Rocz Panstw Zakl Hig 2015;66(3):203-210

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ORIGINAL ARTICLE

TOTAL AND INORGANIC ARSENIC IN FISH, SEAFOOD AND SEAWEEDS - EXPOSURE ASSESSMENT

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ABSTRACT

Background. According to the European Food Safety Authority (EFSA), fish, seafood and seaweeds are foodstuffs that significantly contribute to dietary arsenic intake. With the exception of some algal species, the dominant compounds of arsenic in such food products are the less toxic organic forms. Both the Joint FAO/ WHO Expert Committee on Food Additives (JECFA) and EFSA recommend that speciation studies be performed to determine the different chemical forms in which arsenic is present in food due to the differences in their toxicity. Knowing such compositions can thus enable a complete exposure assessment to be made.

Objectives. Determination of total and inorganic arsenic contents in fish, their products, seafood and seaweeds present on the Polish market. This was then followed by an exposure assessment of consumers to inorganic arsenic in these foodstuffs.

Materials and Methods. Total and inorganic arsenic was determined in 55 samples of fish, their products, seafood as well as seaweeds available on the market. The analytical method was hydride generation atomic absorption spectrometry (HGAAS), after dry ashing of samples and reduction of arsenic to arsenic hydride using sodium borohydride. In order to isolate only the inorganic forms of arsenic prior to mineralisation, samples were subjected to concentrated HCl hydrolysis, followed by reduction with hydrobromic acid and hydrazine sulphate after which triple chloroform extractions and triple 1M HCl re-extractions were performed. Exposure of adults was estimated in relation to the Benchmark Dose Lower Confidence Limit ($BMDL_{05}$) as set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) that resulted in a 0.5% increase in lung cancer (3.0 µg/kg body weight (b.w.) per day).

Results. Mean total arsenic content from all investigated fish samples was 0.46 mg/kg (90th percentile 0.94 mg/kg), whilst the inorganic arsenic content never exceeded the detection limit of the analytical method used (0.025 mg/kg). In fish products, mean total arsenic concentration was 1.48 mg/kg (90th percentile: 2.42 mg/kg), whilst in seafood they were 0.87 mg/kg (90th percentile: 2.23 mg/kg), for inorganic arsenic contamination at the 90th percentile was 0.043 mg/kg with most results however being less than 0.025 mg/kg. The highest inorganic arsenic levels were determined in the Hijiki algal species samples (102.7 mg/kg), whereas the other algal samples gave a mean inorganic concentration of 0.41 mg/kg (90th percentile 0.86 mg/kg). The estimated average adults exposure to inorganic arsenic in fish, seafood and seaweeds was less than 0.5% of the lowest $BMDL_{0.5}$ dose. Only for the Hijiki seaweed it was at 4.9% $BMDL_{0.5}$.

Conclusions. Results demonstrate that dietary arsenic intake from fish, seafood and seaweed along with all their products do not constitute a significant health threat to consumers apart from the seaweed species *Hizikia fusiformis* in which over 40% of all the inorganic arsenic compounds were found.

Key words*: total arsenic, inorganic arsenic, fish, seafood, seaweeds, exposure assessment*

STRESZCZENIE

Wprowadzenie. Ryby, owoce morza i wodorosty morskie należą do grupy środków spożywczych, które według Europejskiego Urzędu ds. Bezpieczeństwa Żywności (EFSA) mogą wnosić istotny udział do pobrania arsenu z dietą. W tej grupie produktów za wyjątkiem niektórych gatunków glonów, dominującymi formami arsenu są mniej toksyczne organiczne połączenia. Zarówno Połączony Komitet Ekspertów FAO/WHO ds. Substancji Dodatkowych do Żywności (JECFA) jak i EFSA wskazują na celowość prowadzenia badań specjacyjnych pozwalających określić różne formy chemiczne, w jakich arsen występuje w żywności, mając na uwadze duże różnice w ich toksyczności. Poznanie informacji o zawartości poszczególnych związków pozwala na dokonanie pełnej oceny narażenia.

