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# Heavy Metal Content of Rice and Shellfish in Taiwan

# HAW-TARN LIN\*, SUE-SUN WONG AND GWO-CHEN LI

Residue Control Department, Taiwan Agricultural Chemicals and Toxic Substances Research Institute, Council of Agriculture, 11 Guangming Rd., Wufong Township, Taichung County 413, Taiwan, R.O.C.

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# ABSTRACT

This investigation surveyed heavy metal content of Taiwanese rice and shellfish. A total of 407 rice samples and 83 shellfish samples were collected from various areas of Taiwan. The content of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn in the rice samples was 0.08, 0.01, 0.10, 2.22, 0.001, 0.29, 0.01 and 14.7 mg/kg, respectively. Notably, the Cd and Hg content in the rice samples was found to be below the food sanitary standards of Cd (0.5 mg/kg) and Hg (0.05 mg/kg) in Taiwanese rice. On the other hand, the As, Cd, Cr, Cu, Hg, Ni, Pb and Zn content in shellfish was 1.55, 0.15, 0.18, 13.1, 0.004, 0.37, 0.17 and 50.3 mg/kg, respectively. Notably, oysters had higher content of Cu and Zn than mussels. The average content of heavy metals in Taiwanese shellfish was always below the limits or standards of the UK, Denmark and Germany. To assess the safety of dietary intake, weekly intake of heavy metals by rice and shellfish was calculated based on the intake of a typical Taiwanese. The results indicated that weekly intake of heavy metals from rice and shellfish was below the provisional tolerable weekly intakes recommended by WHO/FAO and USNAS.

Key words: heavy metals, rice, shellfish, arsenic, cadmium, chromium, copper, mercury, nickel, lead, zinc, weekly intake

# INTRODUCTION

Heavy metals, such as As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, are widely used in industry in Taiwan. Entering the environment from both natural and anthropogenic sources<sup>(9,10)</sup>, these heavy metals contaminate food source and accumulate in both agricultural products and seafood through water, air, and soil pollution if waste discharge is not properly treated. For example, cadmium polluted rice in Taoyuan county and contaminated green oysters in the Erhjen River estuary resulted from the illegal discharge of waste waters from chemical plants and metal recycling factories<sup>(7,15)</sup>. Moreover, heavy metals can enter the food chain from aquatic and agricultural ecosystems and threaten human health indirectly<sup>(3)</sup>. The cases of Minamata and Itai-Itai diseases in Japan were caused by food contaminated with heavy metal<sup>(8,12,16)</sup>.

We have surveyed the heavy metal content of 687 crop samples and evaluated the daily intake of heavy metal by Taiwanese during 1990 to 1991. The results revealed that the daily intakes were within the acceptable daily intakes<sup>(14)</sup>. Rice dominates the Taiwanese diet. However, shellfish, another important part of the Taiwanese diet, easily accumulate heavy metals in water by bioconcentration and biomagnification. Therefore, the heavy metal content of both rice and shellfish is worthy of consideration. This investigation surveyed the heavy metal content of rice and shellfish, and evaluated the safety of consuming these two foods in Taiwan.

I. Materials

# (I) Samples and sample preparation

A total of 137 barn rice samples were collected from major rice production areas in Taiwan, and 280 packed rice samples (weighing from 2 to 5 kg) were purchased from markets around Taiwan. Rice samples were ground using an agate mortar, passed through a 50 mesh (< 0.30 mm) sieve, sealed in a plastic box and stored at  $4^{\circ}$ C until analysis.

MATERIALS AND METHODS

Shellfish samples were collected from the main production areas in Taiwan (Taichung, Changhwa, Yunlin, Chiayi, Penghu and Tainan). Eighty-three samples were collected: 59 samples of oysters (*Crassostrea gigas*) and 24 samples of mussels. The mussel samples comprised 14 clams (*Meretrix lusoria*) and 10 venus clams (*Ruditapes variegata*). All soft tissue from the shellfish samples was removed, homogenized, and then freeze-dried before analysis. Sample water content also was calculated based on the fresh and dried sample weight.

# (II) Reagents and utensils

All chemicals used were of "suprapure" or equivalent quality, and the standard solutions of all metals were sourced from Merck. Moreover, all glassware was of Pyrex or similar quality. Glassware were previously soaked in 15% HNO<sub>3</sub> solution for at least 48 hr and afterwards rinsed with deionized water.

<sup>\*</sup> Author for correspondence. Tel: 886-4-23302101 ext. 412; Fax: 886-4-23324738; E-mail: htlin@tactri.gov.tw

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# II. Methods

# (I) Determination of heavy metal in $rice^{(13)}$

To determine As content in rice, 0.5 g of each sample was refluxed in 10 mL of conc. nitric-sulfuric-perchloric acid mixture (4/1/1, v/v/v) for 1 hr. Formic acid (90%) was then added drop by drop until the red-brown gas disappeared. Afterwards, deionized water was added to bring the digest to 25 mL. The resulting solution was reacted with an aqueous solution of 1% NaBH<sub>4</sub> and 1% NaOH. The analysis of As included a JOBIN-YVON JY-138 ULTRACE ICP-AES equipped with a hydride generator.

