ELSEVIER

Food Chemistry 82 (2003) 491–496

Food Chemistry

www.elsevier.com/locate/foodchem

Analytical, Nutritional and Clinical Methods

Polychlorinated biphenyls in seafood: contamination levels and human dietary exposure

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Received 7 November 2002; received in revised form 22 February 2003; accepted 22 February 2003

Abstract

Different species of fish and crustaceans from the Adriatic Sea, were analysed for polychlorinated biphenyls (PCBs) to estimate the update of daily intake of these contaminants from food. The estimates of the daily TEQs intake b.w. resulted below the range of 1–4 pg TEQs/kg b.w./day set by the World Health Organisation for almost all the species examined except for conger and mackerel. This result supports the assumption of a substantial reduction in intake of these compounds from seafood.

Keywords: Polychlorinated biphenyls; Fish; TEFs; Daily intake

1. Introduction

Polychlorinated biphenyls (PCBs) are a group of toxic and highly persistent organic compounds that consist of 209 congeners differing in the number and position of chlorine atoms on the two coupled biphenyl rings. Although these compounds have low water solubility, particularly in seawater, they are lipophilic and thereby enable fish to absorb, retain and concentrate them particularly in fatty tissues (Cooper, 1991). Surveys carried out in a number of countries on the daily exposure in humans to complex mixture of organochlorine compounds such as, polychlorinated dibenzo-*p*-dioxins, dibenzofurans and coplanar polychlorinated biphenyls, have shown that over 90% occurs through the diet, with foods of animal origin usually being the predominant source (Anderson et al., 1998; Mes, Newsome & Conacher, 1991; Svensson, Nilsson, Hanson, Rappe & Akesson, 1991; Tsutsumi et al., 2001). Fish, particularly those with higher fat levels, seem to be the major conduit of dioxin-like PCB congeners into the human body (Alcock, Behnisch, Jones & Hagenmaier, 1998; Harrison et al., 1998). These coplanar or dioxin-like PCBs, have demonstrated in experimental systems to exert a number of responses similar to those observed for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), the most dangerous organochlorine compound for living organisms. Reports from literature suggest that these dioxin-like compounds have a complex spectrum of toxicological properties, including chloracne, thymic atrophy, liver damage, birth defects, immunotoxicity and cancers (IPCS, 1989, 1993; IARC, 1997).

To facilitate the assessment of the health risks posed by these compounds to man, the concept of Toxic Equivalency Factor (TEF) was developed by the World Health Organization (WHO) Jointly with the European Centre for Environment and Health (ECEH). In this way, the WHO/EURO (1991) recommended a Tolerable Daily Intake (TDI) of 10 pg TEQ/kg b.w./day for these dioxin-related compounds. Subsequently, the WHO European Centre for Environment and Health (ECEH) and the International Programme on Chemical Safety (IPCS), reassessed the health risk of these compounds in the light of the new data (WHO, 1998), establishing a TDI range of 1–4 pg TEQs/kg body weight.

On this basis, in order to assess the risk to man's health, polychlorinated biphenyls were determined in

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0308-8146/03/$ - see front matter
doi:10.1016/S0308-8146(03)00119-5

different species of fish and crustaceans, and the exposure to TCDD equivalents (TEQs) was estimated with particular attention to non-, and mono- *ortho* coplanar congeners which are among the most toxic compounds for humans.

