ISSN 1995-4255, Contemporary Problems of Ecology, 2019, Vol. 12, No. 6, pp. 562–574. © Pleiades Publishing, Ltd., 2019.
Russian Text © The Author(s), 2019, published in Sibirskii Ekologicheskii Zhurnal, 2019, No. 6, pp. 688–703.

Current Levels of Organochlorine Pesticides in Marine Ecosystems of the Russian Far Eastern Seas

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Received February 19, 2019; revised March 25, 2019; accepted March 27, 2019

Abstract—This review provides information on major organochlorine pesticides (OCPs), which were applied in agriculture worldwide in the 1940s–1960s and are still in use in some developing countries (such as India and China). Patterns of their distribution in the environment, toxicity, metabolism, and degradation are described. Their distribution over the components of ecosystems in different regions of the World Ocean, including the Far Eastern seas of Russia (the Sea of Japan, the Sea of Okhotsk, and the Bering Sea), is characterized over the period of 2000–2016. In the Sea of Okhotsk and the Bering Sea, the OCP content of marine organisms is lower than that in other regions of the World Ocean, in particular, in the Sea of Japan. Results show that a pesticide "background" has formed on the planet. The OCP concentrations in the Sea of Okhotsk and the Bering Sea can be considered a background level, while the Sea of Japan is exposed to contamination from countries using these substances in agriculture.

Keywords: organochlorine pesticides, HCH, DDT, marine ecosystems, Sea of Okhotsk, Bering Sea, Sea of Japan

DOI: 10.1134/S199542551906009X

INTRODUCTION

Among persistent organic pollutants (POPs), organochlorines (OCs), primarily organochlorine pesticides (OCPs)—metabolites of dichlorodiphenyltrichloroethane (DDT) and isomers of hexachlorocyclohexane (HCH) previously widely used in industry and agriculture across the world due to their high toxicity and resistance—are considered the most hazardous agents in terms of ubiquitous distribution and impacts on living organisms.

The composition of a technological HCH mixture included various HCH isomers with different cycle conformations: α-HCH made up 55–70%; β-HCH, 5–14%; and γ-HCH, 9–13%. Preparations with a different composition containing 25 and 90–99% γ-HCH (lindane) as the active agent were also produced (Zulidov et al., 2002). A technological preparation of DDT was based on dichlorodiphenyltrichloroethane, represented by two main isomers, *p*,*p*'-DDT and *o*,*p*'-DDT, which differ in the position of chlorine atoms in the benzene rings, and by other isomers with minor concentrations (Tsydenova, 2005).

The leading criteria in assessing the behavior of OCPs are their persistence (stability) in the environment, cumulative properties, and consequences in the case of bioaccumulation. OCPs are accumulated up food chains and are capable of biomagnification, which can result in serious negative impacts on organisms occupying the top of the trophic pyramid, including humans.

Currently, the use of such compounds is prohibited in most countries of the Northern Hemisphere, but they continue to be applied in Southeast Asia. Furthermore, approximately 45–80% of OCPs migrate to other regions of the planet, including the territory of the Russian Federation (Rovinskii et al., 1990; Ivanter and Medvedev, 2007; Agapkina et al., 2010, 2011; German et al., 2010; Chuiko et al., 2010; Mamontova et al., 2012).

Aquatic ecosystems frequently become the terminal link in the accumulation chain of these compounds. The sources of OCP pollution of the marine environment can be leakage from storage sites, surface runoff from catchment areas of waterbodies, falling with precipitation, and atmospheric circulation and transport by sea currents. In marine ecosystems, OCPs are accumulated up the food chain and reach maximum values at the highest trophic levels.

Distribution and transformation of POPs in marine organisms from different parts of the world's

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oceans are described in various works (Apeti et al., 2013; Buckman et al., 2004; Tanabe, 2007; etc.). In this respect, the Far Eastern seas have been studied to a much lesser degree. The goal of the present review is to summarize the results of recent research (2010–2016) on levels of OCPs in marine organisms of various trophic levels from the Sea of Okhotsk, the Sea of Japan, and the Bering Sea for assessing the position of the region in the World Ocean by the distribution pattern of POPs.

Patterns of OCP Distribution in the Environment

After entering the biosphere, POPs become involved in various physical and chemical processes. Their resistance to photochemical, chemical, and biological decomposition in the atmosphere, water phase, and soil lead to their long-term circulation in the environment. Despite the considered group of POPs being characterized by low vapor pressure values, they show a noticeable ability to pass into the vapor/gas phase, i.e., evaporate into the atmospheric air, for instance, from the surface of soil, water, etc., and circulate between different components of the environment (Wania et al., 1998; Wania and Mackay, 1996; Rovinsky et al., 1990).

Currently, POPs are distributed ubiquitously, as is evidenced by the facts of their detection in both abiotic and biological samples from various sites of the planet. Findings of POPs in Arctic and Antarctic regions, geographically remote from the possible sources of emission of these compounds, also confirms their worldwide distribution (Bidleman, 1999; Macdonald et al., 2000; Negoita et al., 2003; van den Brink, 1997). The behavior in the environment and distribution of OCPs in various components of natural ecosystems are determined by their physical and chemical properties.

Metabolism and Degradation of OCPs

The main mechanisms of OCP degradation in the environment can be divided into abiotic (photochemical reactions) and biotic processes of metabolic degradation, with the involvement of living organisms.

The photochemical decomposition of OCPs, whose molecules contain aromatic groups and unsaturated chemical bonds, occurs due to the absorption of solar energy in the ultraviolet and visible regions of the spectrum (Crosby, 1979; Tinsley, 1979). The rate of photochemical decay, as well as the composition of the final products of this reaction, depends on the environment where this process occurs.

