organotin pollution (Horiguchi et al., 1995). Horiguchi et al. (1997) reported that the threshold body concentration at which TBT induces imposex was 0.02 mg/kg in rock shell.

It is well known that tributyltin (TBT) and heavy metals can accumulate in tissues of aquatic organisms and cause deleterious effects. With an increasing amount of public concern about the possible harmful effects on human health resulting from exposure to TBT and other metals, the consumption of contaminated seafood has been reported as an important route of human exposure to metals in Taiwan (Han et al., 1998). Oyster is the most popular seafood in Taiwan. In particular, the potential risk of consuming oysters is relatively higher than for other seafoods due to the high bioaccumulation of contaminants in oysters. However, the consumption of TBT and other metals from oysters for the general population and fishermen in Taiwan has not yet been conducted. We consider the concentrations of TBT, Cu, Zn, Cd, As, and consumption of oysters of Taiwanese to be important information in relation to public health in Taiwan. Therefore, the aim of this study was to evaluate the public health threat associated with TBT, Cu, Zn, Cd and As from oysters for the general population and fishermen in Taiwan. With oyster as one of the most popular seafoods in Taiwan, it is crucial to estimate the acceptable daily intake of oysters, particularly for fishermen since they usually reserve 600–1200 g/day of oysters for their family after harvesting. Thus, fishermen may be a high-risk exposure group. Finally, another purpose of this research was to clarify the major threat contributors of TBT, Cu, Zn, Cd and As.

2. Materials and methods

The oyster samples were collected from the

different coasts of western Taiwan during the period from 1991 to 1998. Sampling locations are shown in Fig. 1. Previous studies have reported the concentrations of TBT, Cu, Zn and Cd and As in oysters, respectively (Hung et al., 1998; Han et al., 2000). Based on these data, we can estimate the daily intake of TBT, Cu, Zn, Cd and As from

oysters for the general population and fishermen. The study population included the general population living in Taipei City and fishermen who live in the Hsiangshan coastal area of Taiwan. To evaluate the oyster consumption rate, subjects were personally interviewed with a questionnaire. The questionnaire included demographic infor-

