Heavy Metals in Fish and Shellfish

2012 SURVEY

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AQUATIC RESEARCH & SCIENCE COMMUNICATION CONSULTANTS

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HOW DO HEAVY METALS GET INTO OUR RIVERS AND ESTUARIES?

Heavy metals such as cadmium and lead, and metaloids like arsenic, are found naturally in the environment. They are stable and cannot be degraded or destroyed, so they tend to accumulate in soils, water, and the atmosphere. We absorb tiny amounts of some heavy metals from our food, drinking water, and the air. These very low levels generally have no adverse effect, and in some cases can be beneficial—for example tiny amounts of zinc and copper are essential to maintain the metabolism of the human body. However, human activities from industry and run-off from urban and agricultural landuse increase the concentrations of these metals in the environment, potentially to levels which could have adverse effects on humans and animals. Small children and infants are more susceptible to ingesting high levels of heavy metals as they consume more food per kilogram of body weight than adults. The toxic effects of certain heavy metals can be particularly detrimental to children's developing organs, especially the brain.

Many heavy metals enter rivers in run-off from roads, factories and agricultural land, and are usually directed to rivers through the stormwater network. Once in the rivers they can accumulate in the sediment. Eventually they get transported down-river to an estuary, which traps the river sediment and thus accumulates metal contaminants. This means that the sediment in rivers and estuaries can have high contamination loads of heavy metals. The metal concentrations vary by site depending on where contaminated sediment is accumulating.

In general marine and freshwater organisms accumulate contaminants from their environment and have been used extensively to monitor heavy metal pollution. Shellfish feed by filtering particles out of the water and often accumulate contaminants. This can have a direct impact on our health if we eat shellfish that have high heavy metal concentrations (e.g., above the safe limits set in the Australia New Zealand Food Standards Code (FSANZ, 2008)) or that are contaminated

by bacteria or viruses. Many signs have been erected around the Avon-Heathcote Estuary/Ihutai warning the public about eating shellfish due to the potential for contamination from the discharge of treated sewage (which ceased in March 2010) and stormwater inputs. Estuary and freshwater fish may also accumulate heavy metals, potentially making them unsafe to eat. Lead, mercury, and cadmium can be present in fish naturally at low levels, or at higher levels as a result of pollution. Mercury also bio-accumulates, meaning that animals further up the food-chain also accumulate the mercury in the smaller animals that they eat.

Christchurch, Canterb

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HEAVY METALS TESTED

The Food Standards Code for Australia and New Zealand (FSANZ, 2008), has set maximum levels for the heavy metals mercury, lead, cadmium and arsenic in our food. These limits are designed to ensure public health and safety when eating.

Hg MERCURY CONSUMING PROPERTY

Mercury occurs naturally in the environment but can also be released into the atmosphere through industrial pollution. It can be transported over large distances and as it has a long life can accumulate in the environment when deposited into surface waters and soils. It is present in fish and seafood products mostly as methylmercury (ENHIS, 2007). Methylmercury accumulates as smaller animals are eaten by bigger animals, so animals higher up the food chain tend to have the highest levels. High amounts of mercury can damage our kidneys and central nervous system which can cause memory loss, slurred speech, hearing loss, lack of coordination, loss of sensation in fingers and toes, reproductive problems, coma, and possibly death (Vannoort & Thompson, 2006). The developing brain of a foetus is especially sensitive.

CADMIUM Cd

Cadmium occurs naturally in low levels in the environment and is also used in batteries, pigments, and metal coatings. Volcanic activity, industrial processes such as smelting or electroplating, and the addition of fertilisers can increase the concentration of cadmium in the environment. Shellfish can also be high in cadmium (Gray *et al*., 2005; WHO, 1992). Long-term or high dose exposure to cadmium can cause kidney failure and softening of bones (Vannoort & Thomson, 2006), and high levels of cadmium have been linked to prostate cancer (Gray *et al*., 2005).

LEAD

Lead is used in batteries, solder, ammunition, devices to shield x-rays, and is found in most consumer electronic items. Most exposure to humans is due to pollution, particularly from lead-based paint and leaded fuel, both of which are no-longer used in New Zealand.