In animals, the degree of OCP accumulation is determined by the ratio of two processes: absorption and excretion. The general trend of metabolism consists in turning exogenous matter into a more polar compound with subsequent binding the resulting product with a highly polar fragment to facilitate its excretion. In plants, which lack excretory systems, exogenous substances (or their metabolites) are usually conjugated with carbohydrates and deposited in parts that are not related with general metabolism. In insects, the set of hydrolases is less than in mammals. Therefore, they cannot perform rapid neutralization, and OCPs can accumulate to lethal concentrations (Isidorov, 1999).

The accumulation of DDT depends on the lipid content of organs. Thus, the dependence of DDT accumulation can be arranged in the following sequence: fat > liver > muscles (Lukyanova et al., 2016). The increased lipid content of fish from pesticide-contaminated areas can be considered obesity and an adaptation to habitat conditions (Maslova, 1981; Tanabe and Subramanian, 2006; Tanabe, 2007).

HCH is less persistent than DDT and is more easily subject to microbial degradation. Studies on model waterbodies have shown that the γ-HCH concentration in water decreases from 0.50 to 0.27 mg/L within a day after introduction; in silt, it increases from 0 to 0.34 mg/kg; in higher aquatic plants, it increases from 0 to 2.3 mg/kg. The accumulation coefficient (C_a) of γ-HCH for sediments was estimated at 1.3–4; for aquatic plants, from 8 to 67 (Vrochinskii et al., 1980; Tanabe and Subramanian, 2006).

As exemplified by the northwestern Pacific Ocean, the α-isomer of HCH dominates surface waters and zooplankton, reaching 55–56%; γ-isomer amounts to 35–40%; and the proportion of the β-isomer is the smallest, lower than 10%. In cephalopods, the β-isomer amount is larger and constitutes approximately 20%. In the tissues of the striped dolphin, the proportions of the isomers are different: the β-isomer dominates (reaching 80%), followed by the α-isomer (10–15%), and the level of the γ-isomer is minimum (Tanabe et al., 1984).

Concentration and proportions of the HCH isomers in the components of the marine environment, open and enclosed waterbodies, and living organisms depend on many factors such as physical and chemical properties of water and sediments, illumination, species-specific characteristics of biotransformation processes, and the duration of pesticide exposure in the environment. The transition of the main part of OCPs dissolved in river water into the suspended form occurs in ecosystems of river deltas and estuaries, at the river–sea interface, where the water salinity sharply increases. With increasing salinity, the solubility of OCPs in water reduces and they pass into suspension (Israel and Tsyban, 2009). Thus, the increased OCP content of organs of riverine and estuarine species, compared to that of marine species, is associated with this process (Tanabe, 2007).

The effects of the abovementioned factors and many others lead to the accumulation of the most persistent β-form of HCH in organisms over time and up food chains (Tanabe et al., 1984). The ratio of concentrations of α- and γ-isomers of HCH is used to estimate the period from the entry of pesticides into an ecosystem. A high value of the ratio, greater than unity, indicates a long presence of OCPs in the environment; a value below unity, i.e., the dominance of the γ-isomer, is the evidence of a "fresh" influx (Rovinskii et al., 1990).

DDT exists in the form of the main product and its metabolites, DDD and DDE. The lifetime of DDT in objects is assessed as the ratio of concentration of DDT to that of DDE, a product of its degradation. High values of the DDT/DDE ratio show a recent entry of DDT into the environment; a low value means that it has long been in the system and is gradually turning into DDE.

Thus, concentrations and proportions of isomers and derivatives of OCPs in the ecosystem are constantly changing and depend on the physical and chemical properties of water and sediments, as well as on the duration of the exposure of the pesticides in the environment.

Toxicity of OCPs

By the middle of the 20th century, the hazard of organochlorines to terrestrial and aquatic organisms was most fully disclosed. The danger that insecticides pose to fish and aquatic invertebrates became apparent due to studies conducted in the 1940s–1950s. In the 1960s, it was found that an acute toxic effect of these substances on the most sensitive aquatic organisms is manifested within a concentration range from 10^{-3} to 10^{-12} g/L (Braginskii et al., 1980). Such a high sensitivity to low concentrations is explained, on one hand, by the extreme toxicity of these substances and, on the other, by the specific pattern of their impact on vital functions. They easily affect any members of arthropods, in particular, crustaceans that constitute the major part of marine and freshwater zooplankton.

The mortality of aquatic organisms of common species reduces the self-purification capacity of an ecosystem, as the life activity of bacteria, algae, crustaceans, mollusks, and other species provides the transformation of organic matter in a waterbody.

OCPs can suppress the endocrine system in aquatic animals, especially during critical periods of their life cycle. Entering through the environment or up a food chain, OCPs can affect the immune system of birds and mammals and cause deviations in the sex characteristics of some individuals, thus, changing the sex structure of populations. It has been found that most animals containing high concentrations of OCPs suffer from tumors (Tanabe and Subramanian, 2006).

According to a report (*State of Canada's environment*, 1996), OCPs pose a threat to living organisms at three levels: genetic, population, and ecosystem. Their negative impact on the hormonal and enzyme systems of terrestrial and aquatic species causes genetic dysfunctions and a reduced survivability of individuals.

As a result of an ecotoxicological assessment of the impact of these compounds on marine mammals (Colborn and Smolen, 1996), a total of 65 epidemiological disturbances were identified, including endocrine suppression, immunological and reproductive dysfunction, neoplasms, and population reduction.