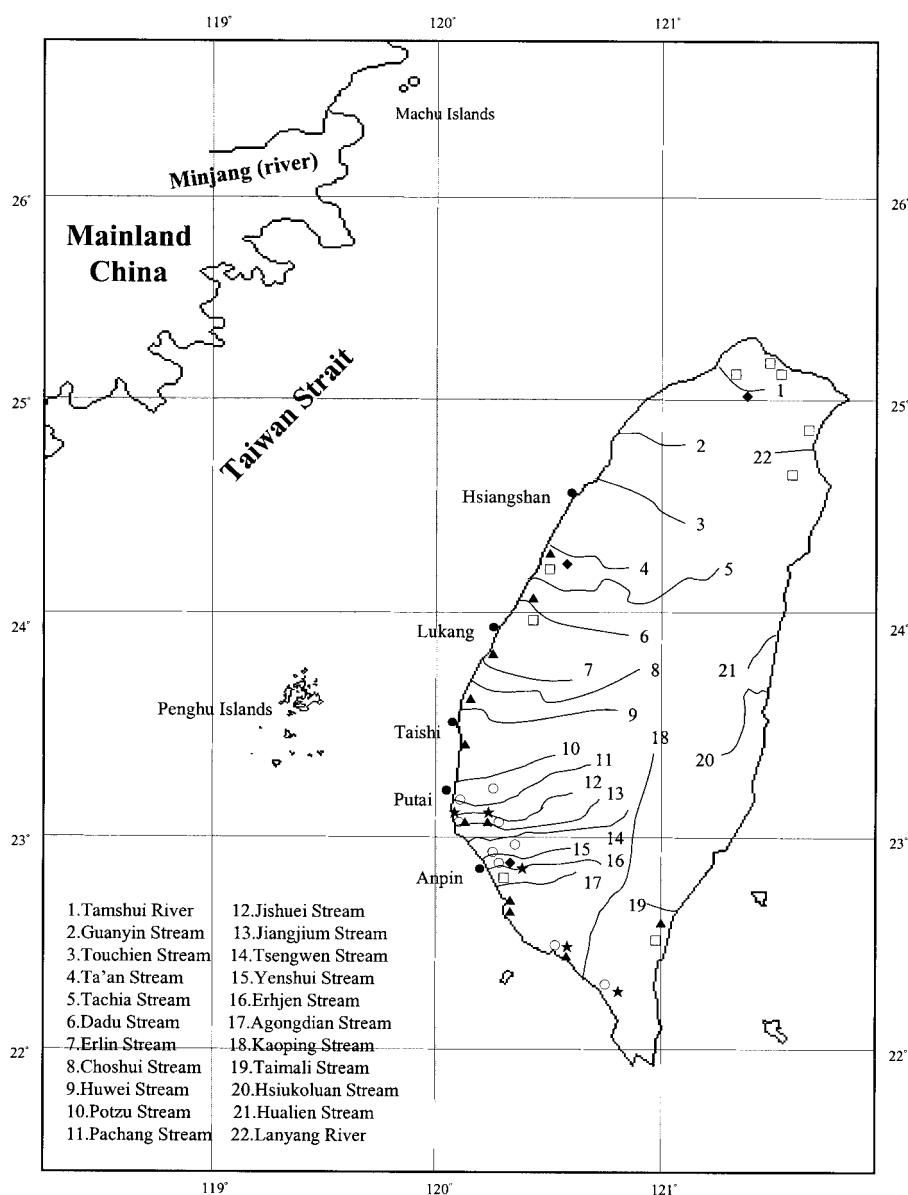


Fig. 1. Sampling locations (●) of oysters and cancer mortality rate for Taiwanese from different coastal areas in Taiwan. [Top 10% cancer mortality rates for liver cancer (▲), gastric cancer (□), bladder cancer (○), breast cancer (◆), kidney (★).]

mation and frequency of oyster consumption. Assuming a 65-g serving per meal of oysters, in the general population, oyster consumers were divided into four groups: a slight-intake group (< 18.6 g/day); a moderate-intake group (18.6–56 g/day); a high-intake group (65–130 g/day); and the highest-intake group (> 139 g/day). In fishermen, oyster consumers were divided into four groups: a slight-intake group (< 94.1 g/day); a moderate-intake group (93–167 g/day); a high-intake group (176–241 g/day); and the highest-intake group (> 250 g/day).

The methodology for the estimation of THQs used was provided in the USEPA Region III risk-based concentration table (USEPA, 2000). According to the report of USEPA, (USEPA, 2000), the dose calculations were made using the standard assumption for an integrated USEPA risk analysis, with the average adult body weights of the Taiwanese assume to be 65 kg. Additionally, based on the USEPA guidance (USEPA, 1989), we assumed that the ingested dose is equal to the absorbed contaminant dose and that cooking has no effect on the contaminants (Cooper et al., 1991).

In general, there are two methods of estimating risks. One is based on carcinogenic effects, and the other is based on non-carcinogenic effects. For non-carcinogenic effects, the risk is expressed as a THQ (target hazard quotients), the ratio

between exposure and the reference dose. Thus, a THQ value below 1 means that the level of exposure is smaller than the reference dose, which assumes that a daily exposure at this level is not likely to cause any deleterious effects during lifetime in human population. In other words, a THQ below 1 means the adverse effects are negligible.

The models for estimating THQs are:

$$\text{THQ} = \frac{\text{Efr} \times \text{EDtot} \times \text{IFR} \times C}{\text{RfDo} \times \text{Bwa} \times \text{ATn}} \times 10^{-3}$$

where THQ is the target hazard quotient; EFr is the exposure frequency (350 days/year); EDtot is the exposure duration (70 years); IFR is the food ingestion rate (g/day); C is the concentration (μg/g); RfDo is the oral reference dose (mg/kg/day); Bwa is the adult body weight (65 kg); and ATn is the averaging time for non-carcinogens (365 days/year × number of exposure years, assuming 70 years). For example, in Table 1, the TBT concentration in oyster is 1.51 μg/g dry wt. for Hsiangshan fishermen and the oyster consumption rate is 94.1 g/day for individuals with typical exposure. It is assumed that TBT concentration in dry-weight is five times the wet-weight. The oral reference dose is 3×10^{-4} mg/kg/day. Based on the above model, THQs is estimated at 1.47 for Hsiangshan fishermen.

