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Bioaccumulation of HCHs and DDTs in organs of Pacific salmon (genus *Oncorhynchus*) from the Sea of Okhotsk and the Bering Sea

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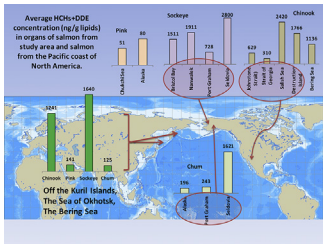
HIGHLIGHTS

- Pacific salmon were collected in the Bering Sea and Sea of Okhotsk.
- OCPs concentration in sockeye and chinook were higher than in pink and chum.
- HCHs concentration was higher than that of DDE.

GRAPHICAL ABSTRACT



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ABSTRACT

Concentrations of isomers of hexachlorocyclohexane (α-, β-, γ-HCH) and dichlorodiphenyltrichloroethane (DDT) and its metabolites (dichlorodiphenyldichloroethane (DDD) and dichlorodiphenyldichloroethylene (DDE) were assessed in organs of the pink (*Oncorhynchus gorbuscha*), chum (*Oncorhynchus keta*), chinook (*Oncorhynchus tshawytscha*), and sockeye salmon (*Oncorhynchus nerka*), caught near the Kuril Islands (the northern-western part of the Pacific Ocean), in the Sea of Okhotsk and the Bering Sea. Pesticides have been found to accumulate in fish organs in the following: muscles < liver < eggs < male gonads. The highest concentrations in muscles and liver have been recorded from sockeye. Of the DDT group, only DDE has been detected. The average concentration of HCHs + DDE in the muscles of pink, chum, chinook, and sockeye was 141, 125, 1241, 1641 ng/g lipids, respectively; and in the liver, 279, 183, 1305, 3805 ng/g lipids, respectively. The total concentration of HCHs isomers was higher than that of DDE. Average HCHs + DDE concentration in organs of salmon from study area is lower than that in salmon from Pacific coast of North America.

1. Introduction

One of the most toxic groups of pollutants in the biosphere is persistent organic pollutants (POPs). These xenobiotics are characterized by a high stability, toxicity, and lipophilic properties, which facilitate their transfer up the food chains and accumulation (biomagnification) in organisms of higher trophic levels. The

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atmospheric transport of volatile POPs is the main route of their global migration and distribution. From the zones, where POPs have been or are being used, they are transported by winds to high latitudes, up to the Arctic, and there accumulate in various components of ecosystems (Wania and Mackay, 1995; Wania, 2003). Marine currents are another route of transfer (AMAP, 1998). As a result, the northern Pacific Ocean has become a polluted zone.

Among POPs, the quite hazardous compounds by their distribution and impact on biota are organochlorine pesticides (OCPs), including isomers of hexachlorocyclohexane (HCHs), dichlorodiphenyltrichloroethane (DDT) and its metabolites (dichlorodiphenyldichloroethane (DDD) and dichlorodiphenyldichloroethylene (DDE), which were particularly widely used from 1950s to 1970s. Now they are distributed in the environment and found in all regions of the planet (Jones and de Voogt, 1999). Marine ecosystems frequently become the terminal sink of accumulation of these compounds (Tanabe, 2007). POPs have a high affinity to lipids, a low rate of disintegration and removal, and may be accumulated in adipose tissues of fish, birds, and marine mammals. Concentration of pesticides in fish, used as an important item in the human diet, is especially hazardous.

Among fish that inhabit the upper epipelagic layer of the northern Pacific, the most abundant group is Pacific salmon of the genus *Oncorhynchus*. By the type of their life strategy, they are referred to as anadromous fish, which have the ocean period of life, spent in feeding grounds, and whose spawning and early development occur in fresh water of rivers and lakes. Juvenile fish migrate downstream to the sea, where they feed for some period, from 1 to 7 years depending on species, and then come back to their breeding grounds to spawn. The geographic range of distribution of these fishes is associated with cold and temperate waters. Their marine ranges are continuous, stretching throughout the North Pacific. Chum and pink salmon have the most extensive distribution during their marine and freshwater periods of life history, from the Arctic to the subtropical zone. The major species spawning on the coast of the Russian zone of the Far Eastern seas are pink (*Oncorhynchus gorbuscha*), chum (*Oncorhynchus keta*), coho (*Oncorhynchus kisutch*), chinook (*Oncorhynchus tshawytscha*), and sockeye salmon (*Oncorhynchus nerka*). The most abundant of them are chum and pink salmon (Shuntov and Temnykh, 2011).