2. Materials and methods

All marine organisms were collected in the southern areas of the Adriatic Sea from May to June 2001. The species chosen are very important commercially and have a significant interest for food use. These species include benthic and pelagic fish and different crustaceans species (Table 1). Within of each species, 30 specimens of similar size were pooled, muscle tissue was taken, and stored below −20 °C pending analysis. To determine polychlorinated biphenyls (PCBs = sum of 17 congeners) concentrations the following method was used. Aliquots (2–10 g) of the homogenised samples were ground with anhydrous sodium sulphate in a mortar. The mixture was extracted with petroleum ether according to Erney's procedure (Erney, 1983). The extracts were then concentrated and subsamples were taken in order to determine the tissue fat content by gravimetry. An aliquot (about 200 mg) of the remaining extract was dissolved in hexane (5 ml) and mixed with H_2SO_4 conc. for the clean up, following the procedure described by Murphy (1972). After centrifugation, the hexane solution was concentrated (about 1 ml) and transferred on a glass column (i.d. 5 mm) filled with 1 g of florisil (100–200 mesh), activated at 120 °C for 16 h, for the separation of PCBs from other organochlorine compounds. The PCBs were eluted from the florisil column using hexane. An aliquot of the eluate (5 ml) was run on a column (i.d. 5 mm) packed with 125 mg of activated carbon (C. Erba, Milano, Italy) for the separation of non-*ortho* PCB congeners, 3,3′,4,4′-T_4CB, (IUPAC 77), 3,3′,4,4′,5-P_5CB (IUPAC 126), and 3,3′,4,4′,5,5′-H_6CB (IUPAC 169) from other PCBs following the method reported by Tanabe, Kannan, Wakimoto and Tatsukawa (1987). The remaining hexane eluate, after concentration, was reserved for the quantification of the other PCBs. Analyses were made on a Carlo Erba Instrument gas chromatograph 8000 Top with automatic injection system (AS-800) and with an electron capture detector ECD-800, Ni^{63}(temperature: 350 °C). The GC was connected to a PC-Pentium III IBM equipped with Chrom-Card version 1.2 software program for integration purposes (C. Erba). For all the analyses a fused-silica capillary column SPB-608 Supelco (length = 30 m, inside diameter 0.25 mm and film thickness 0.25 μm), was used. Helium at a flow rate of 1 ml/min was used as gas carrier, nitrogen as make-up gas 60 ml/min. Temperature was programmed according to the following sequence: injection at 50 °C. Oven steady for the first min and then an increased from 50 to 180 °C at a rate of 15 °C/min. Oven maintained at

Table 1
Concentration range and median of PCBs and ΣPCB (ng/g lipid weight basis) in muscle tissue of fish and crustacean species

	% lipids	PCB 60	PCB 77	PCB 101	PCB 105	PCB 118	PCB 126	PCB 138	PCB 153	PCB 156	PCB 169	PCB 180	PCB 209	Min–max median	ΣPCB
Benthic fish															
Conger conger Conger	0.52	60	3	54	153	308	75	825	1213	243	ND	547	ND	ND-1213 198	3481
Helicolenus dactylopterus Rosefish	0.31	33	5	114	31	237	ND	642	902	183	ND	425	ND	ND-902 183	2572
Lophius boudegassa Angler fish	0.05	ND	8	ND	ND	T	T	246	97	ND	48	T	T	ND-246 73	399
Mullus barbatus Striped mullet	1.82	53	9	69	98	129	ND	174	223	ND	ND	77	ND	ND-223 88	832
Spicara flexuosa Picarel	0.96	ND	8	77	57	128	ND	154	165	58	ND	60	ND	ND-165 69	707
Pagellus blennoides Forked hake	0.15	86	39	916	ND	100	ND	37	272	ND	ND	60	ND	ND-916 86	1510
Pagellus erythrinus Sea bream	0.06	ND	ND	ND	198	192	ND	284	272	ND	ND	ND	ND	ND-284 235	946
Raya clavata Thornback ray	0.11	ND	38	469	28	80	ND	366	292	ND	15	89	T	ND-366 85	1377
Raya oxyrinchus Longnose skate	0.15	ND	46	922	20	40	9	25	38	6	ND	ND	T	ND-922 32	1106
Raya miraletus Winter skate	0.09	ND	25	ND	51	115	ND	90	117	ND	ND	101	T	ND-117 96	499
Pelagic fish															
Sardina pilchardus Pilchard	3.86	ND	8	ND	ND	ND	ND	45	57	ND	ND	28	ND	ND-57 37	138
Merluccius merluccius Hake	0.84	ND	13	54	ND	41	ND	120	105	ND	ND	46	ND	ND-120 50	379
Sardinella aurita Gilt sardine	2.90	ND	2	ND	11	17	ND	68	37	ND	3	58	ND	ND-68 17	196
Scomber scombrus Mackerel	0.42	69	10	143	52	57	15	150	102	29	3	88	ND	ND-150 57	718
Trachurus trachurus Horse mackerel	1.54	ND	5	55	17	39	ND	102	150	11	ND	60	ND	ND-150 47	439
Crustacean Species															
Aristeus antennatus Red shrimp	0.70	ND	1	ND	12	13	ND	31	37	ND	ND	8	ND	ND-37 13	102
Nephrops norvegicus Norway lobster	0.12	ND	ND	360	77	131	ND	232	302	29	ND	285	ND	ND-360 232	1416
Parapenaeus longirostris Pink shrimp	0.15	ND	12	287	ND	38	ND	65	10	ND	7	ND	ND	ND-287 25	419
Plesioniea martia Shirmp	0.07	ND	6	ND	ND	130	ND	131	115	ND	ND	111	ND	ND-131 115	493