Table 1

Geometric mean (GM) concentration (μg/g dry wt.) of TBT and other metals in oysters (*Crassostrea gigas*) collected from different coastal areas of Taiwan

Location	TBT	ΣBTCs	TBT/ΣBTCs (%)	Cu	Zn	Cd	As
Hsiangshan	1.51	1.66	91	1180 (113–2806)	1567 (303–3593)	1.10 (0.167–2.93)	12.1 (7.15–17.6)
Lukang	0.83	0.92	91	378 (60.3–948)	983 (263–1772)	1.50 (0.165–3.75)	13.9 (0.025–29.3)
Taishi	0.40	0.43	91	154 (27.0–520)	547 (137–1145)	1.20 (0.550–2.40)	17.1 (7.05–19.4)
Putai	0.81	0.90	89	139 (124–158)	402 (280–525)	2.24 (1.21–4.15)	4.9 (3.15–7.00)
Anpin	0.32	0.37	86	545 (378–1867)	2545 (574–3662)	2.38 (1.47–4.48)	11.8 (8.41–18.5)
GM	0.66	0.74	89.6	349	970	1.60	11.1

ΣBTCs = MBT + DBT + TBT.

3. Results and discussion

3.1. Daily intake of oysters

The Council of Agriculture of Taiwan, Republic of China categorized 11 major kinds of seafood based on the total amounts of their production. Oyster is the only shellfish on the list (Han et al., 1994). In Taiwan, oyster consumption rates are 18.6 and 139 g/day for typically and maximally exposed individuals of general population, respectively (Han et al., 1998). Fig. 2 shows the oyster consumption rates among fishermen and general population in Taiwan. The values of oyster consumption for fishermen are 94.1 and 250 g/day for typically and maximally exposed individuals, respectively. In particular, the highest intake (250 g/day) from fishermen is almost two times greater than that of the general population (139 g/day). The highest intake of fishermen is associated with the custom in which they preserve 600–1200 g/day of oysters for their family after harvesting.

3.2. Concentrations of TBT and other metals in oysters

Table 1 presents geometric mean (GM) con-

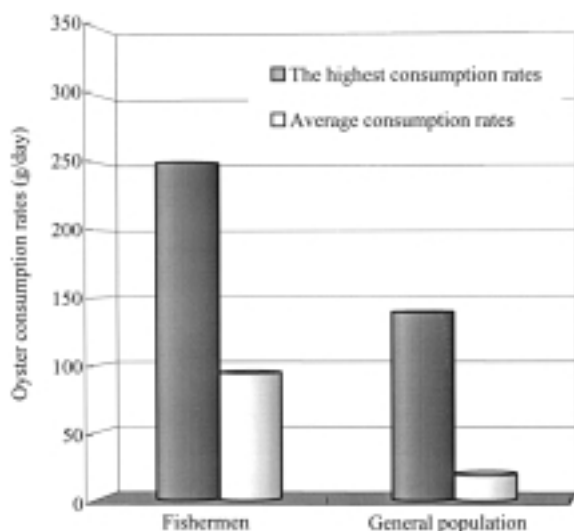


Fig. 2. The oyster consumption rates among fishermen and general population in Taiwan.

