PEER-REVIEWED ARTICLE

Food Protection Trends, Vol 38, No. 1, p. 18–26

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An Assessment of Potential Heavy Metal Contaminants in Bivalve Shellfish from Aquaculture Zones along the Coast of New South Wales, Australia

ABSTRACT

Evaluation of shellfish aquaculture for potential contaminants is essential for consumer confidence and safety. Every three years, between 1999 and 2014, bivalve shellfish from aquaculture zones in up to 31 estuaries across 2,000 km of Australia's east coast were tested for cadmium, copper, lead, mercury, selenium and zinc. Inorganic arsenic was included in the analyses in 2002, and total arsenic was used as a screen for the inorganic form in subsequent years. Concentrations of inorganic arsenic, cadmium, lead and mercury were low and did not exceed maximum limits mandated in the Australia New Zealand Food Standards Code. As maximum limits have not been assigned to copper, selenium and zinc, accepted international dietary guidelines were used as a benchmark. Dietary exposure assessments for these elements demonstrated that shellfish from the aquaculture areas investigated do not present a food safety risk. Continued surveillance is essential, given increasing pressure on Australia's coastal resources.

INTRODUCTION

Certain elements are essential in human physiology; however, an incorrect balance or excess of certain elements in the diet can result in negative health effects. Heavy metals are of particular concern because of their ability to persist and accumulate in the environment. While heavy metals can occur naturally in the environment, human activities and run-off from urban and agricultural land use may increase their concentrations *(6, 29)*. This is particularly important when considering the growing demands on coastal resources due to increasing populations *(3)* and the ability of filter feeding bivalve shellfish to bio-accumulate contaminants, whose presence in the food chain can pose a risk to human health.

Previous studies on heavy metal concentrations in NSW shellfish have focused on individual estuaries *(5, 7, 13, 14, 26)* or a single time period *(17, 21)*. To ensure the sustainability of shellfish aquaculture, an ongoing assessment is required to determine trends in any potentially hazardous substance and to ensure that the classification of the shellfish harvest area is appropriate

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*Author for correspondence: Phone: +61 (0)2.9741.4882; Fax +61 (0)2.9741.4888; E-mail: hazel.farrell@foodauthority.nsw.gov.au to the protection of shellfish consumers. This paper presents the most comprehensive study to date of potential contaminants (inorganic arsenic (As), cadmium (Cd), copper (Cu), lead (Pb), mercury (Hg), selenium (Se) and zinc (Zn)) in aquaculture zones across 2,000 km of the New South Wales (NSW) coastline *(Fig. 1)* between 1999 and 2014. There is no decisive definition for 'heavy metals'; they are broadly classified as elements that have a specific density > 5 g cm^{-3} *(28)*. Metalloid substances such as As are often grouped with heavy metals on the basis of the interchangeable association between heavy metals and environmental toxicity *(8, 15, 28)*. While Se is a non-metal, it can be toxic at high concentrations *(1)*. Of the chemicals tested, only inorganic As, Cd, Pb and Hg have maximum limits (MLs) mandated for bivalve molluscs within the Australia New Zealand Food Standards Code (FSC) (*(12)*, *Table 1*). Cu, Se and Zn are considered as essential micronutrients as well as potential contaminants. Internationally accepted dietary guidelines *(Table 1)* were used to determine whether concentrations of these three elements represented a risk to shellfish consumers.

MATERIALS AND METHODS

Shellfish samples were collected from commercial aquaculture areas every three years between 1999 and 2014 from up to 31 estuaries along the coast of NSW, Australia *(Fig. 1)*, as part of an ongoing monitoring program *(4)*. Samples were representative of the main species harvested in each estuary. This was predominantly Sydney rock oysters (*Saccostrea glomerata*), along with some Pacific oysters (*Crassostrea gigas*), blue mussels (*Mytilus edulis*) and cockles (mainly *Anadara trapezia* and *Katelysia* spp.), where available. Oyster cultivation is predominantly in intertidal racks in plastic trays, baskets or containers, with relatively little stick culture. Mussel cultivation is via long lines. Marketable size shellfish were selected where possible. Within each harvest area, samples were collected from upstream and downstream locations and at various distances from the shoreline. Samples were pooled to provide 150–200 g of tissue (approx. 20–30 shellfish). Not all locations were sampled at every round, because of the prevailing operational status of the harvest area and time constraints. Samples from each estuary were shucked, bottled, labelled and frozen (-20°C) at the NSW Food Authority before being transported to National Association of Testing Authorities, Australia (NATA) accredited laboratories for analysis.

