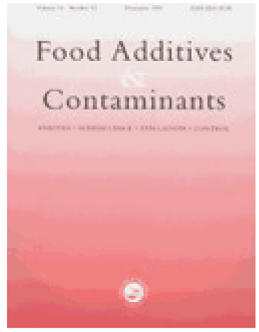
Mercury in fish: concentration versus fish-size and estimates intake dose.

Maria Maddalena Storelli, Grazia Barone, Gaetano Piscitelli, Giuseppe Marcotrigiano



Mercury in fish: concentration versus fish-size and estimates intake dose.

Journal:	*Food Additives and Contaminants*
Manuscript ID:	TFAC-2007-052.R1
Manuscript Type:	Original Research Paper
Date Submitted by the Author:	03-Apr-2007
Complete List of Authors:	Storelli, Maria Maddalena; Università degli Studi di Bari, farmaco-biologico Barone, Grazia; Università degli Studi di Bari, farmaco-biologico Piscitelli, Gaetano; Università degli Studi di Teramo, Zoologia Marcotrigiano, Giuseppe; Università degli Studi di Bari Fac. Med. Vet., Farmaco-Biologico
Methods/Techniques:	Metals - uptake, Risk assessment, Total diet, Toxicology
Additives/Contaminants:	Heavy metals - mercury
Food Types:	Fish

Mercury in fish: concentration versus fish-size and estimates of mercury intake

M.M. STORELLI[1], G. BARONE[1], G. PISCITELLI[2], & G.O. MARCOTRIGIANO[1]*

[1]*Pharmacological-Biological Department, Chemistry and Biochemistry Section, Medicine Veterinary Faculty, University of Bari, Strada prov. le per Casamassima, km, 3, 70010 Valenzano, (BA), Italy;* [2]*Department of Zoology, Faculty of Sciences, University of Bari, Via Orabona 4, 70125 Bari, Italy.*

Correspondence: G.O. Marcotrigiano. E-mail: g.o.marcotrigiano@veterinaria.uniba.it

Abstract

Total mercury concentrations were determined in different fish size classes of commercial importance such as, conger eel (*Conger conger*), starry ray (*Raja asterias*), forkbeard (*Phycis blennoides*), frostfish (*Lepidopus caudatus*), striped mullet (*Mullus barbatus*), red gurnard (*Aspitrigla cuculus*) and yellow gurnard (*Trigla lucerna*) in order to evaluate variations in consumer exposure to mercury as a function of fish consumption of a spectrum of different sizes. The highest mean levels of total mercury were detected in conger eel (0.80 $\mu g\ g^{-1}$) and in starry ray (0.75 $\mu g\ g^{-1}$). Forkbeard (0.67 $\mu g\ g^{-1}$), frostfish (0.59 $\mu g\ g^{-1}$) and striped mullet (0.55 $\mu g\ g^{-1}$) showed slightly lower levels, while red gurnard (0.33 $\mu g\ g^{-1}$) and yellow gurnard (0.22 $\mu g\ g^{-1}$) exhibited the lowest concentrations. The results of the linear regression analysis showed significant relationship between mercury concentrations and fish size for all species. Consequently, dietary consumption of larger size specimens leads to an increase in the exposure level for consumers. Understanding by consumers of all factors leading to an increase of exposure to mercury is the first step to enable them to make decisions about eating fish.

Keywords: *total mercury, fish, risk assessment, PTWI*

Introduction

Fish provide a healthy source of dietary protein, and are relatively low in cholesterol and high in omega-3 (n-3) fatty acids (National Research Council 2000). Several studies have shown that fish consumption reduces the risk of coronary heart disease, decreases mild hypertension, prevents certain cardiac arrhythmias, lowers the incidence of diabetes and appears to alleviate the symptoms of rheumatoid arthritis (Deckere *et al*. 1998, Billman *et al*. 1999, Rosenberg 2002). However, people who eat fish must balance the relative benefits from a low-fat source of protein against potential exposure to contaminants, above all mercury. Seafood consumption is, in fact, the main source of this toxin which accumulates in the human body and causes damage in many of its basic systems, particularly to the nervous system (Dey *et al*. 1999).