To determine Cd, Cr, Cu, Ni, Pb and Zn content in rice, 0.5 g of each rice sample was refluxed in 5 mL of conc. HNO<sub>3</sub> for 1 hr, and then 5 mL of 70% perchloric acid was added. The reflux was continued for another 0.5 hr. The digests were analyzed by JY-138 ULTRACE ICP-AES.

To determine Hg content in rice, 0.3 g of sample was digested with 10 mL of conc. HNO<sub>3</sub> in a water bath ( $80^{\circ}$ C) until the digest was clear. The amount of Hg was determined with a Varian SpectrAA-30 atomic absorption spec-

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trometer equipped with a VGA-77 Hg analysis kit after the digest was reacted with 25% SnCl<sub>2</sub> (in 20% HCl) solution.

#### (II) Determination of heavy metal in shellfish

For analyzing As in shellfish, 0.2 g of each sample was refluxed in 5 mL of conc.  $HNO_3$  for 1 hr, and then 2 mL of  $H_2SO_4$  was added and the reflux was continued for another 1 hr. Deionized water was then added to top the sample up to 25 mL. The instrumental determination of As in the shellfish sample was the same as in the rice sample.

Hg, Cd, Cr, Cu, Hg, Ni, Pb and Zn content in the shellfish sample was determined in the same way as the rice samples.

The analytical methodologies were confirmed using certified reference materials: oyster tissue 1566b, rice flour 1568 and bovine liver 1577a from NIST, rice flour 08510 and 08511 from the PRC, and TORT-2 and DOLT-2 from Canada. Table 1 lists the results of these analyses. The recovery rates range from 71% to 115% (n = 3).

Table 1. Analysis results for standard reference materials (mg/kg, on dry weight basis)

SRM (No.)	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Oyster tissue	$7.65\pm0.65^a$	$2.48 \pm 0.08$	_	$71.6 \pm 1.6$	$0.0371 \pm 0.0013$	$1.04\pm0.09$	$0.308\pm0.009$	$1424 \pm 46$
(SRM1566b)	$(6.96 \pm 0.28)^{b}$	$(2.17\pm0.02)$		$(59.5\pm0.25)$	$(0.03 \pm 0.01)$	$(0.98\pm0.08)$	$(0.29\pm0.02)$	$(1495 \pm 36)$
Rice flour	$0.41\pm0.05$	$0.029 \pm 0.004$	_	$2.2 \pm 0.3$	$0.006 \pm 0.0007$	0.10 <sup>c</sup>	$0.045\pm0.010$	$19.4\pm1.0$
(SRM1568)	$(0.36\pm0.02)$	(ND)		$(1.82\pm0.12)$	(ND)	$(0.16\pm0.01)$	(ND)	$(22.4\pm2.0)$
Rice flour	_	$2.60\pm0.05$	_	_	-	_	_	_
(08510)		$(2.40\pm0.04)$						
Rice flour	_	$0.50\pm0.02$	_	_	-	_	_	_
(08511)		$(0.45 \pm 0)$						
Bovine liver	$0.047\pm0.006$	$0.44\pm0.06$	_	$158\pm7$	$0.004\pm0.002$	_	$0.135\pm0.015$	$123\pm8$
(SRM1577a)	(ND)	$(0.40\pm0.05)$		$(128 \pm 1)$	(ND)		$(0.13\pm0.05)$	$(124 \pm 4)$
Lobster	$21.6\pm1.8$	$26.7\pm0.6$	$0.77\pm0.15$	$106\pm10$	$0.27\pm0.06$	$2.50\pm0.19$	$0.35\pm0.13$	$180 \pm 6$
(TORT-2)	$(16.7 \pm 0.26)$	$(27.8 \pm 1.2)$	$(0.55 \pm 0.20)$	$(82.8\pm1.2)$	$(0.20 \pm 0.05)$	$(2.00\pm0.05)$	$(0.48\pm0.20)$	$(163 \pm 4)$
Dogfish liver	$16.6\pm1.1$	$20.8\pm0.5$	$0.37\pm0.08$	$25.8\pm1.1)$	-	$0.20\pm0.02$	$0.22\pm0.02$	$85.8\pm2.5$
(DOLT-2)	$(11.9\pm0.51)$	$(21.7\pm0.4)$	$(0.40\pm0.01)$	$(22.9\pm0.1$		$(0.30\pm0.05)$	$(0.25\pm0.02)$	$(92.5 \pm 1.2)$
Detection limits	0.10	0.05	0.10	0.05	0.01	0.10	0.20	0.05

<sup>a</sup>Certified value.

