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Spatial Distribution and Ecological Risk Assessment of Residual Organochlorine Pesticides (OCPs) in South American Marine Environments

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Abstract

Purpose of Review Organochlorine pesticides (OCPs) have been intensely used and produced in South America. Although they were banned before the year 2000 (excepting endosulfan, which has been recently banned in several countries), OCPs remain detectable in marine environments of this continent, sometimes at risky levels for biota. This manuscript summarizes studies on OCP levels in the air, water, sediment, bivalves, fish, and marine mammals of the South American coasts and open waters over the last 20 years, tackling their spatial distribution and analyzing their associated ecotoxicological risk.

Recent Findings To the best of our knowledge, this is the first study integrating all available information on current levels of OCPs in South American marine environments. The 63 researches reviewed studied punctual sites or environmental compartments.

Summary The OCP levels were higher in semi-enclosed environments such as bays and estuaries, close to large cities. In terms of individual OCPs, dichlorodiphenyltrichloroethane (DDT) and its degradation products were more abundant than other OCPs in all the environmental compartments, excepting air, for which the most abundant OCP was endosulfan. Depending on the location and the environmental matrix, hexachlorocyclohexanes (HCHs), heptachlors, mirex, and endosulfans followed DDTs. Aldrin, dieldrin and endrin, hexachlorobenzene (HCB), and chlordanes were found at very low levels in most matrices and sites, with some exceptions. Considering their potential ecotoxicological risk, most sites would be safe for biota; however, the levels of some OCPs could damage the structure and function of the communities of several coastal sites in a short or long term, mainly in southeastern Brazil and on the coast of the Argentine Pampas. Moreover, it remains to evaluate many sites potentially contaminated by OCPs.

Keywords Persistent organic pollutants · Organochlorine pesticides · South America · Marine environments · Ecotoxicological risk

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Introduction

Organochlorine pesticides (OCPs) are compounds used mostly for phytosanitary purposes and the control of biological vectors; they are considered persistent organic pollutants (POPs) due to their high toxicity, fat solubility, and degradation resistance [1••]. Many OCPs are semivolatile with relatively high Henry's law constants; therefore, they can migrate to the atmosphere and be transported over long distances in a gaseous state before dry or wet deposition occurs in the oceans and continents [2–5]. Moreover, OCPs are usually adsorbed in sediment and soil particles, which can be a secondary source of pollution of marine environments due to their weathering and transport through surface runoff from the continents to the sea [6]. These processes probably cause a large amount of the

sprayed OCPs that are deposited in marine environments, turning them to the ultimate sink of these compounds [3, 7].

When OCPs enter the ocean, they tend to accumulate in biota and bottom sediments [8]. Smaller organisms incorporate them primarily through their respiratory surfaces whereas larger animals do so through food intake [8]. Due to their fat solubility and high persistence to biological degradation, they are biomagnified along the trophic web [8–10] so that the highest trophic levels, such as marine mammals, top predatory fish, and seabirds, usually show the highest OCP concentrations. Humans and biota can be affected by the toxic effects caused by short-term or long-term exposure to these pesticides. These toxic effects can produce reproductive damage [11], endocrine disruption, immune suppression, or cancer, among others [12, 13].

In South America, most of the OCPs were intensively used and produced between 1950 and 1990, except for endosulfan, dicofol, and methoxychlor, which remained in use until recent years. While they were in use, a large amount of OCPs entered the continental environment and the sea. Consequently, although all South American countries have banned or restricted their use and production, detectable and even harmful environmental concentrations are likely to remain present in seas and oceans for decades because of the intrinsic characteristics and environmental dynamics of OCPs [3, 6, 14].

The harmful effects of OCPs on the environment are particularly of great concern in sensitive environments with a complex interaction of ecosystems and enormous biodiversity, such as mangroves, salt marshes, and coral reefs [15]. Along the 31,080 km of the South American coast, there are hundreds of places with these characteristics, out of which 45 are wetlands of international importance according to the Ramsar Convention (https://rsis.ramsar.org/).

Due to the economic, social, and ecological importance of South American marine environments, the contamination by POPs, including OCPs, have received great attention in recent years [8, 14]. Consequently, OCP levels have been evaluated in several marine compartments, such as the air, sediments, water, mussels, birds, crabs, fish, and marine mammals. However, there is no exhaustive analysis of the current spatial distribution of OCP levels in these South American environments and their associated ecotoxicological effects, that is, the possible damage to the associated ecological communities.

