



Arsenic speciation in fish and shellfish from the North Sea (Southern bight) and Açú Port area (Brazil) and health risks related to seafood consumption



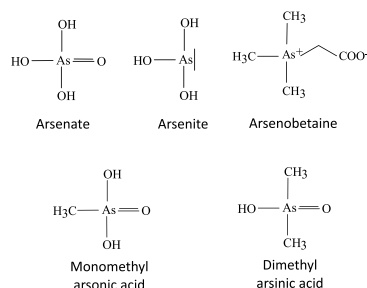
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GRAPHICAL ABSTRACT



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ABSTRACT

In North Sea and Port Açú (Brazil) coastal areas, high arsenic (As) concentrations were observed in water, soil and sediments. Therefore, the impact of this contamination on fish and shellfish species bought from local fishermen was studied. Total As was assessed with ICP-OES (Brazil) and ICP-MS (North Sea) after microwave digestion. Toxic As was assessed with liquid chromatography-ICP-MS (Brazil) and hydride generation-atomic fluorescence spectrometry (North Sea). All analytical methods comply with Quality Assurance/Quality Control procedures.

Several fish species have average Total As concentrations above 1 µg g⁻¹ wet weight (ww), but the highest concentrations are found in less spotted dogfish, lemon sole and whelks from the North Sea, with respectively 50, 49 and 50 µg g⁻¹ ww. High Total As concentrations correspond to high Toxic As concentrations, except for scallops having increased Toxic As concentrations. Toxic As fractions are highest in scallops (almost 10%) but rarely exceeds 2% in all other species. Liver samples were only analyzed in ray, dogfish and catfish and their Toxic As fractions are between 2 and 4 times higher than in muscle.

For a consumption of 150 g of seafood, only 3 samples exceed the provisional total daily intake of 2 µg kg⁻¹ bw, however, cancer risks are non-negligible. Using mean Toxic As concentrations for each of the

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different fish and shellfish species studied, Lifetime Cancer Risk values at the actual global seafood consumption rate of 54 g day⁻¹ are above 10⁻⁴ for whelks, scallops, dogfish, ray and lemon sole.

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1. Introduction

Arsenic is a toxic element, which is widely distributed in the environment, because of both natural as well as anthropogenic sources. As a consequence, humans are exposed to many sources of As (food, water, soil and air), but exposure via diet is by far the most important one. Amongst the food products, seafood supplies most of the total As to humans, with the exception of areas with an endemic high drinking water contamination. The US Food and Drug Administration (US FDA, 1993) indicated that fish and other seafood consumption accounts for 90% of the total human As exposure. Considerable differences in daily intake rates of Total As can be found in the literature mainly resulting from differences in the seafood fraction of the diets. In literature we find that in Canada the total daily intake of As was estimated at 25–35 µg day⁻¹ (Smith et al., 1975), in the US at 56 µg day⁻¹ (Tao and Bolger, 1999), in Tarragona, Spain at 458.5 µg day⁻¹ (Bocio et al., 2005) and in Belgium at 285 µg day⁻¹ (Baeyens et al., 2009). However, total daily As intake rates are not adequate to estimate health risks caused by seafood consumption. Indeed, a distinction between non-toxic and toxic fractions should be made. The inorganic As(III) and As(V) species as well as two organic arsenic species, monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) are the most toxic (Edmonds and Francesconi, 1993; Caumette et al., 2012; Santos et al., 2013). Their sum is considered as the Toxic As fraction (Toxic As) and is used in non-cancer and cancer risk assessment calculations (De Gieter et al., 2002; Xie et al., 2004; Ventura-Lima et al., 2011).

Yet the number of dietary studies making a distinction between total and toxic As species is limited. Daily intake rates for toxic/inorganic As vary between 4.8 µg day⁻¹ (Canada, Yost et al., 1998), 5.6 µg day⁻¹ (UK, TOX/2002), 5.8 µg day⁻¹ (Belgium, Baeyens et al., 2009), 8.3 µg day⁻¹ (US, Yost et al., 1998) and 56 µg day⁻¹ (Bangladesh, Kile et al., 2007). The Bangladesh situation is very special because arsenic deposits cause high inorganic As concentrations in drinking water and foodstuffs (Toxic As is 82% of Total As in food) and as a consequence a high exposure to the population.