<sup>b</sup>Value detected, data are means ± standard deviations of three replications.

<sup>c</sup>Uncertified value.

Table 2.	Content	of heavy	metals in	rice (	(mg/kg,	on fresh	weight basis)

Sample source	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Packed $(n = 280)^{c}$	$\begin{array}{c} 0.10 \pm 0.08 b^a \\ ({<}0.10 - 0.63)^b \end{array}$	$\begin{array}{c} 0.02 \pm 0.05 b \\ (<\!0.05 - 0.20) \end{array}$	$\begin{array}{c} 0.07 \pm 0.20a \\ (<\!\!0.10-1.55) \end{array}$	2.20 ± 0.81a (<0.05 - 7.60)	0.0004 ±0.0026a (<0.01 - 0.02)	0.26 ± 0.33a (<0.10 - 3.75)	$\begin{array}{c} 0.01 \pm 0.04a \\ (< 0.10 - 0.20) \end{array}$	$\begin{array}{c} 14.6 \pm 10.4b \\ (<\!0.05 - 70.1) \end{array}$
Barn (n = 137)	$\begin{array}{c} 0.05 \pm 0.04a \\ ({<}0.10 - 0.14) \end{array}$	$\begin{array}{c} 0.003 \pm 0.02a \\ (<\!0.05 - 0.20) \end{array}$	$\begin{array}{c} 0.17 \pm 0.61b \\ (< 0.10 - 5.45) \end{array}$	$\begin{array}{c} 2.26 \pm 0.55a \\ (0.85 - 3.80) \end{array}$	$\begin{array}{c} 0.003 \pm 0.008 b \\ ({<}0.01 - 0.04) \end{array}$	$\begin{array}{c} 0.35 \pm 0.27a \\ ({<}0.10 - 1.25) \end{array}$	$\begin{array}{c} 0.01 \pm 0.04a \\ ({<}0.10 - 0.20) \end{array}$	10.1 ± 5.6a (<0.05 - 30.5)
Total (n = 417)	$\begin{array}{c} 0.08 \pm 0.07 \\ ({<}0.10 - 0.63) \end{array}$	$\begin{array}{c} 0.01 \pm 0.04 \\ (<\!\!0.05 - 0.20) \end{array}$	$\begin{array}{c} 0.10 \pm 0.39 \\ (<\!\!0.10-5.45) \end{array}$	$\begin{array}{c} 2.22 \pm 0.73 \\ (<\!\!0.05 - 7.60) \end{array}$	$\begin{array}{c} 0.001 \pm 0.005 \\ ({<}0.01 - 0.04) \end{array}$	$\begin{array}{c} 0.29 \pm 0.31 \\ (<\!\!0.10 - 3.75) \end{array}$	$\begin{array}{c} 0.01 \pm 0.04 \\ (<\!0.10 - 0.20) \end{array}$	$\begin{array}{c} 13.1 \pm 9.4 \\ (<\!\!0.05-70.1) \end{array}$

<sup>a</sup>Data are means  $\pm$  standard deviations, and the values in the same column followed by different letters are significantly different, (p < 0.05) by Duncan's multiple range test.

<sup>b</sup>Range.

<sup>c</sup>Number of samples.

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# **RESULTS AND DISSCUSSION**

# I. Heavy Metal Content in Rice

Table 2 lists heavy metal contents of the rice samples. The average content of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn in packed rice sold in markets from various areas was 0.10, 0.02, 0.07, 2.20, 0.0004, 0.26, 0.01 and 14.6 mg/kg, respectively (on a fresh weight basis). Meanwhile, the average content of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn in barn rice from various areas was 0.05, 0.003, 0.17, 2.26, 0.003, 0.35, 0.01 and 10.1 mg/kg, respectively (on a fresh weight basis). The As, Cd and Zn content of packed rice was higher than that of barn rice (Table 2). The 137 barn rice samples were all polished rice, while 29 of the packed rice samples were unpolished rice. Polishing of the bran and embryo may reduce the concentration of some heavy metals. Kitagishi and Obata (1981) indicated that Cd content in polished rice is 20% lower than in unpolished rice<sup>(11)</sup>. The average content of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn in all rice samples was 0.08, 0.01, 0.10, 2.22, 0.001, 0.29, 0.01 and 13.1 mg/kg, respectively (on a fresh weight basis). In comparison, the mean Hg content of unpolished Japanese rice sampled during 1989 to 1997 was  $0.004 \text{ mg/kg}^{(17)}$ . Additionally, the average Cu content in unpolished rice from Japan was 2.9 mg/kg, and that in Javanese rice from Indonesia was  $3.41 \text{ mg/kg}^{(22)}$ . As for the Cd, rice from USA had the content of 0.013 mg/kg<sup>(30)</sup>. In sum, investigation of the Cd content of rice from different countries revealed a range from 0.0008 to 0.13 mg/kg, with the average being 0.03 mg/kg<sup>(23)</sup>. The Pb content in rice samples from various countries ranged from 1.6 to 58.3 ng/g, and the average content was 15.7 ng/g<sup>(31)</sup>. Comparing the results in Table 1 with heavy metal content of rice from foreign countries, we found lower heavy metal content in Taiwanese rice. Additionally, the average heavy metal contents in the rice samples of this survey is lower than in an similar survey conducted in 1992 with the exception of Cu and Hg content. The average As, Cd, Cr,