Therefore, the aims of this review are the following: to summarize the results of the studies on OCP levels in the main environmental compartments of the South American coasts and seas including seawater, the air, sediments, and biota (bivalves, fish, and marine mammals) over the last 20 years; to evaluate the spatial distribution of residual OCP levels in South American marine environments; and to analyze the ecotoxicological risk associated with residual concentrations of OCPs in each evaluated site.

Use, Production, and Legislation

All South American countries have national legislations to restrict or ban the use and production of OCPs, which are normally more restrictive and prior to international conventions (Appendix A, Supplementary material). Moreover, all countries signed and ratified the Stockholm Convention on POPs to eliminate the use and production of the POPs listed there, including many OCPs, and the Rotterdam Convention to eliminate the use of toxic chemicals (Appendix A, Supplementary material).

In South America, OCPs were used, produced, and formulated from 1950 to 1990, with the exception of some pesticides such as endosulfan or methoxychlor that were used until recently. The exact amount of pesticides used, produced, or imported by South American countries before their ban and the current available stocks are unknown. The inventories of the National Implementation Plans (NIP) of the Stockholm Convention and the UNEP regional reports [8, 14] show an approximation of the volumes used and stored in some South American countries.

Endosulfan and DDT were the most commonly used pesticides in South America. Both were intensively used for phytosanitary control of a wide variety of crops [14], and DDT was also used in antimalarial programs in tropical and subtropical areas since 1960 [8, 14]. These pesticides were produced in Argentina [16] and Brazil. In Brazil, 75,500 tons were produced between 1959 and 1982 [17]. Brazil also produced 48,150 tons of endosulfan between 2008 and 2010 [17]. Available data on endosulfan imports show its intensive use; for example, Colombia imported 173 tons in 1999 [14], Brazil imported 26,700 tons between 1997 and 2011 [17], and Argentina imported 4815 tons between 2008 and 2010 [18].

The use of DDT and endosulfan has decreased substantially as a result of their ban, and although DDT is still allowed for public health applications by the Stockholm Convention and local laws from some countries such as Guyana, Suriname, Venezuela, and Brazil [1, 8, 14], there is no reported use of DDT for disease vector control in South America [19]. However, since endosulfan has been recently banned in most countries (Appendix A, Supplementary material) and DDT is extremely persistent, the environmental concentrations of these pesticides could be high due to their past intensive usage for disease vector control [8, 14] and their present illegal use [14]. Moreover, dicofol, which is a toxic OCP that usually contains DDT as impurity [20], has been applied until recent years in many countries.

Drins (i.e., aldrin, dieldrin, and endrin) were the first OCPs introduced in South America in the 1950s; they were used especially in the phytosanitary control of cotton, corn, sugar cane, and citrus products, among others, as well as in the extermination of termites. For example, Brazil imported 17,300 tons of aldrin between 1961 and 1996 [17]; the Shell

Company produced drins in Brazil and Argentina, although the quantities remain unknown [14]; and 1275 tons of drins were produced in Argentina in 1967 [16]. Heptachlor was used to control soil insects and termites; Brazil imported about 6400 tons of this pesticide between 1961 and 2003 [17], using it mainly in sugarcane plantations [14]. A little more intense was the use of hexachlorocyclohexanes (HCHs); for example, Brazil produced 18,400 tons and imported 8064 tons of lindane between 1955 and 2006 [17]. These pesticides were used to control soil insects and ectoparasites in animals, for seed treatment, in household insecticides, and as preservatives for clothing and wood [14].

Less commonly used OCPs include chlordane, HCB, and mirex. Chlordane was only used in specific cases such as ant control [14]; the few available data show that 220 tons were formulated in Argentina in 1967 [14], and 58.5 tons were imported by Venezuela between 1975 and 2003 [21]. On the other hand, HCB was almost unused as a fungicide in the region. However, its generation as a by-product during the manufacturing of dyes, organic solvents, and rubber could be the main source of HCB pollution of the environment [14]. Mirex (or dodecachlor or dechlorane) was mainly used to control ants and as a flame retardant in plastics, paints, and papers. The little information available indicates that it was rarely used: 314 tons were imported by Brazil between 1989 and 1998 [17], and 25 tons were imported by Ecuador between 1997 and 1998 [22].