centrations of TBT and other metals in oysters collected from different coastal areas of western Taiwan (Hung et al., 1998; Han et al., 2000). In general, TBT concentrations in oysters ranging from 0.32 to 1.51 $\mu\text{g/g}$ dry wt. varied with sampling locations. For example, the TBT concentrations in the oysters of Taiwan were relatively higher compared with those of Korea. The range of TBT concentrations in oyster was 95 to 885 ng/g dry wt. (mean $\pm$ S.D., 384 ± 217 ng/g) at the innermost parts of Masan Bay, Kohyonsong Bay, Haengam Bay, and Wonmunpo Bay at the south coast of Korea (Shim et al., 1998). The highest TBT, Cu, and Zn GM concentrations in oysters of 1.51, 1180 and 1567 $\mu\text{g/g}$ dry wt. were obtained from the Hsiangshan coastal area (Hung et al., 1998; Han et al., 2000). Lower Cu, Zn, As values (GM = 139, 402 and 4.9 $\mu\text{g/g}$ dry wt., respectively) were observed at the Putai coastal area compared with those oyster culture areas in western Taiwan. On the whole, oysters with higher concentrations of Cu and Zn have still occasionally been observed in the Hsiangshan coastal area since 1986. The Cu and Zn concentration in oysters for each location gradually increased year by year, especially for Cu (Han et al., 2000). In general, when the Cu concentration in oysters was over 500 $\mu\text{g/g}$ dry wt., the color of the oysters became green (Han and Hung, 1990). The results indicate that local and regional inputs of Cu and Zn are the major cause of green oysters in this area. Because of the industry's growth, their environmental impact increased in Taiwan from 1985 to 2000. In addition, the range of total butyltin compounds concentrations was 0.37–1.66 $\mu\text{g/g}$ dry wt. in oysters. TBT is the major composition (86–91%) of total butyltin compounds. The source of TBT in the area might be industrial and/or agriculture pollutions from the upper stream.

3.3. Health threat of consuming oysters

Using reference doses of 3×10^{-4} , 4×10^{-2} , 0.3, 5×10^{-4} and 3×10^{-4} mg/kg/day for TBT, Cu, Zn, Cd and As, respectively (USEPA, 2000), estimates of target hazard quotients (THQs) for TBT and other metals from different coastal ar-

Table 2

Various estimated target hazard quotients (THQs) for metals and TBT caused by consuming oysters for general population and fisherman in Taiwan

Exposure group	Location	Maximally exposed individuals					Typically exposed individuals				
		TBT	Cu	Zn	Cd	Inorganic As	TBT	Cu	Zn	Cd	Inorganic As
General population	Taiwan area	0.76	2.45	1.12	1.50	1.53	0.10	0.33	0.15	0.20	0.21
Fishermen	Hsiangshan	3.87	20.5	3.87	1.65	2.97	1.47	7.69	1.47	0.61	1.06
	Lukang	2.13	5.68	2.03	1.73	3.24	0.80	2.12	0.76	0.66	1.11
	Taishi	1.00	2.84	1.35	1.76	4.19	0.40	1.06	0.51	0.66	1.47
	Putai	2.07	2.57	0.99	3.29	1.19	0.77	0.96	0.35	1.21	0.40
	Anpin	0.80	10.7	6.26	3.60	2.41	0.30	4.05	2.38	1.36	0.91

eas are given in Table 2. As expected, the maximally and typically exposed fishermen are higher than the general population. THQ values of the general population are below 1.0 for typically exposed individuals. However, 90% (27 out of 30 THQ) THQs exceed 1.0 for maximally exposed individuals. In other words, this indicates that people who are exposed to contaminated oysters have potential health risks. In particular, the THQ values of Hsiangshan's fishermen of 3.87 and 20.5 for TBT and Cu for maximally exposed individuals are higher than other oyster culture areas. It is interesting those consuming oysters from Hsiangshan, Lukang, Taishi causes abnormally high THQs of TBT and other metals (100% over 1.0). This means that the intake of oysters from those oyster culture areas could be potentially dangerous, especially for high-risk fishermen.

Fig. 3 shows the results of estimated total THQs caused by consuming contaminated oysters from different coastal areas for the general population and fishermen. Where exposure to two or more toxicants may result in additive and/or interactive effects (Hallenbeck, 1993), the risk addition hypothesis was adopted here. On the other hand, total THQ is the sum of the following compositions:

Total THQ (TTHQ)

$$= \text{THQ (toxicant 1)} + \text{THQ (toxicant 2)} + \dots + \text{THQ (toxicant } n).$$

For maximally exposed individuals, the values of total THQs for different coastal area fishermen

decreased in the order to of Hsiangshan > Anpin > Lukang > Taishi > Putai. The maximal value of total THQ from Hsiangshan is 3.26-fold greater than Putai (THQ = 32.9 and 10.1, respectively). In general, total THQ values for each population all greater than 1 indicate health risks from exposure to contaminated oysters that are potentially of concern. The total TBT and other metal concentrations give a total THQ in excess of 1; therefore, the source of TBT and metals needs to be remediated.