Cu, Ni, Pb and Zn content in the rice samples in the 1992 survey was 0.15, 0.06, 0.14, 2.16, 0.001, 0.47, 0.37 and 34.1 mg/kg, respectively<sup>(14)</sup>.

The food sanitary standards of Cd and Hg in Taiwanese rice were 0.50 and 0.05 mg/kg, respectively<sup>(5)</sup>, which were not higher than for any other rice samples (Table 2). The average content of As, Pb, Cd and Hg in Taiwanese rice also was below the maximum permitted level for cereals in Germany which were 1.0, 0.3, 0.1 and 0.05 mg/kg, respectively<sup>(2)</sup>.

Table 3 lists the heavy metal content of packed rice from markets in different production areas of Taiwan. Comparing the heavy metal content of rice from each area revealed no difference in the Cd, Hg, Ni and Pb content. However, samples from the central and southern areas of Taiwan have higher As and Cu content. Samples from the central and northern areas had higher Zn content, while samples from the southern areas had higher Cr content. The heavy metal content of packed rice from Taiwan was also compared with that from other countries. The average Cu content of Taiwanese rice was higher than that from other countries. Specifically, the average Cu content of Taiwanese rice was 2.24 mg/kg, and the average Cu content of rice from other countries was 1.45 mg/kg.

Figure 1 shows the distribution of heavy metal contents in Taiwanese rice and reveals that 68.5% of rice samples had As content below 0.10 mg/kg. Moreover, 90% of samples had less than 0.10 mg/kg Cd. Meanwhile, 71.5% of samples had Cr content less than 0.10 mg/kg, and 51.9% of samples had Cu content ranging from 2.0 to 3.0 mg/kg. The Hg content of most rice samples (94.3%) was undetectable (the detection limit was 0.01 mg/kg). In addition, 62.5% of the rice samples had Ni content ranging from 0.10 to 0.50 mg/kg. Over 90% of the Pb content in rice samples was undetectable (the detection limit is 0.10 mg/kg). Regarding the distribution of Zn content in the rice samples, 43.2% of samples had Zn below 10 mg/kg, and 40.5% of samples had 10 to 20 mg/kg of Zn.

Table 3. Heavy metal content in packed rice from different production areas (mg/kg, on fresh weight basis)

Production area	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
	0.07 0.00 0				0.001 0.002		0.01 0.02	
North <sup>a</sup> $(n = 15)^b$	$0.07 \pm 0.08 a^{g}$	$0.04 \pm 0.07a$	$0.02 \pm 0.05a$	$2.23 \pm 0.56b$	$0.001 \pm 0.003a$	$0.25 \pm 0.21a$	$0.01 \pm 0.03a$	$11.7 \pm 5.52b$
West <sup>c</sup> $(n = 92)$	$0.12\pm0.09c$	$0.01\pm0.03a$	$0.06\pm0.16a$	$2.37\pm0.65c$	$0.0001 \pm 0.001a$	$0.28\pm0.28a$	$0.01\pm0.04a$	$17.2\pm8.7b$
South <sup>d</sup> $(n = 69)$	$0.12\pm0.09c$	$0.02\pm0.04a$	$0.13\pm0.31b$	$2.57\pm0.89c$	$0.0004\pm0.003a$	$0.27\pm0.51a$	$0.01\pm0.05a$	$16.7\pm12.9a$
$East^e (n = 90)$	$0.08\pm0.06b$	$0.03\pm0.06a$	$0.03\pm0.11a$	$1.85\pm0.75a$	$0.001\pm0.004a$	$0.25\pm0.21a$	$0.02\pm0.05a$	$11.1\pm8.2a$
Taiwan ( $n = 266$ )	$0.10\pm0.08a$	$0.02\pm0.05a$	$0.07\pm0.20a$	$2.24\pm0.80b$	$0.001\pm0.003a$	$0.26\pm0.33a$	$0.02\pm0.04a$	$14.7\pm10.1a$
Foreign countries <sup>f</sup> $(n = 14)$	$0.08\pm0.04a$	$0.02\pm0.04a$	$0.07\pm0.15a$	$1.45\pm0.43a$	<0.001a	0.18 ± 0.28a	<0.01a	12.3 ± 16.4a
Total (n = 280)	$0.10\pm0.08$	$0.02\pm0.05$	$0.07\pm0.20$	$2.20\pm0.81$	$0.0004\pm0.003$	$0.26\pm0.33$	$0.01\pm0.04$	$14.6 \pm 10.4$

<sup>a</sup>Taoyuan (n = 4), Sinchu (n = 4), Miaoli (n = 6).