All samples were screened for Cd, Cu, Pb, Hg, Se and Zn. In 2002, inorganic As was added to the suite of analyses. In seafood, inorganic As is ~10% of total As *(27, 30)*. A recent analysis of Australian seafood by Stewart and

FIGURE 1. Estuaries and embayments that host currently classified shellfish aquaculture areas in New South Wales, Australia.

TABLE 1. Dietary standards and guidelines for heavy metals in molluscs

Heavy metal	Dietary standards/guidelines	Reference
As (inorganic)	1 mg kg^{-1} (ML)	FSC Standard 1.4.1 (Schedule 19) *(12)*
Cd	2 mg kg^{-1} (ML)	FSC Standard 1.4.1 (Schedule 19) *(12)*
Pb	2 mg kg^{-1} (ML)	FSC Standard 1.4.1 (Schedule 19) *(12)*
Hg	0.5 (mean) 1 (ML)	FSC Standard 1.4.1 (Schedule 19) *(12)*
Cu	0.5 mg kg^{-1} bw day^{-1} PMTDI	JECFA *(16)*
Se	0.0125 mg kg^{-1} bw day^{-1} TL	FSANZ *(10)*
Zn	1 mg kg^{-1} bw day^{-1} TL	FSANZ *(10)*

Maximum limit (ML); Body weight (bw); Provisional Maximum Tolerable Daily Intake (PMTDI); Tolerable Limit (TL), Food Standards Code (FSC); Joint FAO/WHO Expert Committee on Food Additives (JECFA); Food Standards Australia New Zealand (FSANZ).

Turnbull *(27)*, which included data from the current study, suggested that this percentage was appropriate for oysters (0.23–8.7%) and mussels (0.8–7.3%). Based on the low percentage of inorganic As to total As in seafood, total As concentrations were used to determine whether further testing for inorganic As was required between 2005 and 2014. The 10% guidance level was used to determine which samples were tested for inorganic As (i.e., as the ML for inorganic As was 1 mg kg^{-1}, shellfish samples with results > 10 mg kg^{-1} total As underwent further testing for inorganic As).

Each sample was homogenized and freeze-dried prior to analysis to ensure homogeneity of the metal components in the sample. Analysis of samples between 1999 and 2008 was performed through the laboratory contracted through the National Residue Survey (NRS). Analyses were performed by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) and/or Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). Samples collected during 2011 and 2014 were analyzed by the laboratory Advanced Analytical Australia Pty. Ltd. The methods used during 2011 and 2014 were screened by ICP-OES and/or Cold Vapor-Atomic Absorption (CVAAS).

Concentrations of inorganic As, Cd, Pb and Hg were compared to MLs specified in Standard 1.4.1, Schedule 19 of the FSC *(Table 1)*. As MLs were not specified in the FSC for Cu, Se and Zn, dietary exposure assessments were performed on the basis of internationally accepted dietary guidelines *(Table 1)*. For the 2+ age group population (body weight (bw) = 70 kg), Cu, Se and Zn were expressed as a percentage of their daily intake (DI) guidelines according to:

$$\%\,DI = \frac{MC \times C}{bw} \times \frac{100}{DI} \quad (2)$$

Three levels of mollusc consumption (*MC*, kg day^{-1}) were evaluated *(2)*: mean mollusc consumption (consumers and non-consumers (0.0005 kg day^{-1}), and median (0.063 kg day^{-1}) and 90th percentile (0.146 kg day^{-1}) mollusc consumption for consumers only (as shellfish may be consumed in larger proportions by certain groups, e.g., shellfish farmers and their families). In the formula, C refers to the concentration (mg kg^{-1}) of each element. The use of the median concentration for a particular contaminant is accepted as best practice for chronic dietary exposure assessments *(16)*. As a conservative approach, the maximum concentration for each contaminant was also assessed.