Literature data indicate that predator fish occupying high trophic positions and therefore have generally higher amount of mercury because this metal is particularly liable to biomagnify along marine food chains (Burger *et al*. 1992, Storelli *et al*. 1998, Brabo *et al*. 2000). Added to this other studies show differences in mercury concentrations between pelagic and benthic species (Romeo *et al*. 1999, Bustamante *et al*. 2003, Henry *et al*. 2004). Animals living in close association with sediments in which they bury and from where they mainly feed, are more exposed to eventually sediment-associated contamination than other fish. These remarks can have implications for human health.

Earlier studies have, in fact, shown a high human dietary exposure associated with the consumption of predatory fish and bottom-dwelling fish (Storelli *et al*. 2002, Storelli *et* *al*. 2005). Of critical concern is also the fish-size because generally mercury levels in fish increase with body size. This issue has been discussed with supporting data in the environmental health sciences literature (Dixon and Jones 1994, Storelli *et al*. 1998, Joiris *et al*. 1999, Storelli and Marcotrigiano 2000, Storelli and Marcotrigiano 2001) is a crucial point concerning consumer exposure. The positive relationship between fish-size and mercury levels suggests that consumers that eat larger fish might have higher exposure to mercury than those that eat smaller fish. In relation to this to address the people who eat smaller size fish might arise an effective risk reduction. In this contest we are particularly interested in examining variations in consumer exposure to mercury as a function of consumption of different size spectra of fish. Such information will be of value to those involved in risk communication. An understanding by consumers of all factors leading to an increase of exposure to mercury is the first step to enable them to make decisions about eating fish.

Materials and methods

In June-August 2005 during several trawl surveys, 100 conger eel (*Conger conger*, length range: 32.0-85.0 cm; median: 59.3 cm), 253 red gurnard (*Aspitrigla cuculus*, length range: 10.2-24.7 cm; median: 18.5 cm), 263 yellow gurnard (*Trigla lucerna,* length range: 20.4-55.5 cm; median: 35.0 cm), 464 striped mullet (*Mullus barbatus*, length range: 15.8-31.0 cm; median: 21.0 cm), 224 forkbeard (*Phycis blennoides*, length range: 12.4-49.2 cm; median: 26.5 cm), 879 frostfish (*Lepidopus caudatus*, length range: 110.0-144.0 cm; median: 128.0 cm) and 100 starry ray (*Raja asterias*, length range: 23.0-58.8 cm; median: 48.5 cm) specimens were caught in the Adriatic Sea. For each species, from the total number of specimens were formed pools within which individual fish were collected as a function of their similar size length to investigate the influence of size on mercury bioaccumulation.

From the organisms of each pool muscle tissue was removed and preserved at -25 °C until analysis. The tissues were dissected with plastic materials that were washed with HNO_3 and rinsed with distilled and deionized water, in order to avoid metal contamination. For analyses of total Hg, homogenized samples of the tissue (2 g wet weight) were digested to a transparent solution with 10 mL of the mixture H_2SO_4-HNO_3 (1:1) under reflux. The resultant solutions were then diluted to a known volume with deionized water (Official Journal of the European Communities 1990) and the total Hg concentrations were measured by atomic absorption spectrophotometry (Analyst 800 Perkin Elmer) by the cold vapour technique, after reduction by $SnCl_2$ (FIMS-100, Perkin Elmer). Acid washed glassware, analytical grade reagents and double distilled deionized water were used in the tissue analysis.

In order to check on the purity of the chemical used, a number of chemicals blanks were run; there was no evidence of any contamination in these blanks. Analytical quality control was achieved using TORT-1 Lobster Hepatopancreas (National Research Council of Canada). Replicate analyses (n=3) (Hg total 0.32±0.02 µg g^{-1} dry weight) were in the range of the certified material (Hg total 0.33±0.06 µg g^{-1} dry weight). All data were computed on a µg g^{-1} wet weight basis. Mann-Whitney *U* test was used to test significance of differences between data sets. The level of significance was set at P <

0.05.