Methodology

Literature Review and Data Management

This paper reviews scientific articles on OCPs in South American marine environments published between 1998 and 2019. A search was conducted in the most common search engines, official state websites, and government reports using different combinations of words in English, Portuguese, and Spanish (Fig. 1). Since many of these studies express pesticide levels as the sum of the congeners of each pesticide, the spatial distribution analysis of OCPs was performed with these summations: DDTs (as the sum of o,p′-DDE, p,p′-DDE, o,p′-DDD, p,p′-DDD, o,p′-DDT, and p,p′-DDT); HCHs (α-HCH, β-HCH, γ-HCH, and δ-HCH); endosulfans (α-endosulfan, β-endosulfan, and endosulfan sulfate); heptachlors (heptachlor and heptachlor epoxide); and chlordanes (trans-chlordane, cis-chlordane, trans-nonachlor, and cis-nonachlor). In general, when the articles showed the data of each congener, they also showed the sum data of each group of compounds. However, when the sum was not supplied, it was calculated from the sum of the interest items. The levels below the detection limit were taken as zero, since in general, the detection limit was close to zero, and in many research papers, this value was not established. In other articles, only average levels were shown for each matrix or study area, and that value was used for each site.

When necessary, the levels of each congener were used separately; for example, to assess the DDT/DDE ratio (i.e., the age of the DDT input into the environment) or to assess the ecotoxicological risk associated with lindane levels (γ-HCH) in sediments. The levels of pesticides that refer to individual samples, and not averages per site, were used to analyze the distribution and dynamics of pesticides in marine mammals, because the number of individuals in each site used to be low, and the average levels could not be representative of the place.

The data analysis of OCP levels was performed using the ArcGIS 10.3 (Esri, 380 New York Street, CA 92373, USA) and Microsoft Excel (ver. 2013, Microsoft, Redmond, WA, USA). Furthermore, these levels were compared with the Norwegian Environmental Quality Classification System (NEQCS) [23, 24] to determine the ecotoxicological risk associated with OCP levels in different environmental matrices (Appendix B. Supplementary data).

Results

Distribution of OCP Levels in South American Marine Environments

About eighty research or monitoring studies that involved the determination of OCP levels in several environmental compartments of South American marine areas were published over the last 20 years. However, some documents do not report data on OCP levels and just limit to the study of a single group of pesticides or studied OCP levels in environmental compartments that are not relevant to this work (for instance, pellets, suspended particulate matter, penguins, and crabs). Consequently, 63 studies were reviewed [7, 9, 12, 25–83], most of which analyze diverse environmental compartments (Fig. 2). Figure 2 shows that most studies were performed close to important cities such as Rio de Janeiro, Sao Paulo, Curitiba, and Florianopolis in the southeast of Brazil and Buenos Aires, Mar del Plata, and Bahía Blanca in Buenos Aires Province in Argentina. Moreover, most studies were conducted in coastal landforms with great ecological importance, such as estuaries, fjords, bays, and coastal lagoons. The summary of the data from each reviewed study is available in tables S1, S2, S3, S4, S5 and S6 of appendix C of the Supplementary material.

Seawater

Only four studies on OCP levels in the South American seawater were found [7, 25, 26, 27•]. Along the Atlantic Ocean,