The amount of shellfish required to exceed the DI for Cu, Se and Zn *(Table 1)* was estimated according to:

$$\text{Amount of shellfish to exceed } DI = \frac{DI \times bw}{C} \quad (2)$$

where body weight for each age category was based on 2011–12 Australian National Nutrition and Physical Activity Survey (NNPAS) data *(2)*, and calculations were based on median and maximum concentrations, *C*, of each metal reported. These data were used to determine how many standard servings of seafood (adult: 150 g day^{-1}; children up to 6 yrs: 75g day^{-1} *(11)*) were required to exceed an acceptable daily intake *(Table 1)* for different age groups. It should be noted that the assessments did not consider exposure from other sources of heavy metals.

RESULTS AND DISCUSSION

All data were presented as mg kg^{-1} wet weight. Occasionally, sample results were reported below the limit of detection (LOD) for inorganic As (n = 2), Pb (n = 3) and Hg (n = 71). The number of samples below LOD was higher for Hg across all years of testing; however, maximum reported values for Hg (0.06 mg kg^{-1}) were considerably below the FSC's ML of 1 mg kg^{-1} (mean = 0.5 mg kg^{-1}). While the concentration of analytical results below the LOD ranged between 0 and the LOD, previous investigations of metal concentrations in seafood *(21)* have substituted 'zero' when results have been reported as not detectable (ND). This was due to the lower overall contribution of seafood to the total diet. The same approach was used in this study.

The maximum concentration of inorganic As reported was 0.4 mg kg^{-1} (2002 and 2008, *Fig. 2, Table 2*). This was less than the ML of 1 mg kg^{-1} specified in the FSC for inorganic As in molluscs. When both total and inorganic As were tested in samples from 2008 (n = 24), inorganic As was approximately 4.2% of the total As concentration *(Fig. 2)*. The maximum reported concentration of total As 12.01 mg kg^{-1} *(Table 2, Fig. 2)* was in samples collected from a harvest area in Brisbane Water *(Fig. 1)* in 2011. Based on the guidance ratio for inorganic to total As (1:10), additional samples (n = 2) were tested during 2012. Concentrations of total As were less than the maximum values reported in 2011 (1.2–1.5 mg kg^{-1}) and concentrations of inorganic As were < 0.05 mg kg^{-1}.

The presence of inorganic As was low, and total As was considered to be an appropriate screen for inorganic As. Stewart and Turnbull *(27)* noted similar concentrations for inorganic As (oysters: 0.11 mg kg^{-1}, mussels: 0.04 mg kg^{-1}) and total As (oysters: 3 mg kg^{-1}, mussels: 2.9 mg kg^{-1}) in a study that combined data from this report, and data from other Australian state shellfish control agencies (Tasmania, South Australia and Victoria). In the same report *(27)*, concentrations of inorganic As in saucer scallops exceeded the ML (n = 56, median = 1.25 mg kg^{-1} wet weight, maximum = 4.9 mg kg^{-1} wet weight, n > 1 mg kg^{-1} wet weight = 31). This should be a consideration for the NSW food regulator and the shellfish industry if the diversification of commercial aquaculture extends to the harvest of scallops.

Reported concentrations for Cd, Pb and Hg were less than the MLs specified for molluscs in the FSC *(Table 2)*. Maximum Cd concentrations were 1.37 mg kg^{-1}, which was approximately 69% of the ML. Maximum concentrations of Hg and Pb were approximately 6 and 18% of their respective MLs.

Where national health standards for Cu, Se and Zn in molluscs were not available, internationally accepted dietary guidelines were considered. Based on the median concentration of Cu (15.7 mg kg^{-1}), dietary exposure estimates for consumption of shellfish were between 0.02 (general population) and 6.55% (high-level consumers) of the provisional maximum tolerable daily intake (PMTDI) (0.5 mg kg^{-1} bw day^{-1}) *(Table 3)*. For the maximum concentration of Cu (218.42 mg kg^{-1}), dietary exposure estimates ranged from 0.31 to 91.11% of the PMTDI *(Table 3)*. Across all age groups examined, between 7.6 and 16.6 standard seafood servings (median concentration, approximately 15–33 dozen oysters) and 0.5 and 1.2 standard seafood servings (maximum concentration, approximately 1–2 dozen oysters) would have to be consumed in order to exceed the PMTDI *(Table 4)*. Dietary exposure assessments, based on the median concentration of Se (0.49 mg kg^{-1}) were estimated to be between 0.03 and 8.18% of the tolerable limit (TL) (0.0125 mg kg^{-1} bw day^{-1}) *(Table 3)*. At this concentration, between 6.5 and 13.3 standard servings of seafood (approximately 7–27 dozen oysters) would have to be consumed each day in order to exceed the TL *(Table 4)*. Based on the maximum concentration of Se (4.1 mg kg^{-1}), dietary exposure was estimated to be between 0.2 and 68.4% of the TL *(Table 3)*, which corresponded to between