Results and discussion

Among the different fish species examined the highest mean levels of total mercury were detected in conger eel (0.80 μg g^{-1}) and in starry ray (0.75 μg g^{-1}). Forkbeard (0.67 μg g^{-1}), frostfish (0.59 μg g^{-1}) and striped mullet (0.55 μg g^{-1}) showed levels slightly lower, while red gurnard (0.33 μg g^{-1}) and yellow gurnard (0.22 μg g^{-1}) exhibited the lowest concentrations (Table 1). Despite of mercury level variability, statistical comparisons did not reveal significant differences among various fish species, except among Triglidae and the other species ($P < 0.04$).

It is known that differences in feeding habits generally assign the species to a trophic level and species belonging to higher trophic levels are considered to contain higher mercury concentrations (Wiener and Spry 1996; Watras *et al.* 1998, Snodgrass *et al.* 2000). In our case the findings of statistical analysis led to suppose that all species analysed belonged to similar trophic levels. Published estimates of Mediterranean fish trophic levels confirmed this hypothesis being the species in question included in the same functional trophic group with the highest values corresponding to conger eel (Stergiou and Karpouzi 2002). In this picture Triglidae were an exception in that their trophic levels was the lowest. This latter information corroborated further the thesis that mercury levels in fish reflect its trophic level. Significant differences in mercury concentrations were, in fact, detected solely between Triglidae with the lowest trophic levels and the other species assigned to the highest.

Of primary importance in explaining mercury body burden of fish is also the habitat where they live. It is well known that benthic fish are species that tend to concentrate mercury to a higher degree than other organisms (Campbell 1994, Storelli *et al*. 2005), confirming the significant process of sedimentation and persistence of this metal in sea depths. For this reason the tested species were all benthic feeding fish but however, exhibiting different dietary preferences. It is clear from the above discussion that food habits as well as feeding location of fish, are factors of primary importance influencing mercury body load. However, independently from the complexity of interactions leading to different accumulation profiles among fish, a common point to all species in question was that metal concentration seemed to increase with size/age of fish suggesting that larger, older fish had higher mercury levels than smaller, younger fish. The linear regression analyses confirmed this trend being mercury concentrations positively and significantly correlated with fish length (striped mullet: $R = 0.94$, $P < 0.001$; red gurnard: $R = 0.92$, $P < 0.001$; yellow gurnard: $R = 0.96$, $P < 0.001$; starry ray: $R = 0.88$, $P < 0.001$; conger eel: $R = 0.89$, $P < 0.001$; forkbeard: $R = 0.93$, $P < 0.001$; frostfish: $R = 0.82$, $P < 0.001$).

Generally, the evaluation of the toxicological risk of the metal for humans is carried out through comparison of measured concentrations of a certain element in food with the levels imposed by law or following guidelines proposed by different international organizations. In the case of mercury, European legislation (Official Journal of the European Communities 2001) sets the maximum limit in edible parts of fish-products at

0.5 μg g^{-1} wet wt, except for some species for which it is raised to 1.0 μg g^{-1} wet wt. Generally, these latter species are either high trophic level predators, that with their considerable mercury load demonstrate the importance of biomagnification process through food chain (Monteiro and Lopes 1990, Storelli and Marcotrigiano 2001), or benthic organisms that spending a considerable time searching for food on the bottom of the sediment are particularly prone to accumulate higher amounts of mercury (Romeo *et al*. 1999, Kljakovic *et al*. 2002, Storelli *et al*. 2005). For fish analyzed in the present study total mercury concentrations should not exceed 0.5 μg g^{-1} wet wt, except for conger eel, starry ray and frostfish for which the established value is 1.0 μg g^{-1}. It is, in fact, not by chance that these latter species had a more consistent mercury load in their flesh respect to others. However, comparison of mercury mean concentrations detected in the present study with the levels imposed by law revealed that almost all fish in question (see conger eel, red and yellow gurnard, frostfish and starry ray) were suitable for human consumption with the metal concentrations under the prescribed legal limits.