<sup>b</sup>n = number of samples.

<sup>c</sup>Taichung (n = 17), Nantou (n = 2), Changhwa (n = 59), Yunlin (14).

<sup>d</sup>Chiayi (n = 32), Tainan (n = 16), Kaohsiung (15), Pingtung (n = 6).

<sup>e</sup>Yilan (n = 20), Hwalien (n = 54), Taitung (n = 16).

<sup>f</sup>Mainland China (n = 5), California (n = 5), Australia (n = 2), Thailand (n = 1), Japan (n = 1).

<sup>g</sup>Data are means  $\pm$  standard deviations, the values in the same column followed by different letters are significantly different, (p < 0.05) by Duncan's multiple range test.

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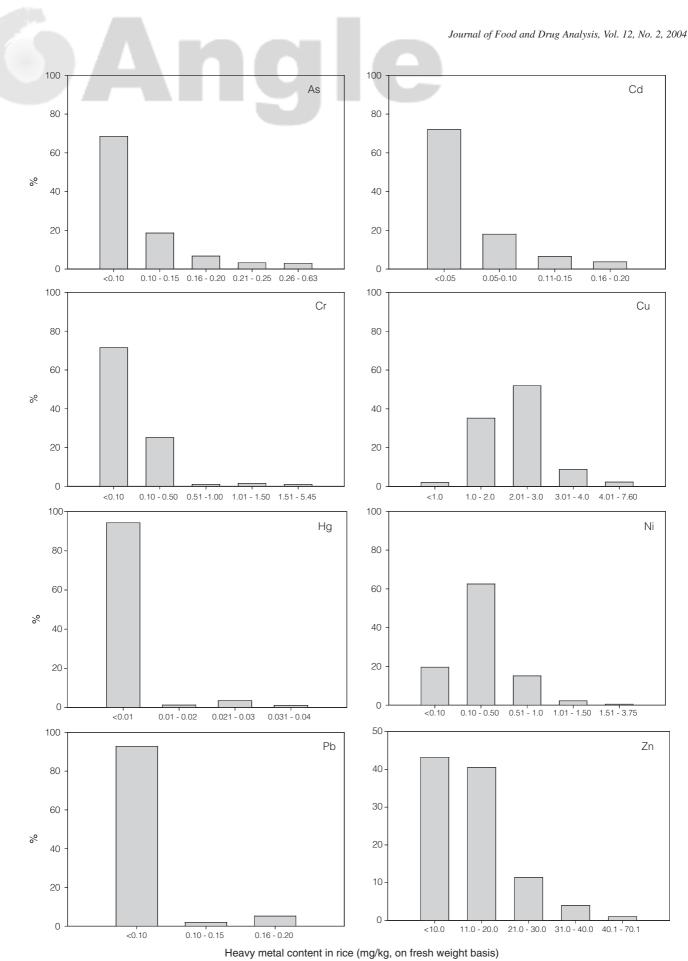


Figure 1. Distribution of heavy metal contents in rice.

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table 4. Heavy metal content in oysters from different areas in farwan (ing/kg, on difed weight basis)									
Area	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	
Taichung $(n = 3)^a$	$10.7\pm0.63d^{b}$	$0.77\pm0.16a$	$1.45\pm0.11a$	$155 \pm 91b$	<0.02a	$1.60\pm0.33a$	$1.58\pm0.08a$	$491 \pm 52.0a$	
Changhwa $(n = 20)$	$9.80 \pm 1.22b$	$0.44 \pm 0.20a$	$1.08\pm0.67a$	$125\pm69.6a$	$0.02\pm0.06b$	$1.55\pm0.93a$	$0.85\pm0.72a$	$474\pm309a$	
Yunlin $(n = 5)$	$9.93 \pm 0.46c$	$0.63\pm0.27a$	$0.73\pm0.47a$	$104 \pm 48.5a$	$0.02\pm0.04b$	$0.64\pm0.50a$	$0.72\pm0.54a$	$299 \pm 167a$	
Penghu $(n = 5)$	$14.5\pm1.15f$	$1.84\pm0.17a$	$1.07\pm0.44a$	$70.5\pm53.5a$	$0.09\pm0.02b$	$0.94\pm0.43a$	$1.16\pm0.74a$	$560\pm310a$	
Chiayi (n = 19)	$10.9 \pm 3.33e$	$2.26\pm5.04a$	$1.41 \pm 1.31a$	$157 \pm 116c$	$0.04\pm0.07b$	$1.81 \pm 1.75 a$	$1.89 \pm 1.70a$	$441\pm232a$	
Tainan (n = 7)	$8.89\pm0.46a$	$0.39\pm0.27a$	$0.97\pm0.67a$	$37.0\pm36.4a$	<0.02a	$0.87\pm0.61a$	$0.54\pm0.36a$	$237 \pm 153a$	
Total (59)	$10.5\pm2.48$	$1.17\pm2.98$	$1.16\pm0.91$	$120\pm93.3$	$0.03\pm0.06$	$1.43 \pm 1.23$	$1.20 \pm 1.21$	$429\pm269$	

Table 4. Heavy metal content in oysters from different areas in Taiwan (mg/kg, on dried weight basis)

<sup>a</sup>n = number of samples.

