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Spatial and temporal trends of contaminants in mussel sampled around the Icelandic coastline

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HIGHLIGHTS

- Contaminants have been determined in blue mussels around Iceland for 20years.
- Concentrations of POPs have decreased at most locations in recent years.
- Concentrations of trace metals were fairly stable except for cadmium.
- Local sources of pollution are suspected at three of the locations investigated.

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ABSTRACT

Contaminants have been determined in blue mussels (*Mytilus edulis*) at 11 locations around the Icelandic coastline from 1990 to 2010. The aim of the present study was to investigate if there has been a change in concentration of contaminants around the Icelandic coastline for the last two decades and if the concentrations and changes, if present, were consistent between locations. Concentrations of the persistent organic pollutants, *p,p′*-dichlorodiphenyldichloroethene (*p,p′*-DDE), hexachlorobenzene (HCB), α-hexachlorocyclohexane (α-HCH), polychlorinated biphenyl (PCB-153) and *trans*-nonachlor, have decreased at most of the sampling locations in Iceland in recent years. However, an increasing trend was found at a few locations that could be explained by anthropogenic activity. The concentration levels of the persistent organics were much lower than found at the Norwegian, USA and Chinese coasts, especially levels of *p,p′*-DDE. The concentration of copper and selenium had a consistent pattern of change and concentration between locations over the period which showed a decreasing trend in recent years. The trace elements arsenic, cadmium, mercury and zinc showed more variation in concentration between locations, the concentration of arsenic, mercury and zinc was fairly stable over the period, whereas there were fluctuations in cadmium concentrations. The concentrations of cadmium and zinc were observed to be somewhat higher than found in mussels from Norway, USA and China but values of mercury and lead were much lower in the mussel sampled in Iceland. The higher concentrations of cadmium and zinc can be explained by the volcanic activity in Iceland but no major anthropogenic sources of trace elements are known in Iceland.

1. Introduction

Trace elements, such as metals in the Arctic have both natural geological and anthropogenic sources. Their output from anthropogenic sources increased during the 20th century (Jarup, 2003). In order to mitigate the emission of trace elements, international agreements, such as the OSPAR convention have been implemented. This is considered a necessary action since trace elements, especially arsenic, cadmium, lead and mercury, can cause adverse health effects (Jarup, 2003). Unlike the trace elements, persistent organic pollutants (POPs) mainly come from anthropogenic sources. They have been used extensively in agriculture and industry in the past. However, most countries have signed the Stockholm convention and have consequently banned or restricted the use of POPs due to their persistence and negative effects on the health, reproduction and survival of wildlife and humans (Fisk et al., 2005; Smith and Gangolli, 2002).

POPs and trace elements can migrate from lower latitudes to higher ones by long-range transport (Pacyna et al., 1985; Wania and Mackay, 1993). The contaminants are mainly transported to the Arctic with air, ocean currents and rivers (AMAP, 1998) and these main

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pathways can be affected by climate change (Macdonald et al., 2005). These changes can make interpretation from time-series difficult as trends may also arise from climate change but not only change in emission (Macdonald et al., 2005). Results from spatial and temporal analyses of POPs in Arctic air indicate that the reducing ice cover, increasing temperature of the sea and biomass burning may affect trends in POP concentrations in the Arctic (Hung et al., 2010).

A meta-analysis was carried out by Riget et al. (2010) on temporal trends of POPs in the Arctic biota. Their results revealed that POPs in the Arctic have mostly been decreasing the last two to three decades, even if increasing trends were observed in some cases. The same results were reported from trend analysis of POPs in the Arctic air (Hung et al., 2010). Research on spatial and temporal trends of metals in the Arctic area has mainly focused on mercury (Hg). Temporal trends of Hg in the Arctic biota have shown no consistent pattern of change over the last 30 years (Riget et al., 2011). Nevertheless, a spatial trend for Hg was observed, higher proportion of time-series from Canada and Greenland showed increasing trends than time-series from eastern regions (Riget et al., 2011). Less is known regarding trends of other trace elements in the Arctic such as arsenic (As), cadmium (Cd), copper (Cu), lead (Pb), selenium (Se) and zinc (Zn). Analysis of time series from the marine biota around Greenland and Norway showed no consistent pattern of change for Cd concentration (Green et al., 2011; Riget et al., 2004). The metal Pb was found to decrease in concentration during the period 1990–2010 in the marine biota at the Norwegian coast, and some of the Norwegian time-series of Cu and Zn had either increasing or decreasing trends (Green et al., 2011).

National monitoring programs for environmental conditions in the sea around Iceland have been running since 1989. The program is under the auspices of the Environmental Agency of Iceland on the behalf of Ministry for the Environment. The execution of the program is coordinated by Matís ltd. in cooperation with the Marine Research Institute and the Department of Pharmacology and Toxicology at the University of Iceland. The program is part of the fulfillment of Iceland's obligations to the Stockholm Convention, OSPAR commission and the Arctic Monitoring and Assessment program (AMAP). Contaminants have been measured in blue mussels (*Mytilus edulis*) for 15 to 21 years at 11 locations around the Icelandic coastline (depending on contaminant and location). Blue mussels are common around the Icelandic coast, they are filter feeding organisms that live in intertidal areas attached to secure substrates and are therefore a good biomonitoring species, indicating local pollution at the sampling site. The aim of the present study was to use the monitoring data collected in Iceland to answer the following questions: 1) Has there been a change in concentration of contaminants around the Icelandic coastline for the last two decades? and 2) Are concentrations and changes, if present, consistent between locations?