Only two species, striped mullet and forkbeard, had mercury mean concentrations (striped mullet: 0.55 μg g^{-1}; forkbeard: 0.67 μg g^{-1}) slightly exceeding the standard of 0.5 μg g^{-1} wet wt established by European legislation. With these results, it is likely to conclude that the consumption of these fish by humans should be safe. However, this assertion was not corroborated by the concentrations relative to each fish-size classes. Within each species, the larger size specimens showed, in fact, concentrations exceeding, to a more or less great extent, the maximum regulatory limits.

What was observed might be particularly relevant with respect to potential risk on consumers' health. To test this hypothesis the metal intakes via dietary consumption of these fish were calculated and compared with the Provisional Tolerable Weekly Intake (PTWI) of 5 μg kg^{-1} body weight, set by the Joint Expert Committee on Food Additives (WHO 2003). The average weekly dietary intakes (0.52-1.90 μg kg^{-1} bw) calculated by taking into account mean mercury concentrations in each species and a mean weekly consumption of demersal fish of 142 g (FAO 2002), were below the fixed safe level (Table 1).

However, the purpose here was not to evaluate the exposure to mercury due to sea product consumption but rather to examine how varied the exposure according to consumption of different sized fish. In this respect, as shown in table 1, weekly mercury intakes were comprised in a wide range from 0.02 to 3.34 μg kg^{-1} body weight. As the figure indicated, mercury intake increased with consumption of larger size specimens for all species. However, the consumption of certain species, even of large size, such as yellow gurnard and red gurnard gave levels relatively small in comparison to PTWI value (25.0-27.4% of PTWI), whereas high exposure values, constituting from 49.2% to 66.8% of PTWI, were associated with consumption of largest specimens of the remained species. In particular, the highest mercury intakes resulted from the consumption of specimens of weight above 400 g of forkbeard (55.4-66.8% of PTWI), conger eel (51.0-62.4% of PTWI) and starry ray (50.2-56.8% of PTWI). These data clearly demonstrate that people that eat larger fish have higher exposure to mercury than those that eat smaller fish. This could be of concern for consumers, particularly if they repeatedly ate the largest individuals of some species. It would be, in fact, sufficient a weekly consumption of 250 g of largest specimens of these species to have a mercury intake close or that surpass the reference limit.

Potential public health risks from dietary exposure to mercury continue to be the subject of much research, regulation and debate. State and federal agencies can respond to potential health risks from mercury in fish by issuing consumption advice. For example, recently the US Food and Drug Administration (FDA 2002) has issued a series of consumption advice notes based on mercury that suggested that pregnant women should limit their fish consumption and avoid eating large predatory fish. However, the question of risk from eating fish is complicated by the positive health benefits of consuming fish. Because of this it becomes extremely important to reduce human exposure to mercury from fish consumption. The data here obtained clearly show that mercury levels vary in fish as a function of their trophic level and size. Consequently this means that the consumers could substantially reduce their exposure to mercury by eating selected species and fish of smaller size. However, such information is not helpful if the general public is unaware of this possibility. Understanding by consumers about the relationship between contaminant, fish-size, trophic level and mercury intake is crucial to enable them to be able to make better decisions about eating fish. On this basis educational programs to foster such an understanding and, thus, changes in the fish species and size consumed would constitute an useful tool to reduce human exposure to this neurotoxin.

References

Billman, G. E., Kang, J. X., and Leaf, A., 1999, Prevention of sudden cardiac death by dietary pure omega-3 polyunsaturated fatty acids in dogs. *Circulation*, **99**, 2452-2457.

Burger, J., Cooper, K., and Gochfeld, M., 1992, Exposure assessment for heavy metal ingestion from a sport fish in Puerto Rico: estimating risk for local fisherman. *Journal of Toxicology and Environmental Health*, **36**, 355-365.

Bustamante, P., Bocher, P., Chérel, Y., Miramand, P., and Caurant, F., 2003, Distribution of trace elements in tissues of benthic and pelagic fish from the Kerguelen Islands. *Science of the Total Environment*, **313**, 25-39.