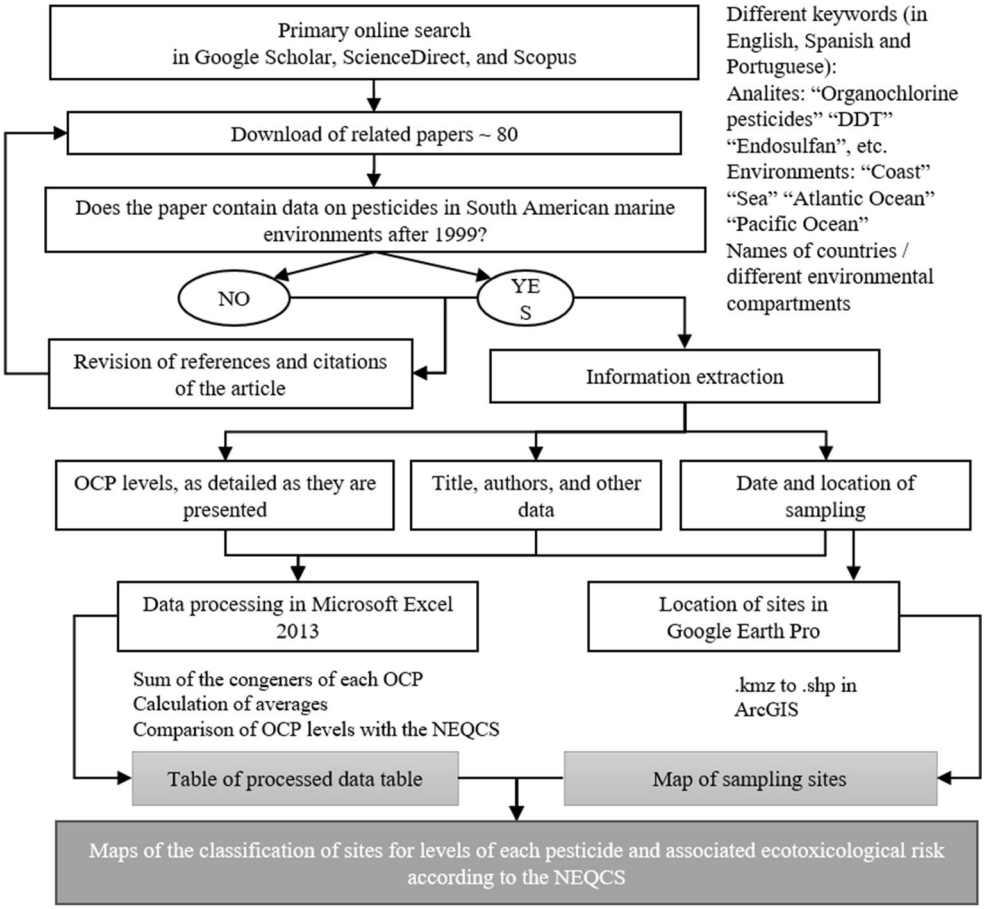

Fig. 1 Flow chart. Identification and inclusion of papers and data management in this review

two oceanographic cruises have studied these contaminants. Their study areas were a NE-SW transect between Gothenburg (Sweden) and McMurdo station (Antarctica), located approximately 250 km east of the South American coast [27•] and a NW-SE transect between Rhode Island (USA) and Namibia, located approximately 500 km northeast of the South American coast [7]. In the first one, the α-HCH, lindane, α-endosulfan, and HCB levels were evaluated whereas in the second one, o,p′-DDT, α-HCH, lindane, and HCB levels were assessed. In both studies, the concentrations found for all the compounds were very low. Near northeastern South America, the only compound found above the detection limit (DL) was HCB, whose maximum value (0.56 pg L^{-1}) was recorded near Martinique Island. In the NE-SW transect, the only compound lower than <DL was HCB. In addition, α-endosulfan and lindane were detected in 50% and 75% of the samples, respectively. The maximum values of both compounds were found in front of the Río de la Plata Estuary: 5.5 pg L^{-1} for α-endosulfan and 13 pg L^{-1} for lindane. On the other hand, α-HCH levels gradually increased from north to south, with values <DL near Natal, Brazil, to 5.3 pg L^{-1} near Tierra del Fuego, Argentina. According to the NEQCS classification, the OCP levels found in these studies would not harm the ecological communities of the evaluated sites.

Two studies analyzed the OCP levels in the Colombian coast waters: INVEMAR (José Benito Vives de Andréis Marine and Coastal Research Institute), 2010 [25], and Menzies et al., 2013 [26]. The first one analyzed OCPs in unfiltered water; although the detection limits were high, DDT and endosulfan levels were detected in 25% and 15% of the samples, respectively. According to the NEQCS, these levels could cause damage to biota in several Colombian coastal sites, including Cienaga de la Virgen, the front of the Agrotijo River, and the mouth of the Guadalito River. On the other hand, Menzies et al. analyzed DDT, drin, HCH, and chlordane levels in sea surface water. The detection percentage was 95% at the selected sites, being DDTs and HCHs the most abundant compounds. Rosario Islands, Santa Marta Bay, and Cienaga were the most polluted sites, with 22.2 ng L^{-1}, 67.9 ng L^{-1}, and 62 ng L^{-1} of DDTs and 52.7 ng L^{-1}, 12.2 ng L^{-1}, and 20.3 ng L^{-1} of HCHs, respectively. Unfortunately, the OCP levels found in this work could not be compared with the NEQCS ones because the sampling methodology was not compatible with the one required by it.

Air

The spatial coverage of air pesticide analyses of South American marine environments was quite broad (Figs. 2 and