Measures to control environmental pollution require certain sanitary and hygienic standards of permissible levels of harmful chemicals in the air, water, soil, and food. Their regulation is based on determining the minimum values of concentrations of pollutants that ensure safety for human health and the environment. The safe levels of OCPs in the air, water, soil, and cattle feed are provided in many studies and regulations (*GN 1.2.2701–10*, 2010; Rovinsky et al., 1990; Tanabe, 2007; etc.). In Russia, the hygienic standards of seafood safety for humans and the requirements for manufacturing, importing, and marketing foodstuffs to comply with these standards (SanPiN 2.3.2.1078–01) establish the highest concentrations of HCH and DDT for fish liver: 1.0 and 3.0 mg/kg, respectively.

Examples of OCP Distribution in Ecosystems of the World Ocean

Due to their lipophilicity, organochlorine compounds are capable of biomagnification and accumulate from the lowest to highest trophic levels. After entering the aquatic environment, OCPs are bound to suspended particles, precipitate onto the bottom, and build up in sediments. Thus, in the Bay of Bengal (Indian Ocean), the maximum OCP concentration of bottom sediments was 11.36 μg/kg dry weight (d.w.) (Rajendran et al., 2005); in the Yamuna River (India), 209.5 μg/kg d.w. (Bhupander et al., 2011). In the rivers of the city of Tianjin (China), the maximum concentration reached 337 μg/kg d.w. (Lu et al., 2013). On the East China Sea coast, the OCP concentration of bottom sediments varied within 0.1–7.2 μg/kg d.w. (Lin et al., 2012); in the northern South China Sea, from 0.04 to 3.9 μg/kg d.w. (Chen et al., 2006); in the northwestern Yellow Sea, from 0.2 to 9.3 μg/kg d.w. (Hu et al., 2009). In the estuarine zone of rivers emptying into Peter the Great Bay (Sea of Japan), the maximum level was recorded from the Razdolnaya River (45.4 μg/kg d.w.) (Lukyanova et al., 2012). Bottom sediments of the continental seas also show presence of OCPs. For instance, in the Mediterranean Sea off the coast of France, the concentration of toxicants amounts to 255 μg/kg d.w. (Wafo et al., 2006); off the Black Sea coast of Turkey, 108 μg/kg d.w. (Bakan and Ariman, 2004).

OCPs are also present in the water column, where they are sorbed by suspended organic particles. Thus, off the city of Osaka (Sea of Japan, Pacific Ocean), the

OCP content of water varies within a range of 0.5–2.7 ng/L (Kawanishi et al., 2005); in the Bay of Bengal (Indian Ocean), from 5.6 to 12.5 ng/L (Rajendran et al., 2005). OCP concentrations along the California coast are distributed almost uniformly. For instance, in three bays of the state of California—Monterey, Half Moon, and San Pablo—the concentrations were 44.3, 39.4, and 47.9 ng/L, respectively; in the water near the Golden Gate Bridge in San Francisco, 16.5 ng/L (Menzies et al., 2013). In waters of the rivers of Ganges (India) and Jiulong (China), the maximum OCP levels reached 174 ng/L (Sinha, 2003) and 415 ng/L (Maskaoui et al., 2005), respectively.

Organochlorine compounds are well accumulated by filter feeders such as bivalves, so these organisms are successfully and widely used as bioindicators for monitoring OCPs in natural waters.

In the framework of the Asia-Pacific Mussel Watch (APMW) program, marine environment pollution was monitored using mussels and oysters as bioindicators during the period 1997–2000 (Tanabe, 2000). OCPs were found in all mussel samples from the countries participating in the program (Cambodia, China, Hong Kong, India, Indonesia, Japan, Korea, Malaysia, Philippines, the Russian Far East, Singapore, and Vietnam) (Tanabe and Subramanian, 2006). Significant residual concentrations of DDT and α-HCH were detected in many samples. The total concentrations of HCH isomers and DDT with metabolites in soft tissues of mussels from waters of the developing Asian countries were higher than in samples from the developed countries. For instance, the maximum HCH content of the mussel *Perna viridis* was 430 ng/g lipids in India and 20 ng/g lipids in the Republic of Korea and Japan. Maximum concentrations of DDT were recorded from Hong Kong (61000 ng/g lipids) and China (34000 ng/g lipids), whereas in Japan they reached only 100 ng/g lipids. In mussels of the genus *Mytilus*, the level of DDT in China amounted to 29000 ng/g lipids (Monirith et al., 2003); in Hong Kong, 8000 ng/g lipids; on the Pacific coast of Russia, 900 ng/g lipids; in India, Japan, Korea, Vietnam, Singapore, Malaysia, Indonesia, and Cambodia, less than 800 ng/g lipids (Minh et al., 2002). HCH in mussels from the above countries did not exceed 120 ng/g lipids (Tanabe, 2007).

Fish, as a link of food chain, accumulate POPs in their organs and tissues during the biomagnification process. Fish are distributed ubiquitously and, in most cases, reflect the OCP levels in the environment. The content of pollutants in them depends on many factors, one of which is migration. Japanese researchers have conducted a number of studies on migratory (*Diaphus theta* and *Ceratoscopelus warmingi*) and nonmigratory (*Stenobrachius nannochir* and *Lampanyctus regalis*) species and concluded that OCP concentration in the former (up to 180 ng/g lipids) is lower than those in the latter (up to 310 ng/g lipids) (Takahashi et al., 2000; Tanabe and Subramanian, 2006; Tanabe, 2007).