Brabo, E., Santos, E., Jesus, I. M., Mascarenhas A. F. S., and Faial, K. F., 2000, Mercury contamination of fish and exposures of an indigenous community in Parà State, Brazil. *Environmental Research*, **84**, 197-203.

Campbell, K. R., 1994, Concentrations of heavy metals associated with urban runoff in fish living in stormwater treatment Ponds. *Archives of Environmental Contamination and Toxicology*, **27**, 352-356.

Deckere. Ede., Korver, O., Verschuren, P. M., and Katan, M. D., 1998, Health aspects of fish and n-3 polyunsaturated fatty acids from plant and marine origin. *European Journal of Clinical Nutrition*, **52**, 749-753.

Dey, P. M., Gochfeld, M., and Reuhl, K. R., 1999, Developmental methylmercury administration alters cerebellar PSA-NCAM expression and Golgi sialyltransferase activity. *Brain Research*, **845**, 139-151.

Dixon, R., and Jones, B., 1994, Mercury concentrations in stomach contents and muscle of five fish species from the north east coast of England. *Marine Pollution Bulletin*, **28**, 741-745.

FAO, 2002, Statistical databases. Available: http://apps.fao.org.

Food and Drug Administration, 2002, Consumer advisory: An important message for pregnant women and women of childbearing age who may become pregnant about the risks of mercury in fish. Washington: FDA, 2001. Available at: http://www.cfsan.fda.gov/~lrd/tphgfish.html. Accessed 2002 Aug 1.

Henry, F., Amara, R., Courcot, L., Lacouture, D., and Bertho, M-L., 2004, Heavy metals in four fish species from the French coast of the Eastern English Channel and Southern Bight of the North Sea. *Environment International*, **30**, 675-683.

Joiris, C. R., Holsbeek, L., and Moatemri, N. L., 1999, Total and methylmercury in sardines *Sardinella aurita* and *Sardina pilchardus* from Tunisia. *Marine Pollution Bulletin*, **38**, 188-192.

Kljakovic, G. Z., Zvonaric, T,, Vrgoc, N., Odzak, N., and Baric, A., 2002, Cadmium and lead in selected tissues of two commercially important fish species from the Adriatic Sea. *Water Research*, **36**, 5023-5028.

Monteiro, L. R., and Lopes, H. D., 1990, Mercury content of swordfish, *Xiphias gladius*, in relation to length weight, age, and sex. *Marine Pollution Bulletin*, **21**, 293-296.

National Research Cuncil (NRC), 2000, Toxicological Effects of Methylmercury. National Academy Press, Washington, DC.

Official Journal of the European Communities, 1990, L 286/33 of 26 September1990.

Official Journal of the European Communities, 2001, L 77/1 of 16 March 2001.

Rosenberg, J. H., 2002, Perspective: fish-cod to calm the heart. *The New England Journal of Medicine*, **346**, 1102-1103.

Roméo, M., Siau, Y., Sidoumou, Z., and Gnassia-Barelli, M., 1999, Heavy metal distribution in different fish species from the Mauritanian coast. *Science of the Total Environment*, **232**, 169-175.

Snodgrass, J. W., Jagoe, C. H., Bryan, A. L., and Burger, J., 2000, Effects of trophic status, and wetland morphology, hydroperiod and water chemistry on mercury concentrations in fish. *Canadian Journal of Fisheries and Aquatic Sciences*, **57**, 171-180.

Stergiou, K. I., and Karpouzi, V. S., 2002, Feeding habits and trophic levels of Mediterranean fish. *Reviews in Fish Biology and Fisheries*, **11**, 217-254.

Storelli, M. M., Giacominelli-Stuffler, R., and Marcotrigiano, G. O., 1998, Total mercury in muscle of benthic and pelagic fish from the South Adriatic Sea (Italy). *Food Additives and Contaminants*, **15**, 876-883.

Storelli, M. M., and Marcotrigiano, G. O., 2000, Fish for human consumption: Risk of contamination by mercury. *Food Additives and Contaminants*, 17, 1007-1011.