2. Materials and methods

2.1. Sampling method and locations

Blue mussels (*M. edulis*) with shell length of 4–6 cm have been sampled in autumn (August to October) since 1990 by the Marine Research Institute in Iceland and contaminants in the mussels have been measured for 15–21 years at 11 locations (depending on contaminant and location) around the Icelandic coastline. Each sample contained 50 individuals which were deshelled, pooled and homogenized, making one sample per location each year (see Tables S1 and S2 in Supporting information for details). The samples were kept frozen at −20 °C until analysis was performed.

The sampling sites are shown in Fig. 1. Two of these sampling sites are located close to villages on the Westfjord peninsula, a village of 145 residents is located 4 km from Dvergasteinn and a village of 2600 people is 2 km from Úlfsá. A small waste incineration plant was in operation from 1994 to 2010 about 2 km from Úlfsá. Hvalstöð, Hvaleyri and Hvítanes are all located in the same fjord, Hvalfjörður, in western Iceland, where an aluminum factory started its operation in 1998, a ferro-silicon plant was in operation for the whole period and a whaling station, located close to the site Hvalstöð restarted operation in 2009 and 2010 after a 20 year cease. The sampling sites Dalatangi, Brekka and Botn are in a fjord in the eastern part of Iceland called Mjóifjörður with a population of about 40 people. Salmon aquaculture was carried out in the fjord from 2001 to 2007 and there is a small fish factory at Brekka. Straumur is located beside an aluminum factory not far from the capital area and Hvassahraun is 6 km southwest from Straumur. Grímsey is a remote island in the north with around 80 residents.

2.2. Chemical analysis

2.2.1. Chemical analysis of POPs

POPs were analyzed at the Department of Pharmacology and Toxicology at the University of Iceland. The mussels were extracted wet, basically according to the method of Jensen et al. (1983) as described earlier (Olafsdottir et al., 1995). In short, the tissue was extracted with hexane/acetone/diethyl ether, solvents evaporated at 40 °C under N_2, the residue resuspended in isooctane containing 1,2,3,4-tetrachloronapthalene (the internal standard) and cleaned with concentrated sulfuric acid. Recovery was checked and corrected for by the addition of ε-HCH, *op′*-DDD, PCB #112 and PCB #198 (no. according to IUPAC) to all samples at the first step of the extraction. The fat content was determined gravimetrically.

The individual polychlorinated biphenyls (PCBs) and pesticides were determined by gas chromatography (HP6890) against a six level standard curve (0.5–200 pg/μl) made from the corresponding individual standards and the internal standard from Promochem, Wesel, Germany and AccuStandard, USA. Twelve chlorinated pesticides or their metabolites, hexachlorobenzene (HCB), α-, β- and γ-hexachlorocyclohexane (HCH), α- and γ-chlordane, *trans*-nonachlor, oxychlordane, *p,p′*-dichlorodiphenyldichloroethene (*p,p′*-DDE), *p,p′*-dichlorodiphenyldichloroethane (*p,p′*-DDD), *p,p′*-dichlorodiphenyltrichloroethane (*p,p′*-DDT) and *o,p′*-DDT and 11 PCB-congeners (#28, 31, 52, 101, 105, 118, 138, 153, 156, 170, 180) were determined using two different capillary columns from JW Scientific (DB5, 60 m, 0.25 mm inside diameter, 0.25 μm film thickness and DB1701, 60 m, 0.25 mm inside diameter, 0.25 μm film thickness) and an ECD detector. Helium was used as the carrier gas (25 cm/s) and nitrogen as a make-up gas, splitless injection of 2.5 min, injector temperature 270 °C. Temperature program for DB5: 85 °C for 2 min, 30 °C/min to 200 °C, hold for 29 min, and 1.5 °C/min to 246 °C, 10 °C/min to 310 °C, hold for 8 min; and for DB1701: 85 °C for 1.5 min, 30 °C/min to 200 °C, hold for 28 min, and 2 °C/min to 250 °C, 7 °C/min to 290 °C, hold for 8 min. The limit of quantification was at least 0.01–0.1 μg kg^{-1} ww (wet weight) (0.05–0.25 μg kg^{-1} dw (dry weight)) for the pesticides and the individual PCB congeners.

Quality assurance was ascertained in the laboratory participating twice annually in QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) BT2 and BT5 exercises, the use of blank samples and a mussel reference sample from QUASIMEME was extracted and run with every analysis. The standard solutions were checked by comparison to certified reference material (NIST1493, Promochem, Germany).

2.2.2. Chemical analysis of trace elements

The analysis of trace elements (As, Cd, Cu, Hg, Pb, Se and Zn) was carried out at Matís ltd. The samples analyzed before 2007 were analyzed using cold vapor atomic absorption (Hg), FAA/impact bead using D_2-background correction (Cd, Cu, Zn, Pb) and hydride generation atomic absorption (As, Se) as described by Yngvadóttir et al. (2006) after sample digestion in 50 ml Parr digestion bombes as described by Rabieh et al. (2007). Samples analyzed in the year 2007 and later were analyzed with microwave digestion (Mars5, CEM, Matthews,