<sup>b</sup>Data are means  $\pm$  standard deviations, the values in the same column followed by different letters are significantly different, (p < 0.05) by Duncan's multiple range test.

 Table 5. Heavy metal content in mussels (mg/kg, on dried weight basis)

Mussels	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Venus clams $(n = 10)^a$	$10.1\pm6.15a^b$	$0.55\pm0.20a$	$2.15\pm3.93a$	$6.08 \pm 1.84a$	$0.03\pm0.04a$	$2.09\pm2.35a$	$1.15 \pm 1.33a$	$45.7\pm26.9b$
Clams (14)	$13.1\pm7.4a$	$1.96 \pm 1.97 b$	$1.86 \pm 1.34a$	$29.5\pm84.1b$	$0.04\pm0.07a$	$12.9\pm7.6b$	$1.85 \pm 1.18a$	$166 \pm 180 b$
Total (24)	$11.8\pm7.0$	$1.37 \pm 1.66$	$1.98 \pm 2.74$	$19.7\pm64.3$	$0.04\pm0.06$	$8.39 \pm 7.94$	$1.56 \pm 1.29$	$116\pm150$

an = number.

<sup>b</sup>Data are means  $\pm$  standard deviations, the values in the same column followed by different letters are significantly different, (p < 0.05) by Duncan's multiple range test.

Table 6. Heavy metal content in shellfish (mg/kg, on fresh weight basis)

			0	,				
Shellfish	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Oyster $(n = 59)^a$	$1.66\pm0.51b^b$	$0.16\pm0.31a$	$0.17\pm0.12a$	$17.8 \pm 12.9 \mathrm{b}$	$0.004\pm0.007a$	$0.21\pm0.15a$	$0.17\pm0.16a$	$66.5\pm45.3b$
	$(0.80 - 3.17)^{c}$	(0.02 - 2.44)	(trace – 0.88)	(0.29 – 19.9)	(trace - 0.04)	(trace - 0.65)	(trace - 0.58)b	(7.44–193)
Mussels $(n = 24)$	$1.29\pm0.80a$	0.19a	$0.19\pm0.19a$	$1.54\pm3.84a$	$0.004\pm0.007a$	$0.77\pm0.69b$	$0.16\pm0.15a$	$10.5\pm9.20a$
	(0.51 – 3.76)	(0.02 – 0.79)	(trace – 0.53)	(1.40 – 48.6)	(trace - 0.02)	(0.03 - 2.91)	(trace – 0.51)	(2.04 - 47.6)
Total (n = 83)	$1.55\pm0.63$	$0.15\pm0.28$	$0.18\pm0.15$	$13.1\pm13.2$	$0.004\pm0.007$	$0.37\pm0.47$	$0.17\pm0.15$	$50.3\pm46.4$
	(0.51 - 3.76)	(0.02 – 2.44)	(trace - 0.88)	(0.29 – 48.6)	(trace - 0.04)	(trace -2.91)	(trace - 0.58)	(2.04 - 193)

an = number.

<sup>b</sup>Data are means  $\pm$  standard deviations, the values in the same column followed by different letters are significantly different, (p < 0.05) by Duncan's multiple range test.

<sup>c</sup>Range.