Storelli, M. M., and Marcotrigiano, G. O., 2001, Total mercury levels in muscle tissue of swordfish (*Xiphias gladius*) and bluefin tuna (*Thunnus thynnus*) from the Mediterranean Sea (Italy). *Journal of Food Protection*, **64**, 1058-1061.

Storelli, M. M., Giacominelli-Stuffler, R., and Marcotrigiano, G. O., 2002, Total and methylmercury residues in tuna-fish from the Mediterranean Sea. *Food Additives and Contaminants*, **19**, 715-720.

Storelli, M. M., Storelli, A., Giacominelli-Stuffler, R., and Marcotrigiano, G. O., 2005, Mercury speciation in the muscle of two commercially important fish, hake (*Merluccius merluccius*) and striped mullet (*Mullus barbatus*) from the Mediterranean Sea: estimated weekly intake. *Food Chemistry*, **89**, 295-300.

Watras, C. J., Bsck, R. C., Halvorsen, S., Hudson, R. J. M., Morrison, K. A., and Wente, S. P., 1998, Bioaccumulation of mercury in pelagic freshwater food webs. *Science of the Total Environment*, **219**, 183-208.

Wiener, J. G., and Spry, D. J., 1996, Toxicological significance of mercury in freshwater fish. In "Environmental Contaminants in Wildlife: Interpreting Tissues Concentrations" (Beyer, W. N., Henz, G. H., Redmom-Norwood, A. W. Eds.), pp. 297-339. Lewis, Boca Raton, FL.

World Health Organization, 2003, Summary and conclusions of the sixty-first meeting of the Joint FAO/WHO Expert Committee on Food Additives (JECFA), JECFA/61/SC. Rome, Italy, 10-19 June 2003.

Table 1. Total mercury concentrations (μg g^{-1} wet weight) and estimated weekly intake (EWI) of total mercury (μg kg^{-1} body weight) in dependence on consumption of differed sized fish.

Pools	Striped mullet		Red gurnard		Yellow gurnard		Starry ray		Conger eel		Forkbeard		Frostfish	
	[Hg]	EWI	[Hg]	EWI	[Hg]	EWI	[Hg]	EWI	[Hg]	EWI	[Hg]	EWI	[Hg]	EWI
1	0.16	0.38	0.07	0.17	0.01	0.02	0.15	0.35	0.26	0.61	0.30	0.71	0.16	0.38
2	0.18	0.43	0.12	0.28	0.09	0.21	0.65	1.54	0.29	0.69	0.30	0.71	0.47	1.11
3	0.39	0.92	0.40	0.95	0.08	0.19	0.30	0.71	0.55	1.30	0.42	0.99	0.39	0.92
4	0.58	1.37	0.33	0.78	0.10	0.24	0.81	1.92	0.42	0.99	0.49	1.16	0.32	0.76
5	0.63	1.49	0.42	0.99	0.08	0.19	0.60	1.42	0.43	1.02	0.60	1.42	0.48	1.14
6	1.04	2.46	0.39	0.92	0.25	0.59	0.78	2.55	1.08	2.55	0.48	1.14	0.27	0.64
7	0.88	2.08	0.58	1.37	0.15	0.35	1.08	2.51	1.32	3.12	0.64	1.51	0.68	1.61
8					0.37	0.88	1.20	2.84	1.30	3.07	0.73	1.73	1.15	2.72
9					0.35	0.83	1.19	2.81	1.08	2.55	1.17	2.77	0.80	1.89
10					0.41	0.97			1.29	3.05	0.87	2.06	0.99	2.34
11					0.53	1.25					1.41	3.34	0.82	1.94
Min	0.16	0.38	0.07	0.17	0.01	0.02	0.15	0.35	0.26	0.61	0.30	0.71	0.16	0.38
Max	1.04	2.46	0.58	1.37	0.53	1.25	1.20	2.84	1.32	3.12	1.41	3.34	1.15	2.72
Mean	0.55	1.30	0.33	0.78	0.22	0.52	0.75	1.85	0.80	1.90	0.67	1.59	0.59	1.40
Median	0.58	1.37	0.39	0.92	0.15	0.35	0.78	1.92	0.82	1.93	0.60	1.42	0.48	1.14
St. Dev.	0.33	0.79	0.18	0.42	0.17	0.40	0.37	0.91	0.45	1.06	0.35	0.84	0.32	0.75