ND: not detecte. T: traces.

steady temperature for 1 min and then increased from 180 to 220 °C at a rate of 4 °C/min; oven maintained at steady temperature for 20 min and then increased from 220 to 275 °C at a rate of 5 °C/min; from this point until the end of the analytical run, the column remained isothermal at a temperature of 275 °C.

The individual PCB congeners were 8, 20, 28, 35, 52, 60, 77, 101, 105, 118, 126, 138, 153, 156, 169, 180 and 209 numbering system (Ballschmiter & Zell, 1980) determined against the corresponding individual standards obtained from ULTRA Scientific, Inc. (chemical purity 99%). Recoveries are determined by adding known amounts of PCBs standards to empty samples and found to be within 80–110%. The limits of detection (LOD) was 5 pg/g on a wet weight basis for the PCB congeners. Quantification was done within the linear range of the detector. Residues in 100% of the samples were confirmed by gas–liquid chromatography–mass spectrometry (Fisons MD 800).

3. Results and discussion

Table 1 shows the range and median values of total polychlorinated biphenyls (PCBs) and individual congener concentrations, expressed on ng/g lipid weight basis in muscle tissue of different fish and crustacean species. Median PCBs fell within 32 and 235 ng/g and 17 and 57 ng/g in the benthic and pelagic fish, respectively, whereas in the crustacean samples ranged from 13 to 232 ng/g. These data revealed that a difference in contamination levels seems to characterize the various species. The interaction organism–sediment may explain most of the variation in pollutant levels among species: organisms living in close relationship with sediment, are exposed to sedimentary PCBs via several routes, such as direct contact with sediment, respiration of interstitial water and incidental ingestion of sediment, consequently they show a contaminant load comparatively higher than in pelagic organisms (Porte & Albaiges, 1993; Pastor, Boix, Fernandez, & Albaiges, 1996). The data in the fish in question fit this general picture well: benthic species (sea bream: 235 ng/g; conger: 198 ng/g; rosefish: 183 ng/g; winter skate: 96 ng/g; striped mullet: 88 ng/g; forked hake: 86 ng/g; thornback ray: 85 ng/g; angler fish: 73 ng/g; picarel: 69 ng/g; longnose skate: 32 ng/g) in contrast to pelagic organisms (mackerel: 57 ng/g; hake: 50 ng/g; horse mackerel: 47 ng/g; pilchard: 37 ng/g; gilt sardine: 17 ng/g) showed higher concentrations. Among crustaceans the highest values were encountered in Norway lobster (232 ng/g), followed by shrimp (115 ng/g) and pink shrimp (25 ng/g), while red shrimp exhibited the lowest levels (13 ng/g). Polychlorinated biphenyl congener profile in fish showed a high proportion of the more chlorinated PCBs, with a clear predominance of hexachlorinated isomers, with percentages above 40% (excepted forked hake), followed by penta- and hepta-, while tetrachlorobiphenyls showed the lowest percentages, generally below 10%. The bioaccumulation pattern in crustacean flesh was quite similar to that of fish, although in some species an enrichment in pentachloroisomers (see pink shrimp and norway lobster) was observed. Dichlorobiphenyl PCB 8, trichlorobiphenyls (PCB 20, 28, 35) and tetrachlobiphenyl PCB 52 were below the detection limit for all samples, as well as decachlorobiphenyl, PCB 209, present at trace level or absent. The individual PCB congener distribution in the various species, showed that the compounds with the highest concentration in most fish and crustacean samples were hexachlorobiphenyls PCB 138 and PCB 153, followed by PCB 180. Other chlorobiphenyls found in major amounts, especially in crustacean flesh, included congeners PCB 101 and PCB 118. In agreement with our data, the above mentioned compounds are the prevailing congeners usually present in marine organisms (Bayarri, Baldassarri, Iacovella, Ferrara, & Di Domenico, 2001; Porte & Albaiges, 1993; Storelli & Marcotrigiano, 2000a, 2000b, 2001b). On the other hand, the International Committee for the Exploration of the Sea recommended to quantify some PCB congeners (PCBs 28, 52, 101 118, 138, 153 and 180), because they proved to be relatively abundant in fish. The major presence of these congeners is a direct consequence of their molecular structure.