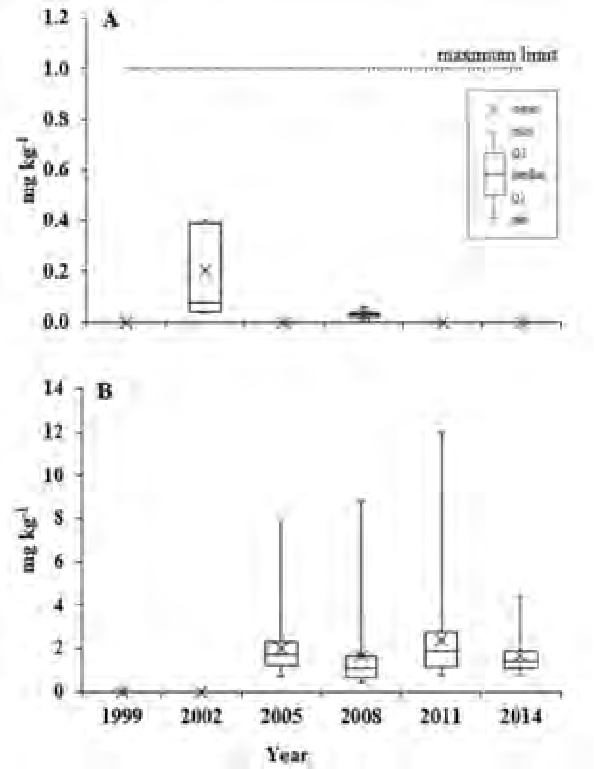

FIGURE 2. (A) Inorganic and (B) Total As concentrations in shellfish tissue tested triennially in classified New South Wales shellfish aquaculture areas (1999–2014). The ML of 1 mg kg^{-1} specified by the FSC for inorganic arsenic in mollusc tissue is represented by a dashed line. Note: differing scales on y-axes.

TABLE 2. Summary statistics for heavy metals tested in NSW shellfish aquaculture areas between 1999 and 2014

		Mean	Median	Min.	Max.	90th Percentile	99th Percentile	Standard 1.4.1 (Schedule 19)
	n	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	ML mg kg^{-1}
As (inorganic)*	62	0.13	0.04	0	0.40	0.39	0.39	1
As (total)^	182	1.93	1.42	0.40	12.01	3.54	8.07	N/A
Cd	305	0.31	0.24	0.08	1.37	0.57	1.30	2
Pb	305	0.05	0.04	0	0.35	0.09	0.17	2
Hg	305	0.01	0.04	0	0.06	0.02	0.04	1 (0.5 mean)
Cu	305	19.47	15.70	0.36	218.42	36.40	65.94	N/A
Se	305	0.55	0.49	0.12	4.10	0.89	1.23	N/A
Zn	305	248.98	230.00	8.40	1333.00	413.00	726.60	N/A

*Inorganic arsenic was measured during 2002 and 2008 only; ^Total arsenic was measured between 2005 and 2014, Maximum limit (ML), Not applicable (N/A).

TABLE 3. Dietary exposure assessment for Cu, Se and Zn for the 2+ years population age group expressed as a percentage of the daily dietary guidelines

Heavy Metal	Concentration	General population	Median consumers	90th Percentile consumers
Cu	Median (15.70 mg kg^{-1})	0.02%	2.83%	6.55%
	Max. (218.42 mg kg^{-1})	0.31%	39.32%	91.11%
Se	Median (0.49 mg kg^{-1})	0.03%	3.53%	8.18%
	Max. (4.1 mg kg^{-1})	0.20%	29.50%	68.40%
Zn	Median (230 mg kg^{-1})	0.16%	20.70%	47.97%
	Max. (1330 mg kg^{-1})	0.95%	119.97%	278.03%