Fig. 4 presents the composition of the relative contribution to THQ by Cu, Zn, Cd, inorganic As and TBT in oysters for the general population and fishermen. Nevertheless, the major risk contributor of pollutants for fishermen of Hsiangshan, Anpin, and Lukang is Cu (62, 46 and 39%, respectively). Inorganic arsenic is the major risk pollutant for the fishermen of Taishi and Lukang (36 and 20%, respectively). Observably, Fig. 4 indicates that the potential health risks of other metals from contaminated oysters are higher than TBT. In other words, TBT is attributed to only 3–21% of the total THQs in different fishermen of Taiwan. The current understanding of TBT toxicity to humans suggests there is a threshold dose below which no toxicity will be observed (USEPA, 1997), although TBT has been demonstrated to be toxic to the thymus-dependent immune system of the rat (Vos et al., 1990; USEPA, 1997). Immunotoxic effects also have been observed in a chronic toxicity study with dogs (Murphy, 1995). In addition, it is of note that oyster consumption in these areas is the major

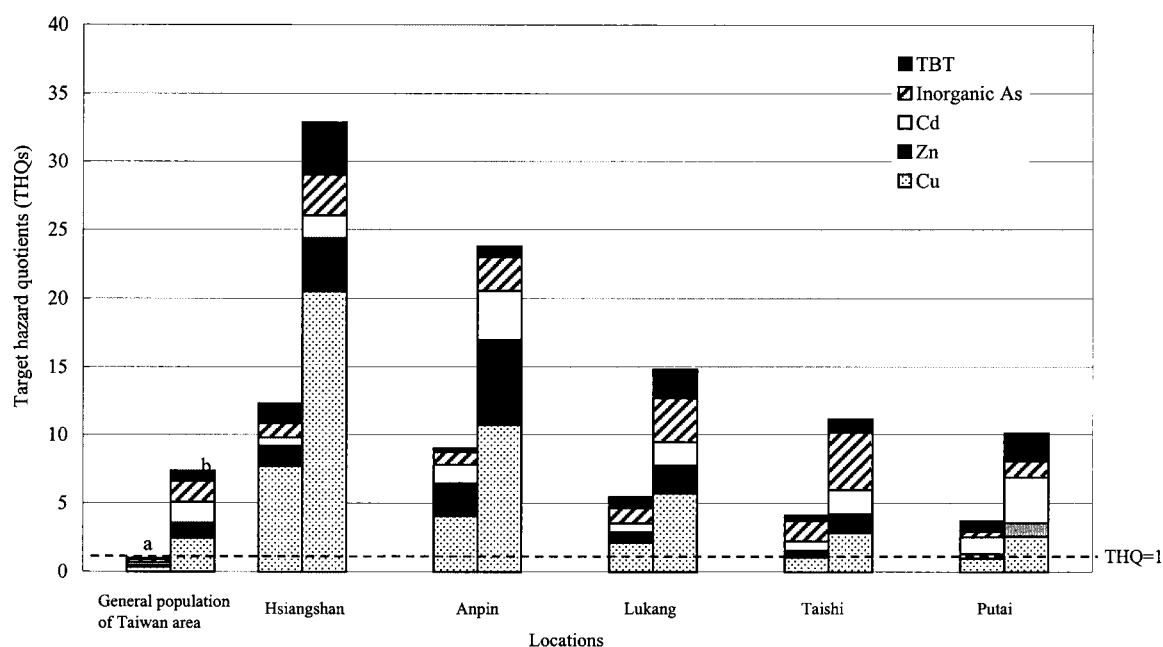


Fig. 3. THQs value of various metals caused by consuming oysters from different locations of general population and fishermen: (a) typically exposed individuals; and (b) maximally exposed individuals.

source of dietary As exposure (Lin and Han, 1999). Basically, the THQs for inorganic As had a larger percentage contribution (ranging from 9–36%) of total THQs from various fishermen. Inorganic arsenic increases the formation of free radicals; lipid peroxides, and decreases selenium levels. As a result, inorganic arsenic exposure has been linked to neurological and cardiovascular disease/disorders (Maitani et al., 1987; Jensen et al., 1991; Chiou et al., 1997).