Ueno et al. (2003) used skipjack tuna for monitoring coastal waters of Japan, Taiwan Island, Philippines, Indonesia, the Seychelles Islands, Brazil, and India. The maximum OCP content was found in fish caught from the coastal waters off China (700 ng/g lipids).

Kajiwara et al. (2003) measured OCP concentrations in five sturgeon species (beluga sturgeon, Russian sturgeon, stellate sturgeon, Persian sturgeon, and bastard sturgeon) from the Caspian Sea, which is the world's largest inland body of water bounded by the territories of Russia, Azerbaijan, Kazakhstan, Turkmenistan, and Iran. The maximum contents were recorded from beluga (17700 ng/g lipids) and Russian sturgeon (3050 ng/g lipids).

Pacific salmon are among the most abundant commercial fish. The level of organochlorine pollutants in them is distributed in such a way that the "high-fat" species (chinook and sockeye salmon) show higher concentrations than the "low-fat" species (chum and pink salmon). For instance, OCPs in chinook salmon from the Salish Sea (Washington) amount to 2420 ng/g lipids (Good et al., 2014); in sockeye salmon from Alaska, 1911 ng/g lipids (Apeti et al., 2013); in chum and pink salmon from Alaska, 243 and 80 ng/g lipids, respectively (Gerlach, 2013).

Seabirds can be both intermediate and terminal links in a trophic chain. By feeding on living organisms, they accumulate toxic pollutants in their organs in the process of biomagnification. The OCP content of their organs and tissues also depends on their type of feeding and migration patterns. Thus, glaucous gull from the Barents Sea, feeding on fish, carrion, and eggs of other birds, contains 216157 ng OCP/g lipids in its body (Knudsen et al., 2007). Great skua (Iceland), whose diet consists mainly of fish, accumulates pollutants to a level of 55600 ng/g lipids (Jorundsdottiro et al., 2010). Fish of the Baffin Sea (Arctic Ocean) accumulate smaller amounts of OCPs due to the remoteness of the region from the potential sources of pollution. For instance, northern fulmar, feeding on crustaceans; fish; squid; plankton; and, by chance, carrion, contains toxicants at a level of 4934 ng/g lipids; black-legged kittiwake, whose type of feeding is similar to that of fulmar, contain 1323 ng/g lipids (Buckman et al., 2004).

All OCPs are highly lipophilic compounds, and fat deposits in the subcutaneous layer of marine mammals serve as a storage site for them. Mammals, when compared to most other marine organisms, live longer and are therefore subject to long-term exposure to xenobiotics. Marine mammals can be considered important species for monitoring long-term manifestations of OCPs in the marine environment and, for this reason, used as indicators of global pollution, because many of them migrate large distances and are unlikely to indicate local pollution (Tanabe and Subramanian, 2006; Tanabe, 2007).

Marine mammals have a long life cycle, and high concentrations of pesticides are found in their organs. However, levels of pollutants in females and males differ substantially. In general, OCP concentrations in immature males and females are comparable and increase until puberty. Subsequently, the concentration in males continues to increase, while in females it reaches a plateau or slightly decreases (Tanabe and Subramanian, 2006). A higher OCP concentration in female Dall's porpoises was recorded in the case of absence of ovulation (120000 ng/g lipids) when compared to animals with a normal reproductive cycle (3000 ng/g lipids) (Kajiwara et al., 2002). The lower OCP concentrations in female marine mammals (cetaceans and pinnipeds) are explained by the mother–fetus transfer of xenobiotics (transplacental transfer via the common circulatory system) during pregnancy and later in the lactation period (with mother's milk having a high lipid content) (Greig et al., 2007; Vanden Berghe et al., 2012).

In addition to the age-related tendency, dietary habits are also of great importance in the accumulation of OCPs by marine mammals. Fish eaters generally accumulate them to higher concentrations than animals that feed on plankton (Tsygankov et al., 2014a; Tanabe and Subramanian, 2006). Thus, in the finless porpoise, feeding mostly on crustaceans and cephalopods, the concentration of OCPs amounted to 48000 ng/g lipids (Park et al., 2010). In the killer whale, a predator characterized by a wide diet spectrum (fish, pinnipeds, etc.), the OCP concentration was 161300 ng/g lipids (Krahn et al., 2007). In the spotted seal, preying mainly on fish, the level of toxicants reached ca. 381400 ng/g lipids (Trukhin and Boyarova, 2013).

The extreme persistence and volatility of OCPs facilitated their spread all over the world's oceans, which has caused both regional and global pollution. They accumulate in various components of marine ecosystems. As a result of biomagnification, their highest concentrations are now recorded from members of the highest trophic levels (seabirds and mammals).

OCPs in Marine Organisms of the Far Eastern Seas (2000–2016)

Studies of organochlorine compounds in marine ecosystems of the Far Eastern seas of Russia have been fragmentary. Some data on OCP levels in the Sea of Japan are considered in the works of Tkalin et al. (1997, 2000). A more detailed monitoring of OCPs in the Sea of Japan is reported in the works of M.D. Boyarova and O.N. Lukyanova (Boyarova, 2008; Boyarova et al., 2004, 2006, 2012; Lukyanova et al., 2007, 2012; Lukyanova, 2013). Recent data on OCP concentrations in ecosystems of the Bering Sea and the Sea of Okhotsk are provided in our studies (Lukyanova et al., 2014, 2015, 2016; Tsygankov, 2012, 2016; Tsygankov et al., 2014a, 2014b, 2015, 2016a, 2016b, 2017, 2018).