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Cel badań. Oznaczenie zawartości arsenu całkowitego i nieorganicznego w rybach i ich produktach, owocach morza i wodorostach morskich dostępnych w obrocie handlowym w Polsce oraz ocena narażenia konsumentów na pobranie arsenu nieorganicznego z tymi grupami środków spożywczych.
Materiał i metody. Analizie na zawartość arsenu całkowitego i nieorganicznego poddano 55 próbek ryb, ich produktów, owoców morza oraz glonów pochodzących z obrotu handlowego. Zawartości arsenu całkowitego i nieorganicznego oznaczono po suchej mineralizacji próbek metodą absorpcyjnej spektrometrii atomowej z wykorzystaniem generacji wodorków (HGAAS), po uprzedniej redukcji arsenu do arsenowodoru za pomocą borowodorku sodu. W celu wydzielenia nieorganicznych form arsenu próbki przed mineralizacją poddano hydrolizie w środowisku stężonego HCl, a następnie redukcji w obecności kwasu bromowodorowego i siarczanu hydrazyny oraz 3-krotnej ekstrakcji chloroformem i reekstrakcji 1M HCl. Oszacowane narażenie w odniesieniu do osób dorosłych porównano z wartością najniższej dawki wyznaczającej (Benchmark Dose Lower Confidence Limit) $BMDL_{0,5}$ ustalonej przez The Joint FAO/WHO Expert Committee on Food Additives (JECFA) powodującej 0,5%-owy wzrost zachorowań na raka płuc (3,0 μg/kg m.c./dzień).
Wyniki. Średnia zawartość arsenu całkowitego w badanych próbkach ryb wyniosła 0,46 mg/kg (90-ty percentyl: 0,94 mg/kg), natomiast arsenu nieorganicznego nie przekraczała granicy wykrywalności stosowanej metody 0,025 mg/kg. W przypadku przetworów rybnych średnie zanieczyszczenie arsenem wyniosło 1,48 mg/kg (90-ty percentyl: 2,42 mg/kg) natomiast owoców morza odpowiednio: 0,87 mg/kg (90-ty percentyl: 2,23 mg/kg), dla arsenu nieorganicznego 90-ty percentyl 0,043 mg/kg przy większości wyników poniżej 0,025 mg/kg.
Najwyższym zanieczyszczeniem arsenem nieorganicznym charakteryzowały się glony z gatunku Hijiki (najwyższa zawartość 102,7 mg/kg), w pozostałych próbkach glonów średnia zawartość arsenu nieorganicznego wynosiła 0,41 mg/kg (90-ty percentyl: 0,86 mg/kg). Oszacowane średnie narażenie na arsen nieorganiczny w odniesieniu do osób dorosłych wyniosło dla ryb, owoców morza i wodorostów poniżej 0,5% wartości najniższej dawki wyznaczającej $BMDL_{0,5.}$ Jedynie w przypadku gatunku Hijiki było ono rzędu 4,9 % $BMDL_{0,5.}$
Wnioski. Na podstawie uzyskanych wyników stwierdzono, że pobranie arsenu w wyniku spożycia ryb, ich produktów, owoców morza, oraz wodorostów morskich nie stanowi istotnego zagrożenia dla zdrowia konsumentów poza gatunkiem *Hizikia fusiformis* zawierającym ponad 40% połączeń nieorganicznych arsenu.

Słowa kluczowe: *arsen całkowity, arsen nieorganiczny, ryby, owoce morza, wodorosty, ocena narażenia*

INTRODUCTION

The health benefits and risks issue arising from consuming foodstuffs of marine origin has been intensely debated in scientific circles [11]. Dieticians have laid stress on the pro-healthy aspects of eating such products, particularly due to the presence of *omega*-3 fatty acids, vitamins, minerals and high protein content [9, 21, 24, 31]. Toxicologists however warn against their excessive consumption because of the high levels of various contaminants harmful to human health like methylmercury, cadmium, biotoxins, dioxins, polychlorinated biphenyls (PCBs) and arsenic [18, 22, 31].

Arsenic and especially its inorganic forms are significant food contaminants. This element is cumulated in large quantities in food of marine origin [37, 38, 48]. The International Agency for Research on Cancer (IARC) has classified arsenic to Group I as substance carcinogenic to humans. Inorganic arsenic compounds are also genotoxic and neurotoxic mainly for the developing central nervous system of foetus, infants and young children. In addition, arsenic causes cardiovascular diseases, peripheral vascular disorders, anaemia, immune and reproductive system disorders together with an impairment of metabolic cellular processes in the liver and kidneys through enzymatic inhibition [38, 46, 47]. The following environmental species of arsenic can be placed in a descending order of toxicity; monomethylarsonous acid [MMA(III)] > dimethylarsinous acid [DMA(III)] > arsenic (III) and the compounds of [As (III)]> acid, arsenic (V) and its the compounds [As (V)] > monomethylarsonic acid [MMA(V)] > dimethylarsinic acid [(DMA(V)] arsenocholine (AC) / arsenobetaine (AB) [8, 50].