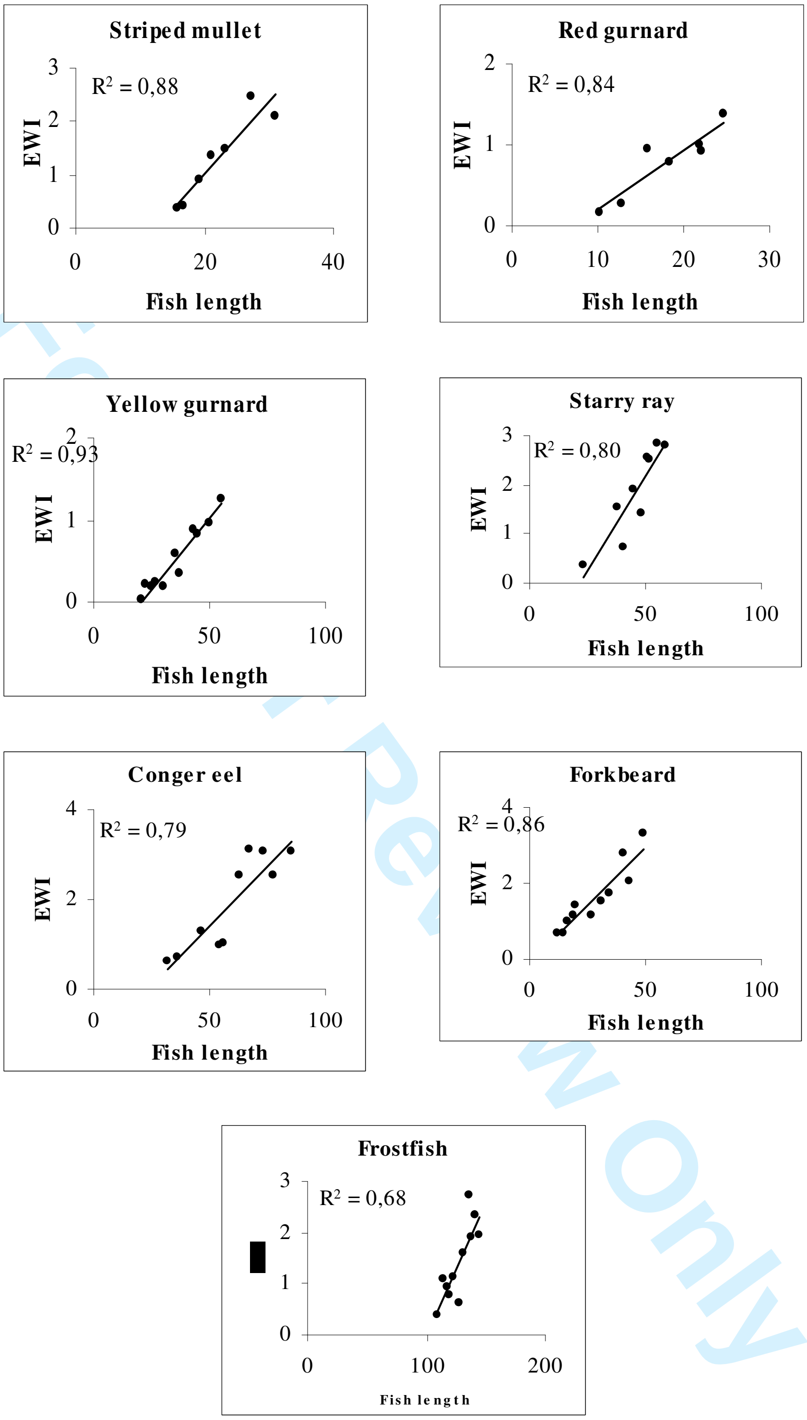


Figure 1. Estimated weekly mercury intake (μg kg^{-1} body wt) versus fish length (cm).