In the mid-1950s, Dr John Higginson made what has become a now widely quoted, and often mis-quoted statement, that 80–90% of all cancers are caused by environmental factors (Hall, 1990; Whelan, 1993). Doll and Peto published research that attempted to quantify cancer mortalities with regards to various environmental factors, showing that 35% of human cancer deaths in United States are attributable to dietary causes (Doll and Peto, 1981). In Taiwan, fish and shellfish are very important in the Taiwanese diet as major sources of certain vitamins, essential trace elements, and polyunsaturated *n*-3 fatty acids. However, potential health risks of TBT and other metals corre-

spond to the amount of oyster consumption. Further data also suggest a possible association between heavy metal exposure and cancer. Fig. 1 shows the top 10% age-adjusted mortality rates of cancers per 100 000 person-years of all townships from 1982 to 1991 in Taiwan. Higher cancer mortality rates are clustered in down-stream and coastal areas (DOH, 1996). Study sites in this study, such as Putai and Anpin, are among the top 10% in mortality rates caused by breast and kidney cancer. Another location that exhibits an association between heavy metals and cancer is in the black-foot disease (BFD) endemic area. Higher mortality from liver and bladder cancers has been observed among residents in the BFD-endemic area, where most residents are fishermen.

Our results suggest that current environmental levels of TBT and other metals are potential threats to the health of fishermen in coastal areas of Taiwan. TBT and other metals were present in the oysters. Furthermore, human health threats associated with oyster consumption were not negligible. However, more research is needed to clar-

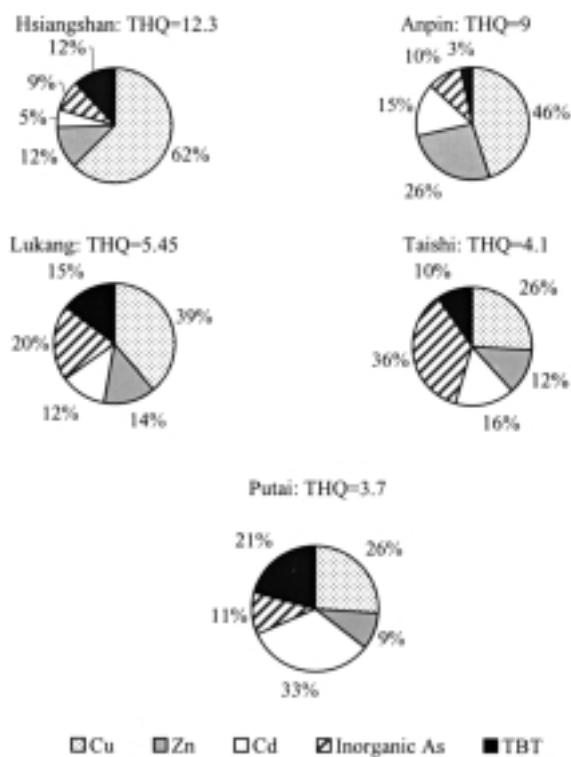


Fig. 4. Composition of relative contribution of THQ by Cu, Zn, Cd, inorganic As and TBT from consuming oysters for fishermen of different coastal areas.

ify and quantify the health risk. Finally, since oysters are a nutritive valuable food, it should not be omitted from a balanced diet, but a limited consumption of oysters and other seafood from polluted water should be considered. We would recommend that a government facing similar issues should actively resolve the pollution problem. Before the pollution problem is resolved, a public awareness campaign and limiting the consumption of contaminated seafood should be considered.

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