Lead can build up in the body and affects the nervous system, reproductive system, and kidneys. Lead can be stored in bones without harm but if calcium intake increases, the lead will be released from the bone. Children and babies are particularly at risk from damage to their central nervous system, which can cause learning difficulties and behavioural changes. In New Zealand the estimated dietary exposure to lead has been decreasing over time and in general our weekly exposure to lead via our diet is under the guidelines developed by the World Health Organisation (WHO, 2000).

As ARSENIC

Arsenic is a naturally occurring element that is common in soils, water, and living organisms. In New Zealand arsenic levels in the environment can increase as a result of mining, geothermal production, treated timber, and erosion caused by intensive land use.

Fish and seafood can accumulate considerable amounts of organic arsenic from their environment, but most foods contain tiny levels of organic arsenic and occasional consumption is not a health concern.

*Inorganic arsenic is estimated to be 10% of total arsenic (USFDA 1993).

An acute high level exposure to arsenic can lead to vomiting, diarrhoea, anaemia, liver damage, and death. Long term (chronic) exposure is thought to be linked to skin disease, hypertension, some forms of diabetes, and cancer (Centeno *et al*. 2005). Arsenic is present in our food in different chemical forms, but inorganic arsenic is more toxic than organic arsenic. Most arsenic in our diet is present in the less toxic organic form (for example fish and shellfish mainly accumulate organic arsenic from their environment; WHO, 2011), and most of this leaves the human body within several days. There is no regulatory limit for total arsenic in fish or shellfish. However, it is difficult to reliably measure the forms of arsenic that are present, so many surveys of arsenic content measure total arsenic levels.

WHERE WE SAMPLED

Estuary fish (sand founder and yelloweye mullet) were collected within the estuary from near the former discharge point of the Bromley Wastewater Treatment Plant (WTP) and from the western side of the Southshore spit. The WTP discharge was operating from the 1970s through to March 2010, with treated wastewater now being discharged directly to the ocean via a 3.2 km pipe out from Southshore Beach. Cockles were collected in these two areas as well as at the southern end of the causeway by Beachville Road, which is a popular shellfish gathering site. Pipi were collected near the end of the Brighton Spit and close to the estuary mouth, while estuary shrimp were collected from the southeastern end of McCormacks Bay. Shortfin eels were collected in the Avon River downstream of Anzac Drive, and in the Heathcote River just downstream of Opawa Road. Whitebait were collected at popular whitebaiting locations. For the Avon River this was opposite Brooker Avenue, while in the Heathcote River this was in Opawa, downstream of Brougham Street. Figure 1 shows these locations.

FIGURE 1: Locations where fish, shellfish and shrimp were collected in the 2012 survey.

Cockles

Shrimp Mullet Flounder Whiteba Eels

HOW WE SAMPLED

SHELLFISH

Cockles (*Austrovenus stutchburyi*) and pipi (*Paphies australis*) were collected at low tide by hand; pipi on the 23 February and cockles on the 6 March 2012. The shellfish were kept cool with ice packs, their length measured, and then delivered live to Hill Laboratories for heavy metal testing. Ten replicate samples per site were collected. For cockles each sample was made up of three specimens, while for the smaller pipi seven specimens were needed per sample. Each sample was tested by the laboratory for mercury, and five samples per site for arsenic, lead, and cadmium.

ESTUARY FISH AND SHRIMP

Sand flounder (*Rhombosolea plebeia*) and yelloweye mullet (*Aldrichetta forsteri*) were collected from the two estuary fish sites over several days in February and March 2012. Sand flounder were caught using a rigid frame benthic drag net (mesh size 25 mm) that was set and dragged behind the boat. Half a dozen drags per site were needed to capture the required number of fish. A fine mesh (38 mm) gill net was used to catch yelloweye mullet. Set netting is no longer allowed in the estuary and so the gill net was instead deployed for less than ten minutes at a time, with the boat and burley used to drive or attract fish into the net. This was supplemented by fishing rods with six hook herring jigs to capture mullet.