For the Bering Sea, the accumulation, biotransformation, and transport of OCPs have recently been studied in Pacific salmon and marine mammals; for the Sea of Okhotsk, in seabirds and Pacific salmon (Table 1); for the Sea of Japan, in mollusks and fish (Table 2).

In the Sea of Japan, mollusks and fish were sampled in some parts of Peter the Great Bay: from Posyet, Amur, and Ussuri bays. Samples of mammals (gray whale and Pacific walrus) were collected from Mechigmen Bay, off the village of Lorino; Pacific salmon (sockeye *Oncorhynchus nerka* and chinook *O. tshawytscha*), were collected from the southwestern Bering Sea off the Commander Islands and eastern Kamchatka. Furthermore, Pacific salmon (pink *O. gorbuscha*, chum *O. keta*, sockeye *O. nerka*, and chinook *O. tshawytscha*) and seabirds (fulmar *Fulmarus glacialis*, crested auklet *Aethia cristatella*, auklet crumb *Aethia pusilla*, Pacific gull *Larus schistisagus*, and grey petrel *Oceanodroma furcata*) were sampled during expeditions of the Pacific Research Fisheries Center (TINRO-Center) in the Sea of Okhotsk off the western coast of the Kamchatka Peninsula, along the western part of the Kuril Islands in coastal waters, and off Hokkaido Island.

Mollusks and Fish from the Sea of Japan

The total OCP content of mussels from different parts of Peter the Great Bay, particularly from waters off the city of Vladivostok, has increased almost tenfold in recent years. According to Tkalin et al. (1997), in 1996, the total OCP content of soft tissues in Gray's mussels from Amur and Ussuri bays amounted to approximately 4.5 ng/g. According to our data, the total OCP content of mussels from Amur Bay in 2004 was 50.0 ng/g; from Ussuri Bay it was 20.0 ng/g.

In 1998, the concentration of OCPs in soft tissues of the mussels collected in the coastal waters off Reineke Island did not exceed 0.8 ng/g (Tkalin et al., 2000); in 2004, the level reached 25.8 ng/g (Boyarova, 2008). The increase in the total OCP content of the mussels both from the inner parts of Amur and Ussuri bays and from the water off Reineke Island, which is considered a conditionally background area, can indicate the continuing economic activity in the south of Primorskii krai, as well as the transfer with atmospheric and sea currents from areas where OCPs are still used (India and China). The time a compound takes to degrade in the environment, which may last for several decades, should also be taken into account. The differences in concentration can also be explained by different methodological approaches and instrumental measurements.

Table 1. Organochlorine pesticides (ΣHCHs and ΣDDTs) (ng/g lipids) in the lipid fraction of marine organisms from the Sea of Okhotsk and the Bering Sea

Species	Sampling area	Sampling period	ΣHCHs	ΣDDTs	References
Pink salmon (*Oncorhynchus gorbuscha*)	Northwest Pacific Ocean (open waters off the Kuril Islands); southwestern Bering Sea (off the Commander Islands and Eastern Kamchatka)	2013	9.2	132.3	Lukyanova et al., 2014, 2015, 2016; Tsygankov, 2016; Tsygankov et al., 2016b
Chum salmon (*O. keta*)			14.1	111.4	
Sockeye salmon (*O. nerka*)		2010–2011	465	833	
Chinook salmon (*O. tshawytscha*)			193.6	2984.3	
Northern fulmar (*Fulmarus glacialis*)	Southeastern Sea of Okhotsk (western coast of the Kamchatka Peninsula and the Kuril Islands)	2012	3335.3	1630.47	Tsygankov et al., 2016a; 2017, 2018
Crested auklet (*Aethia cristatella*)			7575.36	884.78	
Pacific gull (*Larus schistisagus*)			3576.41	674.66	
Grey petrel (*Oceanodroma furcata*)			2017.05	377.1	
Auklet crumb (*Aethia pusilla*)			1583.5	1220.97	
Pacific walrus (*Odobenus rosmarus divergens*)	Mechigmen Bay, Bering Sea (village of Lorino)	2011	4220	2070	Tsygankov et al., 2014a, 2014b, 2015
Gray whale (*Eschrichtius robustus*)		2010	14140	12160	Tsygankov, 2012; Tsygankov et al., 2014b, 2015, 2018

A high total OCP content (616 ng/g) with the absolute predominance of DDT and its metabolites (450 ng/g) was found in mussels from Posyet Bay, which may be associated with the influence of the Tumen River discharging large amounts of various contaminants into the sea. The OCP concentration in the mussels from Posyet Bay was significantly higher than in mussels from Amur and Ussuri bays. Thus, the transboundary transport of pollutants is of great significance in coastal waters of Primorskii krai (Boyarova and Lukyanova, 2006).

An analysis of the ratio of DDT and its metabolites to HCH isomers in bivalve organs has shown that, in the mussels from Posyet Bay exposed to the transboundary transport of pollutants, the DDT group is dominant, as in animals from other coastal zones of Asian countries. At the same time, the level of total HCH isomers in the mussels from Peter the Great Bay was much higher than that in mollusks from other countries. This may indicate the lower use of lindane in agriculture of Asian countries.

The values of maximum OCP concentrations in the mussels from Peter the Great Bay are comparatively low relative to the toxicity and LD_{50} values for humans, estimated at 250–400 mg/kg for DDT and its metabolites and 300–500 mg/kg for HCH (*Spravochnik khimika*, 1967). Thus, the current level of pesticide contamination of mollusks does not pose a threat to human health.