To assess the potential health impact, the tolerable daily intake (TDI) range of 1–4 pg TEQs/kg body weight for these dioxin-related compounds, recommended by the World Health Organization (European Commission, 1999, 2000b; WHO, 1998) for a 60 kg person, was used as a guideline. To compare the values found in the analyzed species expressed in lipid weight basis, with the tolerable daily intake (TDI) range adopted by WHO, the concentrations were also expressed as wet weight values (pg/g; Table 2) and the TEQs were given using the WHO-toxicity equivalent factors (TEFs) (Van den Berg et al., 1998).

Table 3 shows, the person daily dietary intakes (μg/person/day) of the six congeners dioxin-like PCBs (mono-*ortho* PCB 105, 118 and 156; non-*ortho* PCB 77, 126, 169), among the twelve considered by WHO (1999) from each species assuming a gross average seafood consumption of 60 g person day in Italy (Istat, 2000). More, in Table 3 are reported the TEQ exposure from mono- and non-ortho congeners (pg/person/day) calculated by multiplying the daily intakes by the corresponding TEF for each PCB congener, and the daily TEQ intake per kilogram body weight (60 kg). These estimates assumed that non-detected isomer concentrations equal to zero (ND = 0), as well as non-detected isomer concentrations equal to half of the LOD (ND = 1/2 LOD) and LOD (ND = LOD). The values

Table 2
Concentration range and median of PCBs and ΣPCB (pg/g wet weight basis) in muscle tissue of fish and crustacean species

	% lipids	PCB 60	PCB 77	PCB 101	PCB 105	PCB 118	PCB 126	PCB 138	PCB 153	PCB 156	PCB 169	PCB 180	PCB 209	Min–max median	ΣPCB
Benthic fish															
C. conger Conger	0.52	312	16	281	796	5923	49	4290	6508	1264	ND	2985	ND	ND-6508 1030	22424
H. dactylopterus Rosefish	0.31	102	16	353	96	735	ND	1990	2896	567	ND	1363	ND	ND-2896 567	8118
L. boudegassa Angler fish	0.05	ND	4	ND	ND	T	T	133	49	ND	14	T	T	ND-133 37	200
M. barbatus Striped mullet	1.82	965	16	1356	1784	2348	ND	3215	4059	ND	ND	1401	ND	ND-4059 1593	15144
S. flexuosa Picarel	0.96	ND	77	799	547	1229	ND	1478	1584	557	ND	576	ND	ND-1584 658	6787
P. blennoides Forked hake	0.15	129	59	1374	ND	150	ND	109	408	ND	ND	90	ND	ND-1374 129	2266
P. erythrinus Sea bream	0.06	ND	ND	ND	119	115	ND	170	163	ND	ND	ND	ND	ND-170 141	567
R. clavata Thornback ray	0.11	ND	42	554	31	88	ND	403	321	ND	16	98	T	ND-554 93	1515
R. oxyrinchus Longnose skate	0.15	ND	9	1383	30	60	14	58	57	9	ND	ND	T	ND-1383 48	1660
R. miraletus Winter skate	0.09	ND	23	ND	46	104	ND	81	105	ND	ND	91	T	ND-105 86	450
Pelagic fish															
S. pilchardus Pilchard	3.86	ND	20	ND	ND	ND	ND	1826	2200	ND	ND	1281	ND	ND-2200 1409	5327
M. merluccius Hake	0.84	ND	10	454	ND	344	ND	1008	982	ND	ND	386	ND	ND-1008 420	3183
S. aurita Gilt sardine	2.90	ND	8	ND	319	493	ND	1970	1203	ND	9	1682	ND	ND-1970 493	5684
S. scombrus Mackerel	0.42	290	20	601	218	239	16	699	428	122	13	370	ND	ND-699 239	3015
T. trachurus Horse mackerel	1.54	ND	17	847	262	601	ND	1571	2310	169	ND	984	ND	ND-2310 724	6761
Crustacean species															
A. antennatus Red shrimp	0.70	ND	7	ND	84	91	ND	217	259	ND	ND	56	ND	ND-259 88	714
N. norvegicus Norway lobster	0.12	ND	ND	432	92	157	ND	278	362	35	ND	342	ND	ND-432 278	1698
P. longirostris Pink shrimp	0.15	ND	18	431	ND	57	ND	97	15	ND	10	ND	ND	ND-431 38	628
P. martia Shrimp	0.07	ND	4	ND	ND	91	ND	92	81	ND	ND	78	ND	ND-92 81	346