In 2009, EFSA requested a revision of the Provisional Tolerable Weekly Intake (PTWI) of inorganic arsenic from all sources (i.e. 0.015 mg/kg body weight) and established a range of benchmark dose lower confidence limit ($BMDL_{01}$) values between 0.3 and 8 μg/kg b.w. per day [38].

A similar verification in its risk assessment was performed by JECFA that proposed the $BMDL_{05}$ value at 3.0 μg/kg b.w. per day resulting in a 0.5% increase in lung cancer rates as determined from epidemiological studies [47].

Food is main source of arsenic exposure in humans. In particular, food of marine origin has high arsenic contamination compared to other foodstuffs [3, 27, 36]. According to the European Commission Scientific Cooperation Project (SCOOP), intake of arsenic from marine foodstuffs may be in excess of 50% [41].

Arsenic in these foodstuffs is chiefly present in the form of organic compounds which are less acutely toxic than the toxic inorganic ones [14, 20, 26, 29, 34, 38, 41, 43]. Fish and marine crustaceans are mainly contaminated with arsenobetaine (AB,) whilst arseno-

-sugar compounds and arsenocholine are predominantly found in algae and molluscs. Arseno-lipids are present in oily marine fish.

It is of concern that some algal species, for instance the *Hizikia fusiformis* species, contain high amounts of inorganic arsenic, sometimes up to 80% of the total arsenic [51]. Studies have demonstrated that a pretreatment step of washing and soaking of these seaweeds can decrease the inorganic arsenic content to 60% [9]. Marine organism are able to convert those inorganic forms of arsenic which are more toxic into the less toxic organic forms as part of the detoxification process [10]. Algae are universally considered as bioindicators of water pollution due to their ability of accumulating metals from the environment, including arsenic [1, 19]. Some countries like United Kingdom, Australia, New Zealand, France, USA have put forward specific recommendations for consumers for eating seaweed that includes avoiding the *Hizikia fusiformis* species due to the aforementioned contamination by inorganic arsenic [1, 12, 13, 14, 51].

Even though the mean monthly consumption of fish and seafood is not great in Poland, (approx. 0.34 kg per capita in 2013 according to Central Statistical Office data) [5], Poles are ever more frequently changing their culinary habits and traditions, reaching for example, seafood and seaweeds one of the main ingredients of Sushi.

Since 2010, notifications within the Rapid Alert System for Food and Feed (RASFF) where high levels of arsenic have been reported in dietary supplements, especially those originating from dried seaweed, have significantly increased. The highest levels were reported in the *Hizikia fusiformis* species coming from Japan; 122 mg/kg [45].

EU legislation is set to place limits on arsenic, including inorganic arsenic, contamination beginning with January 2016 for certain foodstuffs through amending Commission Regulation (EC) No 1881/2006 [6]. This limit covers foodstuffs like rice and its products including rice destined for the production of food for infants and young children.

Current requirements as stipulated by the Codex Alimentarius standards of FAO/WHO, (amongst others), only deal with total arsenic [16], as do those regarding drinking mineral, table and spring water in the case of additives and pesticide residues [7, 32, 33, 35].

It is therefore not possible to perform an adequate health risk assessment without knowing the proportions of organic to inorganic arsenic compounds in various foodstuffs by just using total arsenic content; this yielding falsely high scores leading to a flawed risk management.

Besides determination of total and inorganic arsenic in certain fish species and their products along with seafood and algae, the main aim of this study was to assess consumer exposure to more toxic inorganic arsenic.

MATERIALS AND METHODS

Test samples

Consisted of 55 samples of fish, their products, seafood and algae that are available on the Polish market.

Apparatus and instrumentation

An ASA spectrometer Philips / Pye Unicam model 9200 was used with an attachment for 9360 PU hydride generation in a continuous system. Also used was a Vortex Multi Reax, shaker (Heidolph, Germany), 5810R centrifuge (HG Eppendorf, Germany), Ceran 500 hotplate (Harry Gestigkeit GmbH, Germany), Pyro Milestone TC microwave oven (Italy), hydrophobic filters (syringe, Millipore, pore diameter: 0.45 µm), Class A laboratory glassware and FALCON polypropylene (PP) tubes.