North Sea and Açú Port area (Brazil) ecosystems are highly impacted by As but the sources are of different nature. In the Southern North Sea, the average dissolved As concentration is around 0.75 µg L⁻¹ (Millward et al., 1996), but locally the river input can substantially modify the baseline As concentration of the Southern North Sea. Along the Belgian and Dutch coasts the riverine inputs of Rhine/Meuse and Scheldt (270 tons y⁻¹) increase the local dissolved As concentration with a value between 0.8 µg L⁻¹ and 2 µg L⁻¹. Exchange fluxes of dissolved As at the sediment/water interface in the Belgian coastal area range between 30 µg As m⁻² d⁻¹ in April and 120 µg As m⁻² d⁻¹ in February (Gao et al., 2009) and can also increase the As levels in the coastal areas of the North Sea. In Port Açú area, high As levels in small lakes and marine sediments originate from calcareous biota and iron oxy-hydroxides rich in arsenic. Surfaces of calcareous particles serve as the substrate for the fixation of Fe(III) oxy-hydroxides enriched with As. Arsenic levels in interdunes' lake water vary between <1 and 20 µg L⁻¹ depending on the season while they vary between 20 and 270 µg g⁻¹ in the sediments of those lakes. Marine sediments contain around 50 µg g⁻¹ of As, what is of the same magnitude as

the levels in sediments of the small lakes in the interdunes (Mirlean et al., 2012, 2013; Mirlean and Baisch, 2016).

The arsenic species entering the marine environment via riverine or sediment inputs are all of inorganic, hence toxic nature. However, fish mainly contains arsenobetaine, a non-toxic As form, which means that somewhere in the food-chain a transformation from inorganic As (water) to arsenobetaine (fish) should take place.

Our main objective for the present study was to answer the question if the high As concentrations in water, soil and sediments of both marine ecosystems (North Sea and Port Açú area) eventually led to high As (total and toxic) concentrations in fish. Further, specific objectives were to compare (1) As levels in liver tissue and muscle; (2) the total daily intake (TDI) of As from fish consumption, with the WHO TDI of 2 µg kg⁻¹ bw (The Joint FAO/WHO Expert Committee on Food Additives, 1989) and (3) the Individual Lifetime Cancer Risk (TR) due to seafood consumption, with the acceptable risk level of 10⁻⁴.

2. Methods and materials

2.1. Sampling

In the North Sea, fish species ray, dogfish, lemon sole, pouting, pollack, brill, cod, whiting and ling and shellfish species whelks and scallops were studied. Fish (N = 79) and shellfish (N = 37) species were bought from local fishermen who caught the seafood by trawling, in different areas of the North Sea and the English Channel in the period 2009–2010. Prior to analysis, the subsamples were weighed, deep-frozen, lyophilized, weighed again to determine the water content and manually homogenized. Each fish and shellfish species was individually analyzed.

In the Brazilian coastal area of the Atlantic Ocean bigtooth corvina, catfish and shorthead drum (N = 7) were studied. The fish species were caught in 2013 by local fisherman in the Açú Port area, Rio de Janeiro. Prior to analysis, the subsamples were weighed, deep-frozen, lyophilized, weighed again to determine the water content and manually homogenized. Each fish and shellfish species was individually analyzed.

2.2. Total As analysis

Methods used for the determination of Total As in fish make use of microwave assisted mineralization and ICP-OES detection (Brazil) and ICP-MS (Belgium). Full methods can be found in Santos et al. (2013), Leermakers et al. (2006) and Baeyens et al. (2009), while a summary can be found in Table 1. The validation of the method in Brazil included the analyses of reference materials CRM DORM-2 (dogfish muscle) from the National Research Council of Canada (NRCC, Ottawa, Ontario, Canada), BCR627 (tuna fish muscle) from the Institute for Reference Materials and Measurements (IRMM, Geel, Belgium), and SRM1566b (oyster tissue) from the National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA). Recoveries between 91 and 104% were obtained. In Belgium, certified reference materials, DORM-2 (Dogfish muscle) and TORT-2 (Lobster *hepatopancreas*) from the National Research Council of Canada were used during the validation study and recoveries of 106% for DORM-2 and 102% for TORT-2 (Table 1). As a

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