At each site ten fish of each type were placed on ice, anaesthetised and measured in the lab, and delivered to Hill Laboratories for testing. Ten fish of each type were analysed for mercury and five for arsenic and lead. The small size of the flounder meant that two fish had to be combined to make a single sample with sufficient flesh for testing in two samples from the Discharge site and one from the Southshore site.

Shrimp (*Palaemon affinis*) were caught on the 4 April 2012 using a fine mesh hand net and ten samples weighing approximately 6–10 g each (wet weight) were delivered to Hill Laboratories for testing. All of these samples were tested for mercury and five samples were tested for arsenic and lead.

Yelloweye mullet

FRESHWATER FISH

Shortfin eels (*Anguilla australis*) were collected from the Heathcote River and Avon River using fyke nets that were baited and set overnight on the 20 February 2012. These nets are a series of hoops connected by mesh. Once the fish enter the inverted funnel entrance they can't find the narrow exit and are trapped. The next day the eels were anaesthetised, their length measured, and either taken to Hill Laboratories for analysis or returned live to the river if too many were caught. Mercury levels were tested in ten eels and arsenic and lead in five eels from each site.

Whitebait (most likely *Galaxias maculatus*) were collected during the whitebaiting season, in October 2011, when the tiny fish are migrating upriver after having spent six months developing in the ocean. Due to sewage inputs into the rivers from sewerage pipes damaged during the 2011 Christchurch earthquakes, whitebaiting on the Avon and Heathcote Rivers was less popular during this season. Whitebait on the Avon River were therefore caught using hand-held nets while whitebait on the Heathcote River were obtained from a whitebaiter who gave us a portion of their catch. Ten samples from each site, weighing approximately 4 g and made up of 10–12 fish, were delivered to Hill Laboratories for testing. All of these samples were tested for mercury and five samples per site were tested for arsenic and lead.

OUR FINDINGS

SHELLFISH

Where possible we collected larger shellfish; the size most likely to be collected and eaten. Cockles, however, were smaller at the Southshore and Discharge sites (average length of 38-39 mm compared to 50 mm at the Causeway site), and the pipi found after an hour of searching were not particularly large (average length of 47 mm) (Table 1).

Both pipi and cockles at all sites had levels of cadmium, lead, and mercury below the Food Standards Australia New Zealand (FSANZ, 2008) maximum allowable level set for safe consumption of shellfish (Figure 2). In fact, the average level of all three metals at each site was at least 1/10 that of the FSANZ maximum allowable metal contaminant levels. Cockles collected from the Discharge site had the lowest levels of cadmium, and mercury, but had the highest levels of lead.

Arsenic was lowest in pipi at the Estuary Mouth and cockles at the Discharge site, and highest at Southshore and Causeway sites. The FSANZ (2008) provides guidelines for levels of inorganic arsenic in shellfish (as well as in fish and shrimp). However, as this is difficult and expensive to measure accurately, most studies measure total arsenic levels instead. In America the US Food and Drug Administration (USFDA) has set maximum allowable levels for total arsenic in shellfish at 86 mg/kg (USFDA, 1993). The levels of total arsenic that we found in the estuary shellfish were much lower than this, with the highest total arsenic level being 4.9 mg/kg in cockles at Southshore, 3.9 mg/kg in cockles the Causeway site, 1.9 mg/ kg in cockles at the Discharge site, and 1.4 mg/kg in pipi near the Estuary Mouth. Thus even the highest concentration of total arsenic was at least 1/10 that of the safe consumption levels set by the USFDA. The USFDA has also conservatively set the inorganic arsenic component at 10% of total arsenic (USFDA, 1993). If we apply this rationale to our samples then the highest estimated inorganic arsenic levels would be 0.49 mg/kg; still below the FSANZ guidelines of 1 mg/kg inorganic arsenic.

TABLE 1: Average shell length (mm \pm 1 std error) of shellfish collected in March 2012.