When OCPs were measured in muscles of fish caught in coastal marine waters of Primorskii krai, the saffron cod from in Posyet Bay showed the maximum total concentration (638 ng/g). In muscles of the yellowfin goby from the same bay, the amount of OCPs was also high and reached 264 ng/g. In muscles of the flathead grey mullet from Sivuchya Bay, the pesticide concentration was 88 ng/g. In all three species of fish from the coastal waters of southwestern Primorskii krai, total OCPs were dominated by DDT and its metabolites, like in mussels from Posyet Bay.

Several flounder species were also sampled from most of the study areas (Sivuchya Bay, Posyet Bay, Amur Bay, and Ussuri Bay). The maximum OCP concentration (150 ng/g) was recorded from muscles of the flounders caught in Amur Bay. In contrast to fish from southern Primorskii krai, in the flounders of Amur Bay the total HCH isomers were dominant, 106.9 ng/g, and the main γ-isomer accounted for 74%.

In all the fish liver samples from Peter the Great Bay, almost all the HCH isomers and DDT and its metabolites were detected. The maximum OCP concentrations were recorded from the liver of the Far Eastern smooth flounder caught in Amur Bay (379–736 ng/g).

Pacific herring is a commercially valuable target species. Determining toxicants in this fish is of particular interest with regards to its safety to human health. The total OCP content of meat of the Pacific herring

Table 2. Organochlorine pesticides (ΣHCHs and ΣDDTs) (ng/g w.w.) in the organs of marine organisms from the Sea of Japan (2001–2005)

Species	Organs	Area	ΣHCHs	ΣDDTs	References
Pacific herring (*Clupea pallasii*)	Muscles	Southwestern Peter the Great Bay	1.4	1.1	Boyarova et al., 2004; Boyarova and Lukyanova, 2012
	Liver		4.6	6	
Sculpin (*Myoxocephalus brandti*)	Muscles		0.6	1.5	
Northern black flounder (*Pseudopleuronectes obscurus*)	Muscles		0	6.1	
	Liver		148	12	
Yesso scallop (*Mizuhopecten yessoensis*)	Hepatopancreas		785.6	5.2	
	Gonads		265.9	15.6	
Starry flounder (*Platichthys stellatus*)	Liver	Sivuchya Cove, Peter the Great Bay	118.1	17.1	Boyarova, 2008; Boyarova et al., 2004
Northern black flounder *(P. obscurus)*	Liver		45.7	11.9	
Sculpin (*M. branti*)	Muscles		0.6	1.5	
Pacific herring (*C. pallasii*)	Liver		4.6	6	
Pacific redfin (*Tribolodon hakonensis*)	Liver		10	42.8	
Flathead grey mullet (*Mugil cephalus*)	Liver		96.4	38.8	
Yesso scallop (*M. yessoensis*)	Soft tissues	Posyet Bay	395	–	Boyarova and Lukyanova, 2006
Gray's mussel (*Crenomytilus grayanus*)	Soft tissues		170	446	
Saffron cod (*Eleginus gracilis*)	Muscles		8	630	
Yellowfin goby (*Acanthogobius flavimanus*)	Muscles		12	252	
Starry flounder (*P. stellatus*)	Muscles	Amur Bay	–	0.2	Boyarova et al., 2004
	Liver		49.0	6	
Northern black flounder (*P. obscurus*)	Liver		62.3	351.8	
Far Eastern smooth flounder (*Liopsetta pinnifasciata*)	Liver		41.0	30.9	
Gray's mussel (*C. grayanus*)	Soft tissues		30.3	16.2	
Far Eastern smooth flounder (*L. pinnifasciata*)	Liver		17.4	122.5	Vashchenko et al., 2005
Far Eastern smooth flounder (*L. pinnifasciata*)	Liver	Ussuri Bay	136	1242	Boyarova, 2008; Boyarova et al., 2004
Far Eastern smooth flounder (*L. pinnifasciata*)	Muscles		9.5	20	
Gray's mussel (*C. grayanus*)	Soft tissues		14.6	7.8	

caught in Peter the Great Bay was 2.5 ng/g wet weight (w.w.), which is two orders of magnitude lower than in herring from the Baltic Sea (Roots, 2004).

In 2008, OCP content was measured in organs of Pacific redfin (*Tribolodon brandtii*) from the estuaries of the Razdolnaya and Artemovka rivers. The total maximum OCP concentrations with an average of 1700 ng/g w.w. were recorded from liver of the redfin caught in the Razdolnaya River estuary; in the fish from the Artemovka River estuary, the total OCP averaged about 1000 ng/g. In fish meat, the total OCP level is much lower than in liver and does not exceed the standards of Sanitary Regulations and Norms (SanPiN), 200 mg/kg w.w. In muscles of the fish from the Razdolnaya River, it amounts to an average of 70 ng/g; in the fish from the Artemovka, 57 ng/g. The main pollutants are DDT and DDE.

When comparing the OCP content of organs between estuarine (redfin) and marine fish species (Far Eastern smooth flounder from Ussuri and Amur bays), it can be noted that estuarine fish accumulate OCP to a much greater extent than marine fish, which agrees with the known pattern of accumulation and sedimentation of OCPs at the marine/fresh water interface.

Results of the studies of OCP content in the biota of Peter the Great Bay show that the level of pollutants in bivalves is lower compared to that in the coastal zone of the Asia-Pacific Region countries; in fish, it fits into the range of these substances recorded from other parts of the World Ocean to date.