Reagents

Nitric acid (65%, Merck), hydrochloric acid (35% - 38%, Avantor), hydrobromic acid (≥48% Sigma-Aldrich), hydrazine sulphate (Fluka), chloroform (99.0% -99.4%, Sigma-Aldrich), magnesium nitrate hexahydrate (Merck), magnesium oxide (Chempur), potassium iodide (Avantor), ascorbic acid (Avantor), sodium hydroxide (Merck), sodium borohydride (Merck), basic standard solution of arsenic (V) at a concentration of 1g/L (Merck) and deionised water. All reagents used were of an appropriately high purity.

Sample preparation for total arsenic determination

Samples were appropriately fragmented as weighed amounts of around 0.5 to 2 g and were placed into a quartz vaporiser and 10 mL of ashing aid was added (20% w / v $MgNO_3$ * $6H_2O$, 2% w/v MgO) plus 5 mL of 50% v/v nitric acid solution. After evaporating to dryness on the hotplate, the residue was digested in the microwave oven using a manufacturer's recommended mineralization programme; the final sample ashing temperature not exceeding 400°C. The final ash was dissolved in 5 mL of 6M HCl, filtered and transferred to a flask and filled to 50 mL with 6M hydrochloric acid. Blanks were prepared under analogous conditions [4].

Sample preparation for inorganic arsenic determination

Samples of approx. 0.5 g to 1g were placed into PP centrifuge tubes and 1 mL deionised water was added followed by thorough shaking to ensure complete wetting. Concentrated HCl (20 mL) was then added and the mixture was left overnight (12 h -15 h). Test samples were, next, reduced by 2 mL of hydrobromic acid and 1 mL of hydrazine sulphate (1.5% m/m) and then shaken by hand for 30 seconds. Next, samples were extracted with chloroform (10 mL) and shaken for 3 minutes (at 1400 rev/min). The mixture was then centrifuged

(20 min, 4000 rev/min) and the organic layer was transferred to a clean PP centrifuge tube. The extraction process was repeated twice more and the organic layers retained and then purified using hydrophobic membrane filters. The purified chloroform extracts were now back extracted twice with 10 mL 1M HCl, where each time the mixture was stirred for 3 min. and centrifuged for 20 min at 4000 rev/min. The resulting acidic solutions were transferred to flasks, evaporated to dryness on the hotplate followed by microwave oven ashing at a temperature ramp programme consisting of a 50ºC stepwise increase from 150ºC, up to, but not exceeding 400°C which was then maintained for 8 hours leading to white ash formation. This ash was then moistened with water, dissolved in 6M hydrochloric acid solution and transferred to 25 mL flasks [4]. Blanks were also prepared under analogous conditions.

Determination of total and inorganic arsenic

Arsenic was analytically measured by an in-house adapted method based on Munoz [28]. Total arsenic was measured by taking suitable aliquots of the mineralised test samples (10-20 mL) which were reduced by 6M HCl along with a mixture of 5% w/v aqueous potassium iodide and 5% w/v aqueous ascorbic acid. The volume was adjusted to a fixed total volume with 6M HCl.

Inorganic arsenic was determined from subjecting the entire mineralized test sample to reduction.

The final analyses of the total and inorganic arsenic contents were performed by atomic absorption spectrometry using an hydride generation technique (HGAAS) after sodium borohydride reduction to arsenic-hydride; according to the manufacturer's instruction manual [4]. A standard curve was prepared from a stock standard solution of arsenic (V) containing 1 g/L of arsenic and ranged from 0 ng/mL to 5 ng/mL. As an internal quality control test, crab meat with a known content of inorganic arsenic was used which our laboratory obtained from its participation in the proficiency testing FAPAS round-0791.

RESULTS

Total and inorganic arsenic were determined in 55 samples of fish, fish products, seafood and together with algae derived ones that are on the market. In order to enable comparison with data from other EU member States participating in the SCOOP (Scientific Cooperation on Questions Relating to Food) programme and according to EFSA guidelines, for results below limit of detection (ie. 0.025 mg/kg inorganic arsenic and 0.031 mg/kg total arsenic), calculations were performed taking half these values [17, 40, 41]. The results are presented Figure 1, whereas the proportions of inorganic arsenic to total arsenic are shown in Figure 2.

DISCUSSION

Mean total arsenic levels in investigated fish samples were 0.46 mg/kg, with a 90th percentile value of 0.94 mg/kg, which were ten-fold less than those found in a USA study on such foodstuffs [25].