During their feeding season and particularly prior to the spawning migration, salmon accumulate reserves of neutral lipids, both to satisfy their energy needs and to develop gonads while migrating. Accumulation of lipophilic pollutants occurs along with the build up of lipid reserves, both in subtropical latitudes and in the temperate zone. The levels of pesticide content in salmon spawning on the Pacific coast of North America were studied by US and Canadian researchers (Hardell et al., 2010; Good et al., 2014; Cullon et al., 2009). In the Russian zone of the northwestern Pacific Ocean, significant OCPs concentrations were earlier found in grey whales, Pacific walruses, and sea birds from the Sea of Okhotsk and the Bering Sea (Tsygankov, 2012; Tsygankov et al., 2014a, 2014b, 2015). It was shown also that pink and chum salmon, as the most abundant species of Pacific salmon, perform the pesticide transfer from ocean to freshwater environments (Lukyanova et al., 2014). In this work, we provide data on pesticide concentrations in four species of Pacific salmon of the genus *Oncorhynchus* and discuss the level of accumulation of the pollutants in these species compared to other fishes of the northern Pacific Ocean.

2. Materials and methods

2.1. Study sites and samples

Samples of organs from fish of four *Oncorhynchus* species—pink (*O. gorbuscha*), chum (*O. keta*), sockeye (*O. nerka*), and chinook (*O. tshawytscha*)—were analyzed. The samples were collected during expeditions organized by Pacific Research Fisheries Center (TINRO-Center): pink and chum were caught in the southern Sea of Okhotsk, off the Kuril Islands, in June 2012 and 2013, during pre-spawning migrations; sockeye and Chinook were caught in the western Bering Sea and in the Sea of Okhotsk in October and November 2010 and 2011, during feeding ground (Table 1, Fig. 1). The fish were dissected, their organs were separated from the body, frozen at −20 °C, and delivered to the laboratory for the further analysis. In pink and chum, the organs subjected to analysis were muscles, liver, male gonads, eggs, and whole fish; in chinook and sockeye, muscles and liver.

2.2. Chemical, QA/QC and data analyses

Before chemical analysis, whole fish and separate organs were homogenized mechanically. Lipids were extracted from homogenized tissues (20 g) by means of *n*-hexane extraction, with subsequent disintegration of the fat components by concentrated sulphuric acid (Tsygankov and Boyarova, 2015).

Concentrations of organochlorine pesticides (HCH isomers (α-, β-, γ-HCH), DDT and its metabolites (DDD, DDE)) in samples were measured by a gas chromatograph Shimadzu GC-2010 Plus with an ECD (electron capture detector) (capillary column Shimadzu HiCap CBP5). Column temperature – 210 °C, injector – 250 °C, and detector – 280 °C. Carrier gas is argon, inlet pressure: 2 kg/cm^2, 1:60 flow divider, and flow rate of carrier gas through the column: 0.5 ml/min.

Laboratory blank samples were extracted and analyzed on a regular basis. Retention times for the standard samples were constant and were therefore relied upon for component identification. To identify individual substances, standard working solutions of OCPs in the concentration range of 1–100 mg/ml were applied. The calibration lines showed excellent linearity in the range of the concentrations of interest. To determine the quality of the methodology, a recovery study was performed using standard addition methods. Seven fish tissue samples were spiked with the mixture of pesticides standards. The spiked samples were extracted and analyzed as described in the method above. The results revealed that the mean recovery values ranged from 85.1 to 98.6%. This indicates that the analytical procedures outlined for the OCPs determination in this study were reliable, reproducible and efficient.

The statistical analysis of the results was performed in the software package IBM SPSS Statistics 21 for Mac OS X. Significance of the obtained data was evaluated by using the Mann-Whitney *U* test with the significance level of $p \leq 0.05$.

3. Results

HCH isomers and DDE were found in all analyzed samples (Tables 2 and 3). The total content of pollutants in various organs varied within a wide range, from 41 to 7103 ng/g lipids. In general, the pesticide concentration increased in the following order: muscles < liver < eggs < male gonads. The maximum OCPs concentration in individual fish was recorded from sockeye liver (7103 ng/g lipids), where HCHs constituted 6453 ng/g that was also the maximum value for the studied salmon (Table 3). The highest concentration of DDE was found in chinook liver (3022 ng/g). DDE in all the samples was the only registered DDT metabolite that indicates destruction of initial DDT, i.e. long-term presence of this pesticide in the ecosystem (Tanabe and Subramanian, 2006). All three HCH isomers were detected in pink and chum; the total content of α- and β-HCH was higher than the level of γ-HCH. α- and β-HCH are the most stable isomer and typically constitute the