Pacific Salmon from the Sea of Okhotsk and the Bering Sea

The mean pesticide content (Table 1, total ΣHCH and SDDT) of the pink *O. gorbuscha*, chum *O. keta*, sockeye *O. nerka*, and chinook salmon *O. tshawytscha* was estimated at 141.5, 125.5, 1298, and 3177.9 ng/g lipids, respectively. A comparison of the total amount of OCPs in muscles and liver of all four fish species from the Sea of Okhotsk and the Bering Sea did not show any significant difference in mean values between pink and chum salmon, but in these species they were significantly lower ($p \leq 0.05$) than in chinook salmon, which, in turn, also had lower values than in sockeye salmon. The concentration increases in the following sequence: chum ≤ pink < chinook < sockeye. The ranges of total OCP concentration, e.g., in muscles were as follows: 78.8–174.1, 89.3–222.8, 265.0–2435.4, and 165.7–3020.1 ng/g lipid, respectively (Lukyanova et al., 2015, 2016; Tsygankov et al., 2016b, 2017).

A comparison of soft tissues of chum and pink salmon showed significantly ($p \leq 0.05$) larger amounts of pesticides in chum, which can be explained both by the greater weight and fat content of this species and by the longer marine life history of its individuals. Pink salmon has an annual life cycle and returns to spawn in the following year after the smolt downstream migration into the sea, whereas chum salmon can feed for 2 to 5 years. The mean body weight of the examined chum salmon was 1863 g; the mean weight of pink salmon, 1130 g. As fish grows and gains weight, various pollutants are accumulated in its organs along with the build-up of its lipid reserves. The studied pink and chum salmon were collected in waters off the southern Kuril Islands in early summer, by the completion of their spawning migration, when organs begin to spend lipids. Before spawning, salmon spend up to 80% of their reserved lipids; in this case, the specific concentration of toxicant per 1 g lipids increases and can reach values hazardous to the health of fish (Brett, 1995).

Samples of sockeye and chinook salmon were collected in the western Bering Sea and in the Sea of Okhotsk in autumn, October and November, when fish feed at sea, where they can stay for several years. At this time, the concentration of toxicants in their organs is continuously increasing, not only in the metabolically active liver, but also in muscles. Thus, 99% of all toxicants in chinook salmon are accumulated during the marine life history phase (Cullon et al., 2009).

The number of salmon migrating to the Russian coast varies from year to year, but the species structure of their runs keeps stable: pink salmon accounts for 60–65%; chum, 20–25%; sockeye, 10–12%; and coho and chinook salmon constitute a small fraction. Chum and pink salmon form the major part of runs in eastern Kamchatka, eastern Sakhalin, the mainland coast of the Sea of Okhotsk, and the Amur River basin (Shuntov and Temnykh, 2008). In the even-numbered year 2008, their total catch amounted to 258 000 t; in the record-breaking odd-numbered year 2009, 542 000 t; in 2010, 324 000 t. The number of fish that escaped to their spawning grounds in certain regions of the Far East in 2008–2010 is published in the annual reports of the North Pacific Anadromous Fish Commission (NPAFC). These fish remain at their grounds after spawning and serve as a food supply for many organisms, thus connecting the marine and terrestrial food chains, or, in other words, transferring organic matter from the ocean to the land.

The concentration of OCPs as an averaged sum of HCH+DDE for males and females was 68.85 ng/g w.w. in whole pink salmon and 182.5 ng/g w.w. in chum salmon. The mean weight of one individual of pink salmon is 1.3 kg; that of one spawning chum salmon is 3.5 kg. Calculations show that one pink salmon contains up to 90 μg pesticides and one chum contains up to 640 μg (Lukyanova et al., 2014). Thus, the total amount of OCPs transported, e.g., in 2009 to eastern Kamchatka by these two salmon species only constitutes 10.4 kg; to the Amur River basin, more than 13 kg; and to the mainland coast of the Sea of Okhotsk, 8.1 kg (Lukyanova et al., 2015).

In 2008, the influx of pesticides with salmon to various regions of the Pacific coast of Russia ranged from 520 g to 4 kg; in 2009, from 470 g to 13 kg; and in 2010, from 350 g to 7.75 kg. Within three years, the Amur basin received the largest amount of pesticides, approximately 23 kg. The high runs of salmon in recent years provided from 13 to 30 kg pesticides transferred to land annually.

Thus, organochlorines found in salmon from the northwestern Pacific Ocean indicate the global pesticide background that has formed both on the planet in general and in the world's oceans in particular. The accumulation of such persistent highly toxic compounds can affect the health of adult individuals, the success of their reproduction, and the survival of offspring. The sea-to-land biotransport of toxicants by salmon poses a threat of contamination to their spawning grounds. Further monitoring of pesticide bioaccumulation in salmon immediately at spawning grounds in our studies will provide new information on this little-known factor that can affect the total stock and catch of this commercially valuable group of fish.

Seabirds of the Sea of Okhotsk

The mean pesticide content of the northern fulmar *Fulmarus glacialis*, crested auklet *Aethia cristatella*, Pacific gull *Larus schistisagus*, grey petrel *Oceanodroma furcata*, and auklet crumb *Aethia pusilla* amounted to 5206, 8460, 4542, 2417, and 2804 ng/g lipids, respectively (Tsygankov et al., 2016a, 2017).