ND: not detected. T: traces.

Table 3
Estimations of daily intake and TEQs exposure from mono-and non-*ortho* PCB n the different species of fish and crustaceans

Species	Daily intake μg/person/day Mono + Non-*ortho*	pgTEQs/person/day Mono-*ortho*			pgTEQs/person/day Non-*ortho*			pgTEQs/person/day Mono + Non-*ortho*			pgTEQs/kg b.w./day		
		0	1/2 LOD	LOD	0	1/2 LOD	LOD	0	1/2 LOD	LOD	0	1/2 LOD	LOD
Benthic fish													
Conger	503.34	78.24	78.24	78.24	294.10	295.60	297.10	372.10	373.84	375.34	6.20	6.23	6.26
Rosefish	93.54	22	22	22	0.10	17.47	33.97	22.97	39.47	55.10	0.38	0.66	0.92
Angler fish	1.68	–	0.11	0.21	14.42	29.42	44.42	14.42	29.53	44.63	0.24	0.49	0.74
Striped mullet	257.76	24.79	24.87	24.94	0.10	17.48	33.98	25.77	42.35	58.92	0.43	0.71	0.98
Picarel	144.60	27.36	27.36	27.36	0.05	16.96	33.46	27.82	44.32	60.82	0.46	0.74	1.01
Forked hake	12.54	0.90	0.99	1.08	0.04	16.85	33.39	1.25	17.84	34.43	0.02	0.30	0.57
Sea bream	14.04	1.40	1.48	1.55	–	16.52	33.03	1.40	18.00	34.58	0.02	0.30	0.58
Thornback ray	10.62	0.72	0.80	0.87	9.85	24.85	39.85	10.57	25.65	40.72	0.18	0.43	0.68
Longnose skate	10.92	0.81	0.81	0.81	84.41	85.91	87.41	85.22	86.72	88.22	1.42	1.45	1.47
Winter skate	10.38	0.90	0.98	1.05	0.14	16.64	33.14	1.04	17.62	34.19	0.02	0.29	0.57
Pelagic fish													
Pilchard	18.54	–	0.11	0.21	0.12	18.35	34.85	0.12	18.46	35.06	0.01	0.31	0.58
Hake	27.18	2.06	2.15	2.24	0.06	17.15	33.65	2.71	19.30	35.89	0.05	0.32	0.60
Gilt sardine	57.54	4.87	4.95	5.02	5.45	20.45	35.45	10.32	25.40	40.47	0.17	0.42	1.67
Mackerel	41.82	6.40	6.40	6.40	103.92	103.92	103.92	110.32	110.32	110.32	1.84	1.84	1.84
Horse mackerel	66.54	10.25	10.25	10.25	0.10	16.96	33.46	10.71	27.21	43.71	0.18	0.45	0.73
Crustacean species													
Red shrimp	10.92	1.05	1.13	1.20	0.04	16.54	33.04	1.09	17.67	34.24	0.02	0.29	0.57
Norway lobster	17.04	2.54	2.54	2.54	–	16.52	33.03	2.54	19.06	35.57	0.04	0.32	0.59
Pink shrimp	5.10	0.34	0.43	0.52	6.11	21.11	36.11	6.45	21.54	36.63	0.11	0.36	0.61
Shrimp	5.70	0.55	0.64	0.73	0.02	16.52	33.02	0.57	17.16	33.75	0.01	0.29	0.56