Cockles

FISH AND SHRIMP

The size of yelloweye mullet caught in this survey (average length of 258 mm across both sites) were generally larger than those caught throughout the estuary in 2010 (Unwin & Hawke, 2011), and although there is no size or catch limit for yelloweye mullet, the larger size of fish we caught would be more desirable for consumption (Table 2). The size of sand flounder caught (average length of 89 mm across both sites) were generally similar to those caught in the low tide channels in 2010 (Unwin & Hawke, 2011), but are smaller than what would be regarded acceptable for eating (Table 2). The shortfin eels caught were of similar size between the two rivers (average length of 460 mm for Avon River and 461 mm for Heathcote River). While we retuned the largest eels to the rivers as they are an important part of the breeding population of this slow growing species, the specimens retained for analysis were still of a size that would be caught and eaten.

The levels of lead and mercury from flounder, mullet, eels, and whitebait were all well below the maximum acceptable levels for eating fish (FSANZ, 2008) (Figure 3). However, in general flounder had higher levels of lead than any other fish, with an average level of 0.16 mg/kg compared to less than 0.03 mg/kg in other fish. In contrast flounder had very low levels of mercury, as did whitebait—with both species sometimes recording levels below the detection limit of 0.01 mg/kg. Shortfin eels had the highest levels of mercury (average of 0.05 mg/kg), followed by yelloweye mullet (average of 0.03 mg/kg).

The safe limit for inorganic arsenic in fish is 2 mg/kg, so the estimated level of inorganic arsenic (i.e., 10% of total arsenic) in sand flounder (estimated average 0.102 mg/kg), whitebait (estimated average of 0.083 mg/kg), yelloweye mullet (estimated average of 0.047 mg/kg), and shortfin eels (estimated average of 0.013 mg/kg) were all below this level. Of the fish tested, sand flounder had the highest levels of total arsenic (average of

While not regularly eaten now, the *Palaemon* shrimps of the Avon-Heathcote Estuary/Ihutai were once a prized delicacy, with the 'Redcliff shrimps' cooked and sent throughout New Zealand through to the 1930s (McMurtrie & Kennedy, 2012). The levels of lead and mercury in shrimp from McCormacks Bay were well below the FSANZ maximum metal contamination levels for food—being 0.5 mg/kg for mercury (no levels set for lead) (Figure 3). Levels of lead in shrimp (average of 0.07 mg/kg), while low, were still second highest after sand flounder. Levels of mercury in shrimp (average of 0.008 mg/kg) were at similarly low levels as that found in sand flounder and whitebait.

Following the USFDA (1993) conservative estimate of inorganic arsenic being 10% of total arsenic, the estimated level of inorganic arsenic in shrimp (estimated average of 0.22 mg/kg) was also well below the FSANZ 2 mg/kg guideline for Crustacea. However, the total arsenic levels (average 2.22 mg/kg) was still more than two times that of the sampled fish (mullet, flounder, shortfin eel, whitebait), and was marginally higher than the levels found in pipi from Southshore and cockles from the Discharge site. However, total arsenic levels in cockles from the Causeway and Southshore sites (3.4 and 3.9 mg/kg) still remained higher even than shrimp.

FSANZ guidelines for unsafe levels: above 0.5 mg/kg

FIGURE 3:

0.5

 $0.4 -$

Heavy metal levels in fish and shrimp collected from the Avon-Heathcoste Estuary/Ihutai and main rivers in 2012. (Whitebait collected October 2011.)

DISCUSSION

THE CHRISTCHURCH 2011 EARTHQUAKES

On 22 February and 13 June 2011, 6.3 and 6.4 magnitude earthquakes in Christchurch caused substantial damage to the city's sewerage infrastructure. Over 60,000 m3 /day of untreated raw sewage entered the city's rivers and estuary for one month after each event, with discharges of 10-20,000 m³/day prior to sewerage repairs in November 2011. The collection of whitebait in October 2011 therefore occurred while there was still a small discharge of raw sewage into the Avon River (estimated at 8,654 m^3 /day in August 2011) and estuary (direct discharges estimated $5,643 \text{ m}^3/\text{day}$ in August 2011) as a result of the 2011 earthquakes. In contrast the rest of the fish, shrimp and cockles were collected in March-April 2011, approximately four months after all raw discharges into the City's rivers or estuary had ceased. Thus while the discharge of treated wastewater ceased in March 2010, because of the large volumes of raw sewage that entered the estuary in the nine months following the February 2011 earthquake, the current survey is more indicative of an estuary still impacted by sewage discharges.