## II. Heavy Metal Content in Shellfish

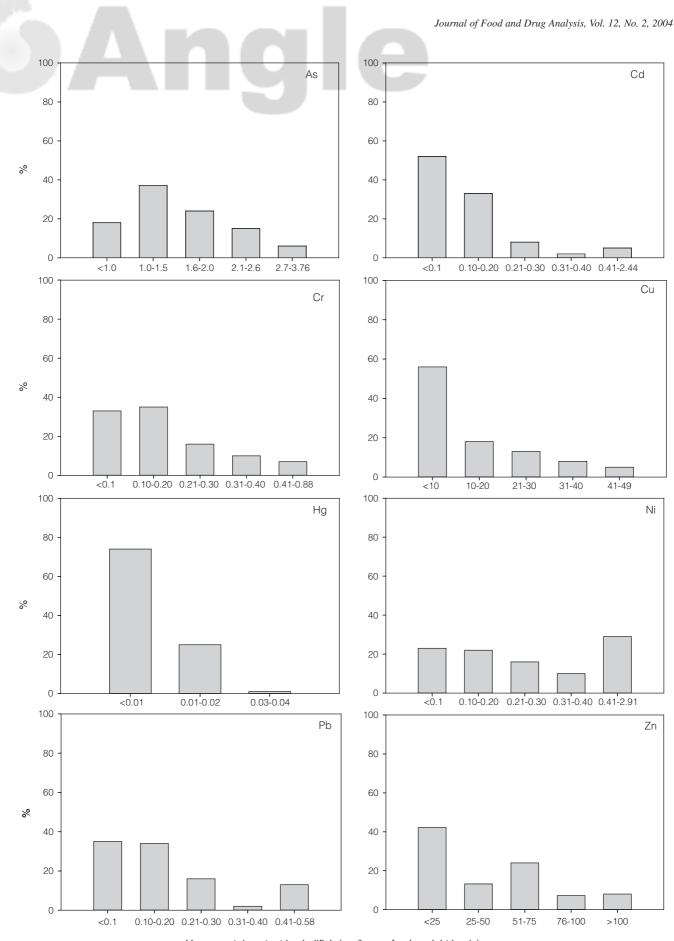
Table 4 lists heavy metal content (on a dried weight basis) of oysters (*Crassostrea gigas*) from different areas. As and Cu content in oysters differed significantly among different areas. Ranking from high to low, As content in oysters was as follows: Penghu > Chiayi > Taichung > Changhwa > Tainan. Cu content was the highest in Chiayi (157 mg/kg), followed by Taichung (155 mg/kg). Oysters from other areas all shared the same Cu content. The results also revealed that Zn was the most prevalent heavy metal in oysters, followed by Cu. The As, Cd, Cr, Cu, Hg, Ni, Pb and Zn content of oysters based on dried weight was 10.5, 1.17, 1.16, 120, 0.03, 1.43, 1.20 and 429 mg/kg, respectively.

Table 5 shows heavy metal content of mussels. The As, Cd, Cr, Cu, Hg, Ni, Pb and Zn content of mussels based on dried weight was 11.8, 1.37, 1.98, 19.7, 0.04, 8.39, 1.56 and 116 mg/kg, respectively. Oysters thus had higher Cu and Zn content than mussels (Tables 4 and 5). Moreover, clams had higher content of Cd, Ni, and Zn than venus clams. The As content of both oysters and mussels was lower than 14.9 mg/kg (dried weight basis), which was the average As content in shellfish from various countries<sup>(19)</sup>. The average H<sub>2</sub>O% of oysters was 84.5%. Table 6 lists the average

heavy metal content of shellfish based on fresh weight. The average content of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn in shellfish was 1.55, 0.15, 0.18, 13.1, 0.004, 0.37, 0.17 and 50.3 mg/kg, respectively. Capar and Yess (1996) found that the As, Cd, Cr, Cu, Ni, Pb and Zn content in shellfish from USA was 1.95, 0.07, 0.65, 3.75, 1.0, 0.27 and 17 mg/kg, respectively<sup>(1)</sup>. Comparing the heavy metal content of shellfish from Taiwan, demonstrated that Taiwanese shellfish had higher content of Cu and Zn. Moreover, the As, Cd, Cr and Ni content were higher in Taiwanese shellfish than in Danish shellfish. The As, Cd, Cr, Hg, Ni and Pb content of Danish shellfish was 6.5, 0.095, 0.098, 0.036, 0.23 and 0.21 mg/kg, respectively<sup>(21)</sup>. Notably, the maximum permitted concentrations of As, Cd, Cr, Hg and Pb in fish and fish products in Hong Kong was 6, 2, 1, 0.5 and 6 mg/kg, respectively<sup>(6)</sup>. The action levels of Cd, Hg and Pb in Denmark was 0.5, 0.3 and 1.0 mg/kg, respectively<sup>(21)</sup>. Additionally, the maximum permitted level of Pb in shellfish in the UK was 10 mg/kg<sup>(4)</sup>. Finally, the limits of As, Pb and Cd in German oysters was 5.0, 0.5 and 1.0 mg/kg, respectively<sup>(2)</sup>. The average contents of heavy metal in Taiwanese shellfish thus were all below the above mentioned limits and standards.

Figure 2 illustrates the distribution of heavy metal content in shellfish. Figure 2 reveals that 18% of samples

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Heavy metal content in shellfish (mg/kg, on fresh weight basis)

Figure 2. Distribution of heavy metal contents in shellfish.

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contained less than 1.0 mg/kg As and 37.3% of samples had 1.0 to 1.5 mg/kg of As. Furthermore, 52.5% of the shellfish samples contained less than 0.1 mg/kg of Cd, and 32.5% of the samples had 0.1 to 0.20 mg/kg of Cd. Regarding the distribution of Cr, 67.8% of the samples had less than 0.2 mg/kg. 62.7% of the samples contained below 20 mg/kg Cu. Hg content was undetectable in nearly 75% of samples (the detection limit for Hg was 0.01 mg/kg). 70% of samples contained less than 0.2 mg/kg Pb. 42.2% of samples had Zn content less than 25 mg/kg, but 13.3% of samples had Zn content of more than 100 mg/kg.