0.8 and 1.6 standard servings of seafood (approximately 1–3 dozen oysters) being consumed each day in order to exceed the TL *(Table 4)*. For the median concentration of Zn (230 mg kg^{-1}), dietary exposure estimates were between 0.16 (general population) and 47.97% (high-level consumers) of the TL (1 mg kg^{-1} bw day^{-1}) *(Table 3)*. Based on the maximum concentration of Cu (1,330 mg kg^{-1}), dietary exposure estimates ranged between 0.95 and 278% of the TL *(Table 3)*. Across all age groups examined, between 1 and 2 standard seafood servings (median concentration, approximately 2–4 dozen oysters) and 0.2 and 0.4 standard seafood servings (maximum concentration, approximately < 1 dozen oysters) would have to be consumed in order to exceed the PMTDI *(Table 4)*.

Some variability was observed in metal concentrations between sampling years. This was to be expected, particularly as the samples were collected at different times throughout each sampling year. Because of the paucity of coincidental environmental data, this variability was not explored in detail; however, elevated results were not consistent. For Cu, Se and Zn, the maximum values observed during the study were reported from the same harvest area in Brisbane

TABLE 4. Estimate of number of standard servings of seafood required per day to exceed the daily intake guidance values for Cu, Se and Zn

		Age group/Body weight (kg)				
Heavy Metal	**Concentration**	**2–6 yrs/ 19**	**7–12 yrs/ 36**	**13–17 yrs/ 62**	**18+ yrs/ 78**	**2+ yrs/ 70**
Cu	Median (15.70 mg kg^{-1})	8.1	7.6	13.2	16.6	14.9
	Max. (218.42 mg kg^{-1})	0.6	0.5	0.9	1.2	1.1
Se	Median (0.49 mg kg^{-1})	6.5	6.1	10.5	13.3	11.9
	Max. (4.1 mg kg^{-1})	0.8	0.7	1.3	1.6	1.4
Zn	Median (230 mg kg^{-1})	1.1	1.0	1.8	2.3	2.0
	Max. (1330 mg kg^{-1})	0.2	0.2	0.3	0.4	0.4

Note: A standard serving of seafood = 150 g day^{-1} and 75 g day^{-1} for adults and children (up to 6 yrs), respectively *(11)*.
These serving sizes are equivalent to approximately 2 dozen (150 g of shellfish tissue) and 1 dozen (75 g of shellfish tissue) oysters.

Water *(Fig. 1)* during 2011. This prompted retesting of this location during 2012, with findings of reduced concentrations of each metal (Cu: 16.96–28.14 mg kg^{-1}, Se: 0.54–0.66 mg kg^{-1}, Zn: 112.36–192.96 mg kg^{-1}). These results may have been due to increased sediment suspension caused by strong winds and currents prior to sample collection, and seasonal variability may have also contributed. All oysters tested during this study were sampled directly from the harvest area (i.e., no depuration) prior to analysis. Depending on the harvest area classification, shellfish stock may be subject to mandatory depuration prior to sale. This process purges the particulate matter recently filtered from the estuarine environment, which could also contribute to elevated results.

The use of maximum reported concentrations in the dietary exposure assessments was a conservative approach, as the 90th and 99th percentile values were considerably lower for Cu, Se and Zn *(Table 2)*. Additionally, when considering maximum concentrations, the lower number of standard serves needed to exceed the recommended dietary guidelines were due to the smaller body weight of the age category groups below 18 yrs. Young children generally tend to eat more food per kilogram of body weight than older children or adults, and should be considered separately in dietary exposure studies. However, these age groups, particularly the 2–6 year age group, are unlikely to eat large amounts of molluscs *(2)*.

As well as being considered potential contaminants, Cu, Se and Zn are also essential elements for correct human physiological processes. Based on the current study, these elements are not considered to be a risk to NSW shellfish consumers. Toxicity from Cu is uncommon because of a number of body systems that regulate Cu levels *(9, 19)*. While Hg does not appear to be a contamination issue in NSW shellfish harvest areas, diets rich in Se can inhibit methylmercury toxicity *(24)*. Intoxication from Zn is rare, and the negative harmful effects of excessive Zn consumption appear to be from supplementary intake rather than from naturally occurring Zn in food *(10)*.