THE INFLUENCE OF SITE LOCATION

Because shellfish are sessile (i.e., they live in the sediment and don't move around a lot) they probably provide the best opportunity to look at differences between sites. CBD (1988) found that smaller cockles had higher levels of heavy metals than larger ones, but we did not find this to be the case. In fact we found that the largest cockles had the highest levels of cadmium, but this may have been more due to site location, with the Causeway site having the largest cockles. Our results therefore imply that site location has a greater influence on heavy metal levels in cockles than size does.

Our results showed that cockles from the Causeway and Southshore sites had higher levels of cadmium, mercury and arsenic than those from the Discharge or Estuary Mouth sites, while lead was higher in cockles from the Discharge site. These patterns are similar to those found in the 2010 survey. The pattern of higher lead levels in the western side of the estuary is also the same as that found over 20 years ago by the Christchurch Drainage Board (CDB, 1988). Because the levels of cadmium, mercury, and arsenic have always been lowest at the Discharge site it is possible that these contaminants originate from the rivers or stormwater drain inputs rather than the (now decommissioned) sewage discharge. The Causeway and Southshore sites are located close to areas where a number of stormwater pipes discharge directly into the estuary (Figure 4), and so may be more exposed to the contaminants in the stormwater discharges than the Discharge site would be. This may similarly mean that the higher lead levels at the Discharge site could be an historic artefact of the discharge of treated wastewater from the City's wastewater treatment ponds, which discharged into the estuary at this point between 1958 and 2010 (McMurtrie & Kennedy, 2012). The fact that the lead levels remain high in cockles from this site despite the sewage discharge having ceased since March 2010 may indicate that there is a historic load held in the estuary sediments around this location. However,

Cockle collection site

● Stormwater discharge

FIGURE 4:

Small stormwater pipe discharges (locations provided by Environment Canterbury) into the Estuary in relation to the location of the cockle monitoring sites . It is possible the stormwater discharges near to the Causeway and Southshore sites contribute to the higher levels of cadmium, mercury, and arsenic in cockles from these sites compared to the Discharge site .

sediment testing in 2011 indicated that total lead levels in sediment at the Discharge site were little different to other areas around the estuary, with the exception of the Avon River mouth (which had higher levels; EOS Ecology, unpublished data).

For freshwater fish (whitebait and shortfin eels), the river that they were collected from made little difference to the levels of mercury or arsenic, with fish collected from both rivers having similar levels. However, lead levels were only marginally higher in both whitebait and eels caught in the Avon River. For the estuarine fish, there was also no relationship between site and heavy metal contamination, with levels in each fish species relatively similar between sites. The obvious exception was the slightly higher levels of arsenic found in sand flounder collected at the Southshore site. Given the transient nature of both types of fish (but in particular yelloweye mullet) it is unlikely that any differences would be associated with where they were caught.

Because fish move around so much it is difficult to attribute any differences in heavy metal levels to the location where the fish were caught. Although typically regarded as marine species, flounder and mullet do not just live in the sea and estuary area, but move up into the lower reaches of rivers to feed. Sand flounder will move a short distance up-river, although they stay within the tidal zone. Yeloweye mullet however, regularly move considerable distances up-river, into freshwater above the tidal zone, where they may remain and feed for several tide cycles before returning to the estuary. The whitebait caught would have spent around six months developing in the ocean before their spring migration into rivers and streams, where they will stay for 1–2 years before moving down into the tidal reaches of rivers to lay their eggs in grasses along the streambank during autumn high tides. When the young hatch they are washed out to sea to develop and will return in the next season's whitebait run. In contrast eels will typically spend most of their life in freshwater, only migrating to the sea to spawn later in their life.

View of the estuary from Kinsey Terrrace, looking towards the oxidation ponds

THE INFLUENCE OF LIFE HISTORY

Some of the differences in heavy metal loadings between the animals collected may be due to differences in life history, habitat preferences, feeding behaviour, and even how metals behave and accumulate, rather than site-specific differences.