The studied bird species from the Sea of Okhotsk have different sizes, and, consequently, differ in the amount of subcutaneous fat. Since OCPs are known to concentrate mainly in the subcutaneous fat, bigger birds show higher OCP content values (Table 1). Pacific gull (with its mean body length of 64 cm) and fulmar (47 cm) have similar concentrations of OCPs in plumage with skin (5962 and 5949 ng/g lipids, respectively). In grey petrel, which is much smaller (22 cm) than the bird species above, the pesticide content was 3128 ng/g lipids. The different levels of trophic relationships between these species also result in different degrees of accumulation. The mean pesticide content of internal organs in birds indicates their different diet spectra. The food of fulmar includes organisms characterized by a high coefficient of OCP accumulation (fish, fish eggs, mollusks, crustaceans and other invertebrates, carrion, whale entrails, various fatty waste, etc.), which explains the high pesticide content of its internal organs (5874 ng/g). Crested auklet, auklet crumb, and grey petrel feed on small-sized crustaceans, marine invertebrates, amphipods, etc., accumulating pesticides to a lesser degree than the items of the fulmar diet. Consequently, the OCP content of the internal organs in these birds is lower: 1730, 2804, and 1705 ng/g, respectively. The accumulation and distribution of pesticides in the organs of birds can be also controlled by other factors. For example, in a crested auklet having a body size of 26 cm, the OCP content of plumage with skin amounted to 15604 ng/g. Further research is required to identify the causes of the high OCP concentration in this species (Tsygankov et al., 2016a).

The accumulation of pesticides in birds affects various aspects of their physiology, causing serious deterioration of, e.g., reproductive function and thinning of egg shells, which leads to disturbance of embryonic development and loss of offspring. The significant OCP concentrations detected in seabirds from the Sea of Okhotsk, remote from regions with intensive agricultural activities, are a manifestation of the general global POP background being currently formed on the planet.

Marine Mammals of the Bering Sea

In the organs of the gray whale (*Eschrichtius robustus*), the lipid content ranged from 1 to 4% in muscles and up to 8% in the liver. In the organs of Pacific walrus (*Odobenus rosmarus divergens*), the lipid content ranged from 1 to 6% in muscles and up to 10% in the liver. Calculations showed (Table 1) that total OCPs in the fat of the gray whale averaged at 6290 ng/g; in Pacific walrus, 26300 ng/g (Tsygankov et al., 2014a, 2014b, 2015, 2017).

The different levels of pesticide accumulation in the gray whale and Pacific walrus from the Bering Sea indicate mainly the different degrees of contamination of their habitats by these pollutants. The species-specific patterns of accumulation of lipophilic xenobiotics are determined largely by the total fat content of the subcutaneous tissue and certain organs. Of great importance is also the degree of sexual maturity of individuals. The species studied by us occupy similar habitats; the fat content of their organs does not vary significantly between them, ranging within 8–10%, and the substantial differences in pesticide level may be associated with the stage of reproductive cycle or feeding habits. The food of gray whales mainly includes bottom crustaceans and other small benthic organisms inhabiting the surface and deeper layers of soft sediments (infauna). The main part of the walrus diet is comprised of benthic invertebrates: bivalves, some shrimp species, lobsters, polychaete worms, priapulids, octopuses, and holothurians, as well as some fish species. Furthermore, walruses can sometimes prey on seals: there are documented cases of their attacks on ringed seals and harp seal pups (Burdin and Filatova, 2009). The prey species of walrus accumulate more pesticides in their body than the items of the gray whale diet, since the coefficients of accumulation of pollutants for mollusks and fishes are higher than those for crustaceans. Based on this fact, we may conclude that the dietary factor is responsible for the differences in bioaccumulation of pesticides between whales and walruses (Tsygankov et al., 2015, 2017).

A comparison of these results with those for other marine mammals shows that marine mammals from the Bering Sea have lower OCP concentrations than mammals from other parts of the World Ocean. This again confirms that the level of OCP pollution of the Bering Sea can be considered background.

CONCLUSIONS

This work is a stage of the research on OCP accumulation by members of various links of the food chain such as Pacific salmon, seabirds, and marine mammals in the subarctic region of seas of the Russian Far East. OCPs are accumulated most actively in marine mammals and birds that hold the top position in the trophic pyramid. Organic pollutants appear to be deeply involved in the directed transport of nutrients by salmon, which connects the oceanic and terrestrial ecosystems. The annual influx of pesticides into spawning grounds and the continuous increase in their concentration in some local zones poses a probable ecological risk to certain populations whose spawning success can be reduced due to the toxicity of their habitats. Marine mollusks, fish, mammals, and birds are bioindicators of OCPs on a global and long-term scale. These organisms make it possible to study the integrated temporal trends in the distribution of pesticides over the marine environment, particularly in the oceans. Currently, the levels of POPs in organisms from the seas in the Far East are lower than the limit values established for seafood by the technical regulations of the Customs Union.

The OCP concentrations in the Sea of Okhotsk and the Bering Sea can be considered background, whereas the Sea of Japan is exposed to contamination from the countries using these substances in agriculture.

The prospects for further research in seas of the Far East are as follows:

- extend the range of studied POPs in the course of nontargeted screening analysis of “new” xenobiotics;

- study the patterns of toxicant distribution along food chains using stable carbon and nitrogen isotopes;
- investigate the accumulation and transformation of POPs in residents of the coastal regions of the Far East Federal District continuously consuming seafoods from local waters.

FUNDING

This study was financially supported by the Russian Science Foundation (agreement no. 18-14-00120).

COMPLIANCE WITH ETHICAL STANDARDS

Conflict of interests. The authors declare that they have no conflict of interest.

Statement on the welfare of animals. All applicable international, national, and/or institutional guidelines for the care and use of animals were followed.

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Translated by E. Shvetsov