While heavy metals occur naturally in the environment, anthropogenic influences from diffuse and point pollution sources, run-off and stormwater from urbanized areas, agricultural land use, mining and other catchment disturbances and atmospheric deposition can contribute to the heavy metal load in estuarine settings *(22)*. As filter feeders, bivalve shellfish can accumulate potential contaminants from their environment as suspended organic (food) and inorganic particles as well as those dissolved in the surrounding solution *(6, 23, 26)*. Illnesses related to metal contamination in Australian seafood are extremely rare. A recent risk assessment of the NSW Food Authority's Seafood Safety Scheme *(20)* reported that the likelihood of heavy metal contamination from seafood was low. There are potential health concerns with mercury contamination for certain groups, and pregnant and breastfeeding women, women planning a pregnancy, and children up to 6 years to are advised to avoid eating certain fish and to limit portion sizes *(20)*. From the available literature, this is the largest survey of heavy metals in bivalve shellfish conducted along the NSW coast of Australia, from the perspectives of both time and geographic extent. Several studies have been carried out on heavy metal concentrations in NSW shellfish

TABLE 5. Summary of mean concentrations of heavy metals tested in shellfish tissue from NSW Food Authority data (1999–2014, this report) and historical studies of heavy metal concentrations in shellfish sampled in NSW. (Notes: *data reported in dry weight concentration, moisture content of 85% assumed for conversion to wet weight concentration; $^{\$}$ values estimated from graphed results; BDL = below detection limit)

		Heavy metal (Mean mg kg^{-1})							
Species & NSW location	***n***	**As (inorganic)**	**As (total)**	**Cd**	**Cu**	**Pb**	**Hg**	**Se**	**Zn**
S. glomerata, with some *C. gigas*, *M. edilus* and cockles (this report)	62–305	0.13	1.93	0.31	19.47	0.05	0.01	0.55	248.98
Crassostrea commercialis = *Saccostrea cucullata*, NSW estuaries *(17)*	49		0.30	0.20	20.00	0.80			277.00
Mollusc species (Sydney rock oysters, pipis, squid, octopus, cuttlefish and calamari), NSW *(21)*	23–27	0.16	5.59	0.17	8.05	0.02	0.02	0.35	
Saccostrea commercialis, Hawkesbury River & Brisbane Water *(14)*$^{\$}$	42		0.5–2.75	0.1–0.26	20–83	0.18–0.6		0.5–1.12	180–690
S. commercialis, wild oysters, NSW estuaries *(25)*	27		2.70	0.90	30.30	0.14	0.01	0.57	414.2
S. commercialis, Woolooware Bay (1999) *(26)**	20		1.50	0.21	25.50	0.06			390.00
S. commercialis, Jervis Bay (1997) *(26)**	76			0.11	3.30	0.07			152.25
S. commercialis, Clyde River (1994) *(26)**	20			0.15	9.60	0.09			232.05
S. commercialis, wild oysters, Botany Bay *(7)*	8				14–93	0.3–0.7			555–1970
S. commercialis, Botany Bay *(5)*	5–11				12–95				440–970
Pinctada imbricata, Port Stephens *(13)*	20		2.73	1.61	0.15	BDL		0.16	145.26

previously *(Table 5)* with samples typically collected in a single time frame or year. In terms of geographic distribution, the studies by Mackay et al. *(17)*, Scanes and Roach *(25)* and NSW Health *(21)* were the widest ranging, with samples collected across NSW. Other studies focused on individual estuaries (Botany Bay/Georges River: Batley et al. *(5)*, Brown and McPherson *(7)*, Spooner et al. *(26)*; Jervis Bay: Spooner et al. *(26)*; Clyde River: Spooner et al. *(26)*; Port Stephens: Gifford et al. *(13)*; Hawkesbury River and Brisbane Water: Hardiman and Pearson *(14)*). The same suite of metals in the present study was not available for comparison in every past study, and the surveys by Brown and McPherson *(7)* and Scanes and Roach *(25)* were based on sampling of wild oysters, rather than oysters obtained directly from commercial harvest areas.