Inorganic arsenic content did not exceed the analytical detection limit (i.e. 0.025 mg/kg) of the method used, which confirms the fact that inorganic arsenic in fish is present mainly in the form of less toxic organic arsenic compounds reaching as high as 90% of the total [23, 48].

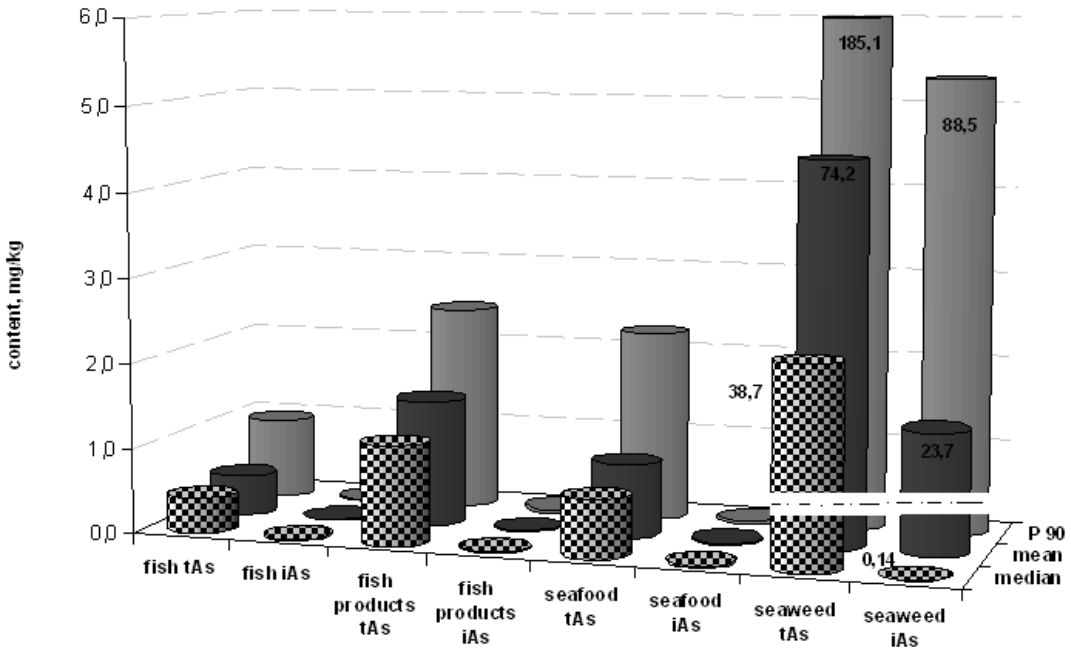

Figure 1. Total (tAs) and inorganic arsenic (iAs) in tested samples of fish, fish products, seafood and seaweed; mg/kg

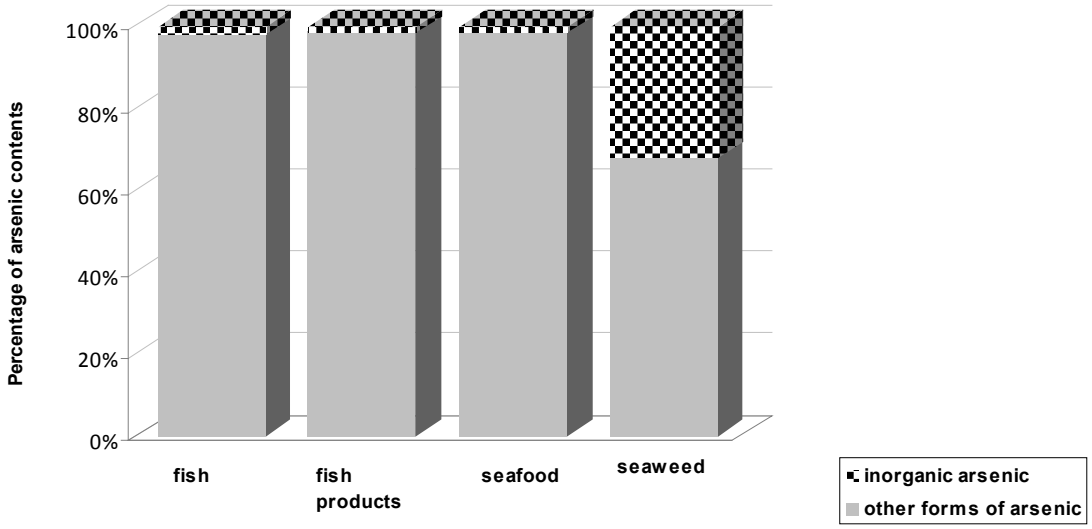

Figure 2. Proportions of different arsenic forms of arsenic measured in tested samples.