# III. Dietary Intake of Heavy Metal from Rice and Shellfish in Taiwan

According to a food consumption survey by the Department of Health, Taiwan, daily intakes of rice and shellfish are 178 and 23.7 g/person/day<sup>(20)</sup>. Weekly heavy metal intake through rice and shellfish was calculated based on the average heavy metal content and weekly intake of these two foods. The weekly intakes of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn from rice was 1.66, 0.21, 2.08, 46.1, 0.02, 6.02, 0.21 and 305 µg/kg body weight/week (average body weight is 60 kg/person), respectively. The weekly intake of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn from shellfish consumption was 4.29, 0.41, 0.49, 36.2, 0.01, 1.02, 0.47 and 139 µg/kg body weight/week, respectively. Table 8 lists the provisional tolerable weekly intake of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn recommended by WHO/FAO and USNAS<sup>(18,24,25,26,27,28)</sup>. Table 8 reveals that weekly heavy metal intake from both rice and shellfish were did not exceed the provisional maximum weekly intake recommended by WHO/FAO and USNAS.

## CONCLUSION

This study found that Taiwanese rice samples had Cd and Hg content below the food sanitary standards of 0.50 mg/kg for Cd and 0.01 mg/kg for Hg. The average content of As, Pb, Cd and Hg in rice also was lower than the tolerance limits of 1.0 mg/kg for As, 0.3 mg/kg for Pb, 0.1 mg/kg for Cd and 0.05 mg/kg for Hg in German cereals. Notably, 68.5% of the rice samples contained less than 0.1 mg/kg As. Furthermore, 90% of the samples contained under 0.10 mg/kg Cd. The Hg content of most rice samples (94.3%) is undetectable (with the detection limit being 0.01 mg/kg). Notably, 62.5% of rice samples have Ni content between 0.10 and 0.50 mg/kg. Over 90% of the Pb content in rice samples is undetectable (with the detection limit being 0.10 mg/kg).

The investigation of heavy metals in shellfish demonstrated that oysters have the highest concentration of Zn, followed by Cu, and they have higher concentrations of Cu and Zn than do mussels. The average concentrations of heavy metals in Taiwanese shellfish were all below the limits or standards of the UK, Denmark and Germany. The measured

 Table 7. Provincial tolerable weekly intake of heavy metal

Heavy metal	Provincial tolerable weekly intake	Reference
	(µg/kg body weight/week)	
As	14	(28)
Cd	7	(27)
Cr	23.3 <sup>a</sup>	(18)
Cu	1400 <sup>b</sup>	(25)
Hg	5	(28)
Ni	35 <sup>c</sup>	(26)
Pb	25	(24)
Zn	5250 <sup>d</sup>	(25)

<sup>a</sup>Calculated based on the recommended dietary allowances (200  $\mu$ g/day) and the body weight of 60 kg.

<sup>b</sup>Calculated based on the upper limit (12 mg/day) and body weight of 60 kg.

<sup>c</sup>Calculated based on the recommended dietary (300  $\mu$ g/day) and body weight of 60 kg.

<sup>d</sup>Calculated based on the upper limit (45 mg/day) and body weight of 60 kg

**Table 8.** Weekly dietary intake of heavy metal by eating rice and shellfish ( $\mu$ g/kg body weight/week)

Foodstuff <sup>a</sup>	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Rice	1.66	0.21	2.08	46.1	0.02	6.02	0.21	305
Shellfish	4.29	0.41	0.49	36.2	0.01	1.02	0.47	139
Provincial tolerable	14	7	23.3	1400	5	35	25	5250
weekly intake								

<sup>a</sup>The daily intake of rice and shellfish is 178 and 23.7 g/person/day, respectively.

results also indicated that 55% of shellfish samples have As content between < 0.10 and 1.5 mg/kg (on a fresh weight basis). 52.5% of the shellfish samples contained less than 0.1 mg/kg of Cd. Moreover, almost 75% of samples had Hg content of below 0.01 mg/kg. Additionally, 62.7% of samples contained less than 20 mg/kg of Cu. 70% of samples contained less than 0.2 mg/kg Pb. Finally, 42.2% of samples had Zn content below 25 mg/kg, while 13.3% contained more than 100 mg/kg of Zn.

Weekly intake of heavy metal from rice and shellfish was calculated based on average heavy metal content and weekly intake of rice and shellfish. In conclusion, weekly intakes of heavy metals from rice and shellfish were below the provisional tolerable weekly intakes recommended by WHO/FAO and USNAS.

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