The majority of heavy metal results reported in this study were comparable to results in historical reports of heavy metals in NSW shellfish. Variations in metal concentrations to those reported in the study of Gifford et al. *(13)* may be due to the smaller sample size and/or different species (Pinctada imbricata, pearl oyster). The mean concentration of Cu during the current study was higher than that recorded by NSW Health *(21)*, but similar to the concentration reported Mackay et al. *(17)*. The difference may be due to the large variety of mollusc species sampled by NSW Health *(21)* (Sydney rock oysters, pipis, squid, octopus, cuttlefish and calamari) compared to Mackay et al. *(17)* and this study, which focused primarily on oysters. Nielsen and Nathan *(18)* found that *C. gigas* had a greater tendency to accumulate Cu than other molluscs. The triennial data in the current study found the lowest concentrations of Cu (< 1 mg kg^{-1}) in cockle and mussel samples.

CONCLUSIONS

Shellfish have long been used as biological indicators of contamination *(23)*, and their ability to accumulate potential contaminants can give an indication of the overall health of a water body. This study provides a long-term assessment of potential metal contaminants over a fifteen-year period. The concentrations of heavy metals in shellfish farmed along the NSW coast were generally low. All samples tested in the current study were found to be compliant with the FSC, with maximum concentrations of inorganic As, Cd, Pb and Hg being less than the MLs specified for bivalve molluscs. The dietary assessments for Cu, Se and Zn demonstrated that these elements did not represent a food safety risk. Further investigation into concentrations of these elements pre- and post-depuration may explain the elevated results observed in certain harvest areas. Ongoing monitoring is necessary to ensure that shellfish consumers are protected from any changing trends in potential chemical contaminants.

ACKNOWLEDGMENTS

The authors thank local program coordinators from NSW shellfish harvest areas for their assistance with sample collection, acknowledge Amanda Cassidy (NSW Food Authority) for helping with sample preparation, and thank Alison Imlay (NSW Food Authority) for important feedback on the revised manuscript. The authors appreciate the constructive comments of three anonymous reviewers.

REFERENCES

1. Agency for Toxic Substances and Disease Registry. 2003. Public health statement for selenium. Available at https://www.atsdr.cdc.gov/phs/phs.asp?id=151&tid=28. Accessed 16 May 2017.
2. Anonymous, 2015. Preliminary dietary exposure assessment — commercial oysters — Tilligerry Creek and Fullerton Cove, Williamtown NSW. Available at http://www.epa. nsw.gov.au/resources/epa/152670-preliminary-assessment-commercial-oysters-211015.pdf. Accessed 16 May 2017.
3. Australian Bureau of Statistics. 2016. Australian demographic statistics, Sep 2016. Available at http://www.abs.gov.au/ausstats/abs@.nsf/mf/3101.0. Accessed 16 May 2017.
4. Australian Shellfish Quality Assurance Advisory Committee. 2016. The Australian Shellfish Quality Assurance Program Operations manual. Available at http://safefish.com.au/wp-content/uploads/2016/07/Australian-Shellfish-Quality-Assurance-Program-Operations-Manual-2016.pdf. Accessed 16 May 2017.
5. Batley, G. E., M. S. Scammell, and C. I. Brockbank. 1992. The impact of the banning of tributyltin-based antifouling paints on the Sydney rock oyster, *Saccostrea commercialis. Sci. Total Environ.* 122:301–314.
6. Birch, G. F. 1996. Sediment-bound metallic contaminants in Sydney's estuaries and adjacent offshore, Australia. *Estuar. Coast. Shelf Sci.* 42:31–44.
7. Brown, K. R., and R. G. McPherson. 1992. Concentrations of copper, zinc and lead in the Sydney rock oyster, *Saccostrea commercialis* (Iredale and Roughley) from the Georges River, New South Wales *Sci. Total Environ.* 126:27–33.
8. Duffus, J. H. 2002. "Heavy metals" a meaningless term? (IUPAC Technical Report). *Pure App. Chem.* 74:793–807.
9. European Food Safety Authority. 2015. Panel on dietetic products, nutrition and allergies: scientific opinion on dietary reference values for copper. *EFSA Journal.* 13:4253.
10. Food Standards Australia New Zealand. 2011. 23rd Australian total diet survey. Available at http://www.foodstandards.gov.au/publications/Pages/23rdaustraliantotald5367.aspx. Accessed 16 May 2017.
11. Food Standards Australia New Zealand. 2011. Mercury in fish. Available at http://www.foodstandards.gov.au/consumer/chemicals/mercury/Pages/default.aspx. Accessed 16 May 2017.
12. Food Standards Australia New Zealand. 2016. Australia New Zealand Food Standards Code. Available at http://www.foodstandards.gov.au/code/Pages/default.aspx. Accessed 16 May 2017.
13. Gifford, S., H. Dunstan, W. O'Connor, and G. R. Macfarlane. 2005. Quantification of in situ nutrient and heavy metal remediation by a small pearl oyster *(Pinctada imbricata)* farm at Port Stephens, Australia. *Marine Poll. Bull.* 50:417–422.
14. Hardiman, S., and B. Pearson. 1995. Heavy metals, TBT and DDT in the Sydney rock oyster *(Saccostrea commercialis)* sampled from the Hawkesbury River estuary, NSW, Australia. *Marine Poll. Bull.* 30:563–567.
15. Järup, L. 2003. Hazards of heavy metal contamination. *Br. Med. Bull.* 68:167–182.
16. Joint Expert Committee on Food Additives. 2012. Joint FAO/WHO expert meeting on dietary exposure assessment methodologies for residues of veterinary drugs: final report including report of stakeholder meeting. Available at http://www.fao. org/fileadmin/user_ upload/agns/pdf/jecfa/Dietary_ Exposure_Assessment_ Methodologies_ for_Residues_of_Veterinary_Drugs.pdf. Accessed 16 May 2017.
17. Mackay, N. J., R. J. Williams, J. L. Kacprzac, M. N. Kazacos, A. J. Collins, and E. H. Auty. 1975. Heavy metals in cultivated oysters (*Crassostrea commercialis = Saccostrea cucullata*) from estuaries of New South Wales. Mar. Freshwater Res. 26:31–46.
18. Nielsen, S. A., and A. Nathan. 1975. Heavy metal levels in New Zealand molluscs. *New Zeal. J. Mar. Fresh.* 9:467–481.