Feeding habitats and life history patterns could influence heavy metal levels in fish. Sand founder live and feed from the estuary floor and so may be more exposed to contaminants in the sediment than other free-swimming fish such as mullet and whitebait. This could explain the higher levels of lead in flounder compared to other fish. The whitebait caught would be little more than six months old, with much of this time having been spent in the ocean where they feed on tiny zooplankton in the water. Thus heavy metal levels in whitebait could be a reflection of their time spent at sea and in the estuary as much as their time spent in the river.

The age of fish caught and their feeding habits could help explain the level of mercury in fish, as it accumulates over an animal's life time as well as up the food chain (e.g., predators also accumulate the mercury from the prey they eat). The much higher level of mercury in eels than all other animals tested may be related to their age and predatory status. The eels caught in this study could be somewhere between 14 and 22 years (as they grow very slowly and are longlived) and would feed on smaller fish as well as invertebrates. The bioaccumulation of mercury could also explain the higher levels in yelloweye mullet compared to either whitebait or sand flounder. While yelloweye mullet would not be nearly as old as the shortfin eels (the mullet were estimated to be only 1-2 years old) their age and size is still greater than that of sand flounder and whitebait, which were estimated at around six months of age.

Pipi and cockles are relatively stationary animals that live in the sediment and filter particles out of the water column. Compared to fish, they actively ingest heavy metals bound to particles (organic and inorganic), meaning that they would be more exposed to heavy metals while feeding. Shrimp also feed by stirring the sediment up and collecting very small particles of organic matter, and so they too would be exposed to the heavy metals bound to this food. Our study and other studies (FSA, 2005) have found that cockles accumulate more arsenic than fish do. This may be due to their feeding or habitat preferences, or other factors. The feeding habits of shrimp may also explain the high levels of arsenic in them compared to the fish tested.

ARE FISH AND SHELLFISH SAFE TO EAT?

Cockles, pipi, shrimp, yelloweye mullet, sand flounder, shortfin eels, and whitebait all had metal concentrations (e.g., mercury, cadmium, lead, arsenic) below the FSANZ (2008) limits and so based on heavy metal levels they are safe for consumption. However, the on-going high arsenic levels in cockles and shrimp could warrant further investigation, with testing of inorganic arsenic in cockles and shrimp to properly ascertain the relationship between total arsenic and inorganic arsenic levels.

Despite this clean bill of health, the consumption of shellfish in particular should still be cautioned. Bacteria (*E. coli*, *Salmonella*) and enteric viruses (norovirus)—which can cause vomiting, diarrhoea, and abdominal pain—are still being found in shellfish collected from the estuary as a result of faecal contamination from either human (sewage overflows) or wildlife (birds, dogs) sources. Quarterly monitoring by EOS Ecology has shown that both *E. coli* and norovirus concentrations at sites near to the Avon and Heathcote River mouth dramatically increased after the February earthquake when there were sewage overflows into the city's rivers and estuary. Bacteria (*E. coli*) concentrations increased to 16,000 MPN/100g (MPN stands for 'most probable number') and norovirus was detected at extremely high concentrations (>10,000 genome copies/g shellfish digestive tissue). While no specific microbiological guideline criteria exist for shellfish gathered for personal consumption or non-commercial purposes, the safe *E. coli* limits for commercial food set by the Australian New Zealand Food Authority in 2011 is 700 MPN/100g in bivalves (FSANZ, 2011). The levels recorded in cockles following the February 2011 earthquake were far in excess of this level and they would have been dangerous to eat raw or lightly cooked. These high levels also illustrate how responsive shellfish are to bacterial and viral contamination in water.

The discharge of raw sewage into the City's rivers and estuary **Cockles being collected from the Estuary**

ceased in November 2011, but there are still chances of further sewage discharges as the City continues to repair its badly damaged sewerage infrastructure. When these sewage overflow events occur the public are being kept informed through warning signs and warning notices on Environment Canterbury's website. The CCC still maintains their warning signs around the estuary advising against taking shellfish for consumption. Once the sewerage network is repaired it is hoped that the viral and bacterial levels in shellfish will drop.

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