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Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota

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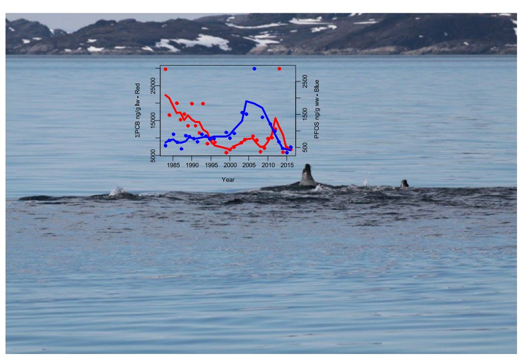
HIGHLIGHTS

- Temporal trends of POPs in Arctic biota.
- Downward trends constitute the majority of statistically significant trends.
- "Newer" POPs show a more mixed pattern of trends.
- Continuing existing time-series would generally lead to more powerful trend detection.

GRAPHICAL ABSTRACT



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ABSTRACT

More than 1000 time-series of persistent organic pollutants (POPs) in Arctic biota from marine and freshwater ecosystems some extending back to the beginning of 1980s were analyzed using a robust statistical method. The Arctic area encompassed extended from Alaska, USA in the west to northern Scandinavian in the east, with data gaps for Arctic Russia and Arctic Finland. The aim was to investigate whether temporal trends for different animal groups and matrices were consistent across a larger geographical area. In general, legacy POPs showed decreasing concentrations over the last two to three decades, which were most pronounced for α-HCH and least pronounced for HCB and β-HCH. Few time-series of legacy POPs showed increasing trends and

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only at sites suspected to be influenced by local source. The brominated flame retardant congener BDE-47 showed a typical trend of increasing concentration up to approximately the mid-2000s followed by a decreasing concentration. A similar trend was found for perfluorooctane sulfonic acid (PFOS). These trends are likely related to the relatively recent introduction of national and international controls of hexa- and hepta-BDE congeners and the voluntary phase-out of PFOS production in the USA in 2000. Hexabromocyclododecane (HBCDD) was the only compound in this study showing a consistent increasing trend. Only 12% of the long-term time-series were able to detect a 5% annual change with a statistical power of 80% at $\alpha < 0.05$. The remaining 88% of time-series need additional years of data collection before fulfilling these statistical requirements. In the case of the organochlorine long-term time-series, 45% of these would require >20 years monitoring before this requirement would be fulfilled.

1. Introduction

Persistent organic pollutants (POPs) are chemicals that have a long lifetime in the environment and due to their physical-chemical properties, they are transported over long distances. POPs enter food-webs and accumulate, and in some cases biomagnify in wildlife and humans. Several global and regional conventions were developed with the goal of eliminating or reducing emissions of POPs. The Stockholm Convention on POPs initially addressed twelve priority POPs and since then these have been extended to 28 POPs as of 2017 (http://chm.pops.int). Despite the fact that several of these POPs were banned in the 1970's and 1980's and others have been restricted, they are still found in the environment at levels that may cause adverse effects to the health of top predators in Arctic food chains (Letcher et al., 2010). Humans living in the Arctic and consuming significant amounts of high trophic traditional food are exposed to legacy POPs, which may lead to adverse health effects (AMAP, 2009).

Temporal trend studies are an important means of assessing the fate of contaminants in ecosystems. They can provide a first warning that potentially harmful compounds may be increasing in the environment, e.g. in biota regarded as indicator organisms. Temporal trend studies may also indicate whether regulatory actions aimed at reducing harmful chemicals in the environment are proving successful, or whether environmental levels are approaching or exceeding threshold values for biological and possibly toxic effects. Several Arctic countries perform POP monitoring in biota, focussing on freshwater and marine ecosystems, resulting in time series of varying statistical power. Statistical power is an important consideration in relation to temporal trend monitoring. The power of a temporal trend represents the statistical probability of detecting a change of a given magnitude when this change actually occurs. It is desirable that monitoring data series have sufficient statistical power to minimise false negatives, i.e. the risk of incorrectly concluding that no change has occurred.

Rigét et al. (2010) assessed temporal trends of POPs in Arctic biota based on the time-series available up until 2008 and in the context of the Arctic Monitoring and Assessment Programme (AMAP). A large number of new time-series data has become available since then and additional years have been added to the time-series included in Rigét et al. (2010). Therefore, it was expected that this update would lead to a more robust assessment than the previous one. The objective of this study was to analyze trends for POPs in all time-series available from the Arctic, including an evaluation of their statistical power.

2. Datasets and statistical analyses

Time-series of POPs were available from seven Arctic countries for a total of 64 location-species-tissue combinations. Fig. 1 shows the locations, together with the species or species group monitored. No series in biota are currently available for the Arctic areas of Russia and Finland. Time-series were available for marine mammals, seabirds, marine and freshwater fish and blue mussels.

Asmund et al. (2004) focused early on the quality assurance/quality control (QA/QC) of the Danish and Canadian laboratories, which were involved in the Greenlandic AMAP trend program. AMAP has worked towards harmonized programs with respect to methodologies and QA/QC in order to ensure the quality and credibility of AMAP assessments. For example, AMAP has established guidelines that cover all aspects of data generation from sample collection, handling and processing, to analysis, and data management, and that must be followed by the laboratories contributing to AMAP assessments. The laboratories responsible for the POP analyses included in this study participate in a number of QA/QC programs including the AMAP/NCP inter-laboratory studies (e.g. Tkatcheva et al., 2013), the QUASIMEME laboratory performance testing scheme (www.quasimeme.org), and equivalent QA/QC programs run by NOAA/NIST. All laboratories have established common internal QA/QC procedures and measures such as the use of internal and certified reference materials, analyses of blank and duplicates etc. Each individual time-series has been analyzed by the same laboratory with the same method over several years.

Time-series were separated into long-term time-series starting before the year 2000 and short-term time-series starting in or after the year 2000. Several of the short-time time-series were derived by deleting data from years before 2000 of the long-term time-series. In total, 1074 long-term time-series and 735 short-term time-series were available covering the following POPs: Polychlorinated biphenyls (PCBs) congeners, dichlorodiphenyltrichloroethane (DDT) and its transformation products, hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and pentachlorobenzene (PeCB), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and pentachlorobenzene (PeCB), chlordane-related pesticides (CHL) and heptachlor, dieldrin, mirex, octachlorostyrene (OCS), toxaphene, polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD) and perfluoroalkyl substances (PFASs). Octachlorostyrene (OCS) was included although not included in the Stockholm Convention because time-series were available and only few time-series study has been published. The distribution of available time-series by country, species group and time-series type are shown in Table 1.

Datasets were treated as in previous assessments of temporal trends of contaminants in Arctic biota (Rigét et al., 2010; AMAP, 2014, 2016). Only time-series with six or more years of data were included. Generally, time-series were considered inappropriate for trend analyses if three or more years had annual medians that were less than the reported detection limit. However, time-series were included if these years were concentrated in the end of the time-series, indicating a decreasing trend or in the beginning, indicating an increasing trend. In these cases, half of the detection limit was used to represent the actual annual medians.

A statistically robust method was applied to the time-series data. Covariates such as age, sex and lipid content were treated as recommended by data originators such as selecting a subset of similar characteristics e.g. sex and/or age. All time-series were treated with the same statistical method and therefore highly comparable results have been generated.