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Factors determining accumulation of bisphenol A and alkylphenols at a low trophic level as exemplified by mussels *Mytilus trossulus*☆

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A B S T R A C T

The aim of the study was to investigate abiotic and biotic factors influencing the accumulation of endocrine disrupting compounds (EDCs) such as bisphenol A (BPA), 4-*tert*-octylphenol (OP) and 4-nonylphenol (NP) in mussels *Mytilus trossulus* from the Gulf of Gdansk (Southern Baltic). The key abiotic factor influencing BPA, OP and NP accumulation in mussels is their hydrophilicity/lipophilicity, which affects their main assimilation routes - by digestive tract for the more lipophilic OP and NP, and additionally by the gills for the less lipophilic BPA. As a result, high condition index (i.e. higher soft tissue weight) is more often correlated with high concentrations of OP and NP in mussels than with BPA. Furthermore, alkylphenols have 6–8 times greater accumulative potential than BPA. Concentration of the studied compounds was lower in females than in males following spawning, and the effect lasted longer for BPA than for alkylphenols. The influence of season and hydrological conditions on BPA, OP, NP in the mussel was more pronounced than the proximity of external sources of these compounds. An increase in water temperature in summer probably stimulated the solubility of BPA, the least lipophilic of the studied compounds, and led to increased assimilation of this compound from water (through gills). On the other hand, high OP and NP concentrations in mussels occurred in spring, which was caused by increased surface run-off and sediments resuspension.

1. Introduction

Bisphenol A (2,2-bis(4-hydroxyphenyl) propane - BPA) and alkylphenols (4-nonylphenol – NP, 4-*tert*-octylphenol – OP) are worldwide high-volume chemicals used in the production of plastics. However, NP in the environment mainly originates from the degradation of nonylphenolethoxylates (NPEOs), common nonionic surfactants used in industrial applications and daily life (Soares et al., 2008). BPA, OP and NP are well known as Endocrine Disrupting Compounds (EDCs) which may interfere with the function of hormones in living organisms (e.g. 17β-estradiol) (Chapin et al., 2008; US EPA, 2010) and are toxic to aquatic organisms (algae, invertebrates, fish and mammals) (Jobling et al., 1996; Servos, 1999; Du et al., 2008). Concentrations of EDCs do not need to be high in order to cause negative and harmful effects for organisms, trace amounts are capable of influencing the activity of natural hormones (Zhong et al., 2012). It is also worth pointing out that some of EDCs metabolites are xenoestrogens. In the mussels *Mytilus edulis*, for example, BPA and its metabolites were shown to affect spawning, by inducing oocyte and ovarian follicle damage (Aarab et al., 2006).

Nonylphenols and octylphenols are considered by the European Parliament in the Framework Water Directive to be substances posing a particular threat to the water environment (Directive 2000/60/EC). Since 2003, a reduction policy has been implemented in the EU for NP and their ethoxylates (Commission Directive 2003/53/EC, 2003), for other compounds the limitations on usage are inconsiderable (BPA – only in products that come into contact with children) or nonexistent (OP). In many countries outside Europe there are still no restrictions concerning the production and usage of phenol derivatives. Historical records of alkylphenol pollution in the Baltic Sea indicated that despite a 50% reduction in NP production in European countries since 2005 (UBA, 2006), the loads of this compound deposited in the region of accumulation bottom of the Baltic exhibit a rising trend (Graca

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et al., 2016). Also in the case of OP, an analogous trend has been observed. In addition, OP reaches very high concentrations in the water of rivers which flow into the Southern Baltic. In extreme cases, these concentrations exceed the so-called Predicted No Effect Concentration (10 ng dm^{-3}, Helsinki Commission, 2010) by nearly sixteen times (Staniszewska et al., 2015a). Not surprisingly then that despite the relatively low usage of bisphenol A and alkylphenols in the catchment of the Gulf of Gdansk (Southern Baltic), where the studies were conducted, and the fast (about two-week) exchange of water in the gulf (Witek et al., 2003), the compounds are detectable on a quantifiable level in the surface water (<1.0–834.5 $ng \cdot dm^{-3}$), in sediments (<0.08–249.08 $ng \cdot g^{-1}$ d.w.) and in organisms (<0.8–798.4 $ng \cdot g^{-1}$d.w.) (Helsinki Commission, 2010; Koniecko et al., 2014; Staniszewska et al., 2014, 2016a; 2015a,b; Graca et al., 2016,b).

BPA and alkylphenols are quite persistent and are expected to bioconcentrate (Jonkers et al., 2003; Ying et al., 2003; Gatidou et al., 2010). According to Flint et al. (2012), because of the low (BPA) and moderate (OP and NP) lipophilicity of these compounds, their bioconcentration generally occurs only with their higher concentrations in the environment. Local hydrodynamic, physicochemical and biological conditions can also influence the distribution and bioavailability of, for example, nonylphenol and their ethoxylates (Ahel et al., 1993). Furthermore, the ability of marine organisms to bioaccumulate EDCs depends on their feeding strategy. A higher concentrations of BPA, OP and NP were found in organisms with the highest exposure to resuspended sediments (e.g. filter feeding organisms) and on the lower trophic levels (David et al., 2009).