calculated at ND = 0, at ND = 1/2 LOD and ND = LOD were comprised between 0.01–6.20 TEQ/kg b.w., 0.29–6.23 TEQ/kg bw, and 0.56–6.26 TEQ/kg bw, respectively. These values resulted below to the range of 1–4 pg TEQs/kg b.w./day set by the World Health Organization (WHO, 1998) for almost all the species except for conger. In agreement with literature, this result supports the assumption of a substantial reduction in the intake of these compounds from food. Recent studies from countries that began to implement measures to reduce dioxin emissions in the late 1980s show, in fact, decreasing PCDD/PCDF and PCB levels in food and consequently a lower dietary intake of these compounds (Duarte-Davidson & Jones, 1994; Van Leeuwen, Feeley, Schrenk, Larsen, Farland, & Younes, 2000). However, since it has been stated that fish is the main dietary source of PCBs (Alcock, Behnisch, Jones, & Hagenmaier, 1998; European Commission, 2000a; Harrison et al., 1998), in order to carry on an assessment of the potential human risk deriving from the consumption of seafood, it is necessary to consider communities and individuals according to the amounts they consume. Population that ingests high amounts of fish could be, in fact, of importance in terms of risk assessment. Cole, Kearney, and Gilman (1995) found that blood in fish eaters aged >50 years old contained twice the PCB-TEQ compared to a non-fish eating group. Dewailly et al. (1994) found elevated levels of dioxin-like PCBs in the breast milk of Inuit women in the Arctic, whose principal protein sources are fish and sea mammals. The population of Italy is exposed to a different diet, because of its geographical position, mainly outstretched towards the sea, except for the northern zone boardered by the Alps. In Northern Italy there is a low consumption of seafood with respect to meat products, while in Central and Southern Italy and the Islands, seafood products occupy a relevant part of the diet (Storelli & Marcotrigiano, 2001a; Storelli, Storelli, & Marcotrigiano, 2002). Therefore, organochlorine compound intake and, consequently, health risk may differ considerably depending on a person's location and the amount of food consumed. On this basis other quantities of consumption of seafood (200, and 250 g day^{-1}), usually eaten by some segments of the population (see fishermen and their families), were considered to calculate the TEQs exposure from PCB dioxin-like, and establish whether it approaches or exceeds the value range fixed by the WHO (1998). These esteems indicated values higher than the limit established by the WHO (1998), not solely for conger and mackerel but also for the benthic species longnose skate. For this latter a value (5.92 pgTEQs/kg b.w.) slightly above 4 pg TEQs/kg b.w./day was observed consequently to the consumption of 250 g day^{-1}.

Although the human intake of these compounds is almost totally through the diet with the largest potential contributor being fish, there were no regulatory requirements for PCBs residues in seafood in Italy. Only recently, the Legislative Decree n. 336/1999 (G.U., 1999) established in different foods of animal origin a maximum residue limit for PCBs of 100 ng/g on a lipid basis. In this light, PCB concentrations, in all pelagic fish analyzed were well below this value, whereas in benthic species concentrations exceeded or approached the limiting value in almost all species, except in picarel and winter skate. Among crustacean species, only in Norway lobster the levels were above the tolerance limit. From a public health aspect, though in many species the concentrations of PCBs exceeded the value of 100 ng/g on a lipid basis, there is no indication of important risks associated with the consumption of these fish products because the estimate of TEQs exposure was considerably below the limit established by the World Health Organization (WHO, 1998) except for two species. However, it must be borne in mind that a population is also subjected to polychlorinated biphenyls contamination via other sources. A recent report regarding the daily intake of PCB from food in Italy include among main contamination dietary sources, besides meat and fish, also vegetables containing PCB 126 (Zuccato et al., 1999) the most toxic congener within the polychlorinated biphenyls family. In addition, it should be pointed out that since the WHO (1998) TDI of 1–4 pg TEQs/kg bw/day includes not only the dioxin-like PCBs but also the PCDDs and PCDFs, it is highly likely that the daily intake we calculated was underestimated. In this perspective and considering what underlined by WHO (1998) "that the upper range of the TDI of 4 pg TEQ/kg b.w. should be considered a maximal tolerable intake on a provisional basis and that the ultimate goal is to reduce human intake levels below 1 pg TEQ/kg b.w./day" it is necessary a continuos monitoring of the levels of these compounds in foods, with particular attention to seafood considered primary vectors of these substances for man.

Acknowledgements

The authors thank Dr. Grazia Barone and Dr. Maria Blasi for their technical support.

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