Marine fish were more contaminated by total arsenic compared with fresh water fish; 0.516 mg/kg versus 0.297 mg/kg, respectively. A similar trend was observed by other authors; however contamination with total arsenic was much higher among demersal species e.g. flounder of 9.36 mg/kg [37]. The contamination levels in fresh water fish were found to be comparable to an Asian study [36] but lower than a study from Belgium [37].

Mean arsenic contaminant levels in fish products were 1.48 mg/kg (90th percentile of 2.42 mg/kg) which were similar to those found in other EU countries [38]. Smoked fish however had higher levels of both total and inorganic arsenic compared to the other products. But, no such trend was observed in a French study [42]. The highest total arsenic level that was measured by our study was in sample of smoked sprats from the Baltic Sea; 2.52 mg/kg.

Other countries taking part in the SCOOP programme showed higher mean arsenic contamination levels in fish and seafood compared to our study, with the exception of Greece, where levels were significantly lower [41].

The mean total and inorganic arsenic contamination in seafood were found to be: 0.87 mg/kg (90th percentile of 2.23 mg/kg) and mostly below 0.025 mg/kg (90th percentile 0.043 mg/kg), respectively, with levels in oysters and mussels being higher. Despite this, arsenic contamination (both inorganic and total) was lower in such foodstuffs when compared to other countries [2, 10, 15, 25, 38].

Definitively much higher levels of inorganic arsenic were found in dried algae. The highest content of 102.7 mg/kg was found in the Hijiki (*Hizikia fusiforme*) species from Japan, at 188.6 mg/kg for total arsenic.

In the other algae samples, the mean inorganic and total arsenic was 0.41 mg/kg (90th percentile of 0.86 mg/kg) and 34.8 mg/kg (90th percentile of 54.1 mg/kg) respectively. Arsenic contamination found in investigated samples of algae was comparable to results from other countries [13, 30]. The much higher inorganic arsenic content in brown algae of the species Hijiki even up to 117.0 mg/kg was also observed in other countries [1, 51]; Our studies demonstrated a doubling of total arsenic content in fish, their products and seafood as compared to those found from monitoring studies conducted in 2004-2008 [51].

Taking into account the data of the Central Statistical Office, along with the data of WHO, GEMS/Food Consumption Cluster Diets [5, 44] on consumption of fish, seafood and algae, the intake of inorganic and total arsenic from these foodstuffs was assessed and compared to the Benchmark Dose Confidence Limit ($BMDL_{05}$) that results in 0.5% increase in lung cancer at 3.0 μg/kg body weight/ per day. Intake of inorganic arsenic compounds for a 70 kg person from only the Hijiki algae would be 4.9% $BDML_{05}$ (0.07 μg/kg b.w./day), whereas with all the other tested products does not exceed the 0.5% $BDML_{05}$, which does not pose a health threat.

Nonetheless, inclusion in the exposure assessment of only total arsenic contamination without differentiating speciation forms leads to much higher intake for the tested samples.

Taking into account mean and 90th percentile contamination, total arsenic intake from particular foodstuff groups will be: 3% and 5% $BDML_{05}$ from fish, 4% and 6% $BDML_{05}$ from fish products, 4% and 9% $BDML_{05}$ from seafood 2% and 3% $BDML_{05}$ from seaweed, respectively.

Estimated exposure based on obtained results from investigated foodstuffs is lower than that reported in other countries [39, 42].

CONCLUSIONS

1. The content of inorganic arsenic in the investigated fish, seafood and seaweed is low and does not pose a health concern.
2. In all of the samples, organic arsenic compounds constitutes over 95% of arsenic, whereas only in the *Hizikia fusiforme* species above 40% were inorganic compounds.
3. It is expedient to conduct speciation studies on different forms in which arsenic is present in foodstuffs, especially the most toxic inorganic compounds, as well as studies on the metabolism and toxicity of organic compounds.
4. The obtained results will provide input to future legislative work on setting maximum permissible levels in EU legislation which would thus facilitate health risk assessments in this area.

Acknowledgement

The study was performed under the scientific project financed by the National Institute of Public Health--National Institute of Hygiene, Warsaw, Poland (No 8/ ZŚ.1. 2011.)

Conflict of interest

The authors declare no conflict of interest.

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Received: 06.04.2015
Accepted: 03.08.2015