This paper determines the main factors influencing the accumulation of bisphenol A and alkylphenols in the mussel *Mytilus trossulus* (Bivalvia), which in the shallow zone of the Southern Baltic is the main component of 1st level consumers (Sokołowski, 2009). This knowledge is essential to the understanding of the accumulation mechanisms of these compounds. Invertebrates are frequently used as biomonitors in the environmental and laboratory studies focusing on bioconcentration of pollutants (Flint et al., 2012). A particularly important group here are mussels, which are commonly used as biomonitoring organisms indicating the pollution levels of aquatic ecosystems. Species belonging to the genus *Mytilus* are particularly well fitted for this role, owing to: widespread occurrence (it is found in both hemispheres), sessile life model, relatively long lifespan, way of feeding (filter feeder), a poorly developed system for metabolising most pollutants, high tolerance to environmental pollution (Farrington, 1991; Kim et al., 2002). In the Baltic Sea a midget form of this species is found - *Mytilus trossulus,* whose size of shell rarely exceeds 6 cm. Their diet consists mainly of suspension (phytoplankton and dead organic matter) and detritus accumulated in the near-bottom water. Mussel colonies filter hundreds of cubic meters of water daily. Both fish and birds feed on mussel beds. Mussels are also gathered and bred for human consumption (Bayne et al., 1976; Wołowicz et al., 2006).

The aim of the present paper was to determine the influence of environmental factors and specimen characteristics (condition, sex) on the accumulation of BPA, OP and NP in mussels. Since the compounds under this study are lipophilic, the assumed hypothesis was that the most important factor affecting the concentration of the studied compounds in the mussel tissues would be their concentration in the ingested food, i.e. in the phytoplankton. Most conclusions on the bioaccumulation of bisphenol A and alkylphenols in mussels are based on laboratory research (Flint et al., 2012). In the present paper the thesis was verified, based on comprehensive environmental studies of BPA, NP and OP concentrations in biotic and abiotic elements of the environment, taking into account seasonality, local hydrodynamic conditions and the distance from potential pollution sources. The study was conducted in the area being under strong anthropogenic pressure (the Gulf of Gdansk), located in the coastal zone of the brackish, semi-enclosed Baltic Sea. Although the study was carried out in a specific area, the obtained conclusions may be universal and they can be extrapolated to mussels from other aquatic water bodies of the world.

2. Materials and methods

2.1. *Mytilus trossulus* collection and preparation

Individuals of *Mytilus trossulus* were taken from onboard a research vessel "Oceanograf II", in the coastal zone of the Gulf of Gdansk (southern Baltic Sea), at 5 stations (Fig. 1), in three seasons of the growing period: spring, summer and autumn of 2011 and 2012.

The following stations were located between 1 and 19 km from the coast, at different depths: UW station (depth 40 m), SP station (depth 17 m), GDY station (depth 12 m), GN station (depth 37 m), ME station (depth 4 m). Station UW was located on the north-east profile of the largest river in Poland - the Vistula (average water flow – 1046 $m^3 \cdot s^{-1}$). This river is 1047 km in length and has a drainage area (urban, agriculture and forest) of 194 424 km^2. Station SP was located near a recreational beach, to the north-east of the mouth of the Kacza River, which flows through forests and urban areas of Gdynia. The river is 14.8 km long and its drainage area is 53.8 km^2 (average water flow – 5.5 $m^3 \cdot s^{-1}$). Potential sources of EDCs at GDY station include the harbour of Gdynia and a yacht marina, outlets of rainwater sewer systems and a recreational beach. ME station, the shallowest, was situated close to the pipe from Dębogórze, the largest wastewater treatment plant in the Tricity agglomeration (Gdansk, Sopot & Gdynia: total population around one million; GUS, 2011). GN station was located ca. 19 km from the Vistula River plume, on the main shipping route to the harbours of Gdansk and Gdynia (Fig. 1).

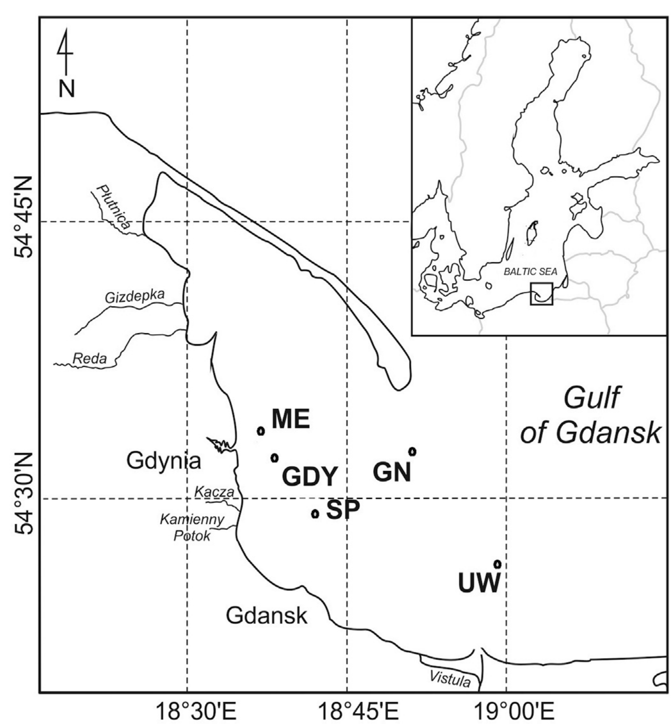

Fig. 1. Location of investigated stations in the Gulf of Gdansk (Southern Baltic).