19. NPI. 2014. Copper and compounds. National Pollution Inventory. Available at: http://www.npi.gov.au/resource/copper-and-compounds. Accessed 16 May 2017.
20. NSW Food Authority. 2017. Metal contamination of major NSW fish species available for human consumption. NSW Food Authority, Australia. Available at http://www.foodauthority.nsw.gov.au/_Documents/scienceandtechnical/risk_assessment_seafood_safety_scheme.pdf. Accessed 7 August 2017.
21. NSW Health. 2001. Metal contamination of major NSW fish species available for human consumption. NSW Health, Australia.
22. OzCoasts. 2015. Metal Contaminants. Available at: http://www.ozcoasts.gov.au/indicators/metal_contaminants.jsp. Accessed 7 August 2017.
23. Phillips, D. J. H. 1977. The use of biological indicator organisms to monitor trace metal pollution in marine and estuarine environments—a review. *Environ. Pollut.* 13:281–317.
24. Ralston, N. V. C., and L. J. Raymond. 2010. Dietary selenium's protective effects against methylmercury toxicity. *Toxicology* 278:112–123.
25. Scanes, P. R., and A. C. Roach. 1999. Determining 'natural background' concentrations of trace metals in oysters from New South Wales, Australia. *Environ. Pollut.* 105:437–446.
26. Spooner, D. R., W. Maher, and N. Otway. 2003. Trace metal concentrations in sediments and oysters of Botany Bay, NSW, Australia. *Arch. Environ. Contam. Toxicol.* 45:92–101.
27. Stewart, I., and A. Turnbull. 2015. Arsenic in Australian seafood: a review and analysis of monitoring data 2000–2013. Available at http://safefish.com.au/wp-content/uploads/2015/ 12/Monitoring-Seafood-for-Arsenic-in-Australia-December-2015.pdf. Accessed 16 May 2017.
28. Tchounwou, P. B., C. G. Yedjou, A. K. Patlolla, and D. J. Sutton. 2012. Heavy metal toxicity and the environment, p. 133–164. *In* A. Luch (ed.), Molecular, Clinical and Environmental Toxicology, vol. 3. Environmental Toxicology. Springer Basel.
29. United States Environmental Protection Agency. 1999. Background report on fertilizer use, contaminants and regulations. Available at https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=20001KEV.TXT. Accessed 16 May 2017.
30. United States Food and Drug Administration. 1993. Guidance document for arsenic in shellfish. Available at http://www.speciation.net/Database/Links/US-FDA-Guidance-Document-for-Arsenic-in-Shellfish-;i762. Accessed 4 August 2017.