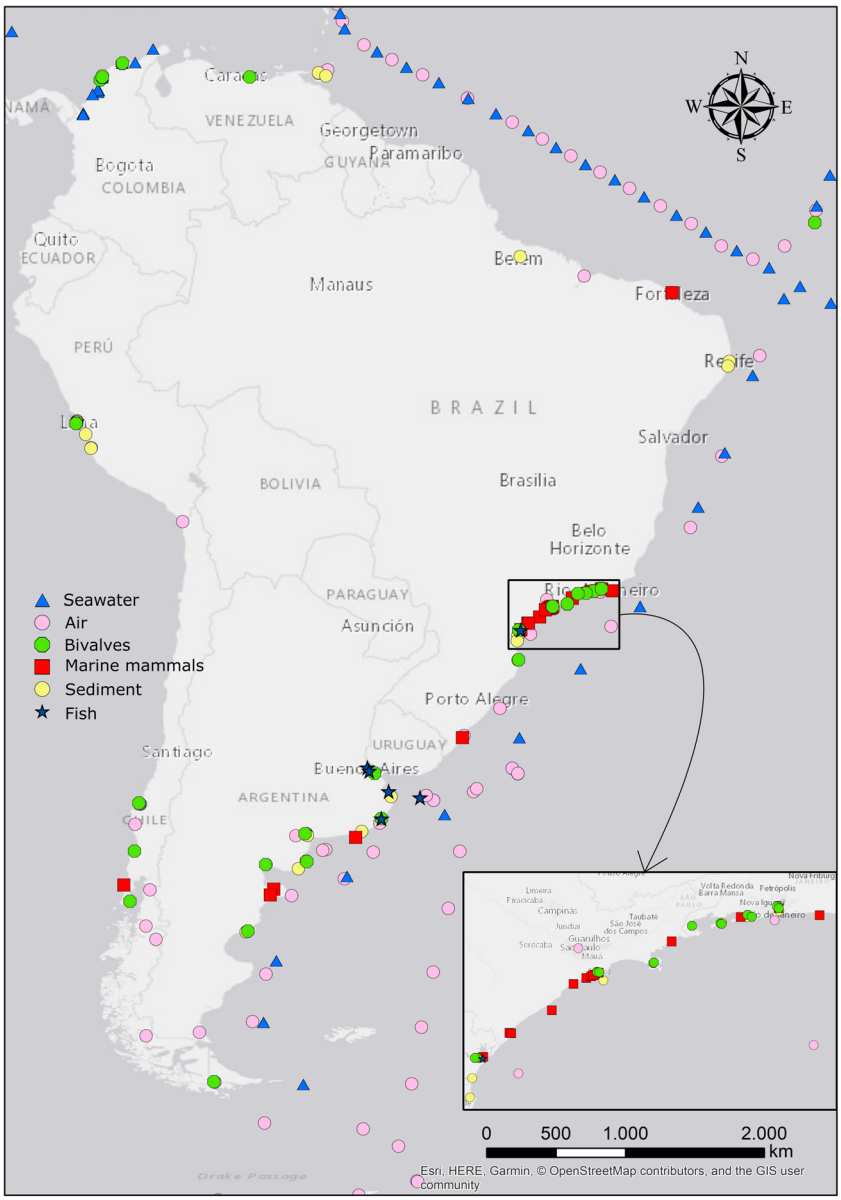

Fig. 2 Geographic location of data published over the last 20 years on organochlorine pesticides in seawater, the air, sediment, fish, bivalves, and marine mammals in South American marine areas

3). In the present work, 13 studies were selected and analyzed, out of which 8 were conducted in coastal environments [31–38] and 5 in the open sea [7, 27•, 28, 30•]. Figure 3 shows the distribution of DDT, HCH, endosulfan, and HCB levels, which were the most frequently analyzed pesticides. Other OCPs such as chlordanes, drins, and heptachlors were studied in less than 37% of the sites, so they were not included in the figure. These pesticides showed low overall levels in relation to the other OCPs.

Generally, OCP levels were higher near urban and coastal areas. The Brazilian coasts showed the highest DDT and HCH levels in South American coast, with a mean of 38.4 pg $m3^{-1}$ and 21.5 pg $m3^{-1}$, respectively. The Argentinean coasts showed the highest endosulfan levels, 1185 pg $m3^{-1}$ on average and a maximum of 4699 pg $m3^{-1}$ in Bahía Blanca. The Chilean coasts showed the highest average HCB levels, 38.8 pg $m3^{-1}$, and chlordane levels, 22.8 pg $m3^{-1}$; both compounds are related to urban use and production. The highest DDT levels in South America were found in the Brazilian coasts, 127.7 pg $m3^{-1}$ in Sao Luis, NE of Brazil [35•], and 102.6 pg $m3^{-1}$ in the Rio de Janeiro coast, SE of Brazil [28]. The highest endosulfan levels in the air from the South American coasts were found in Bahía Blanca, Argentina, with an average of 4699 pg $m3^{-1}$ and a maximum of 18,739 pg $m3^{-1}$ in the summer of 2005 [33].

The OCP levels in the air over the open sea were, in general, lower than those found on the coast. Furthermore, when comparing the data available for northern Brazil, eastern Brazil, eastern Uruguay, Buenos Aires Province, and eastern Argentinean Patagonia, it was observed that the highest

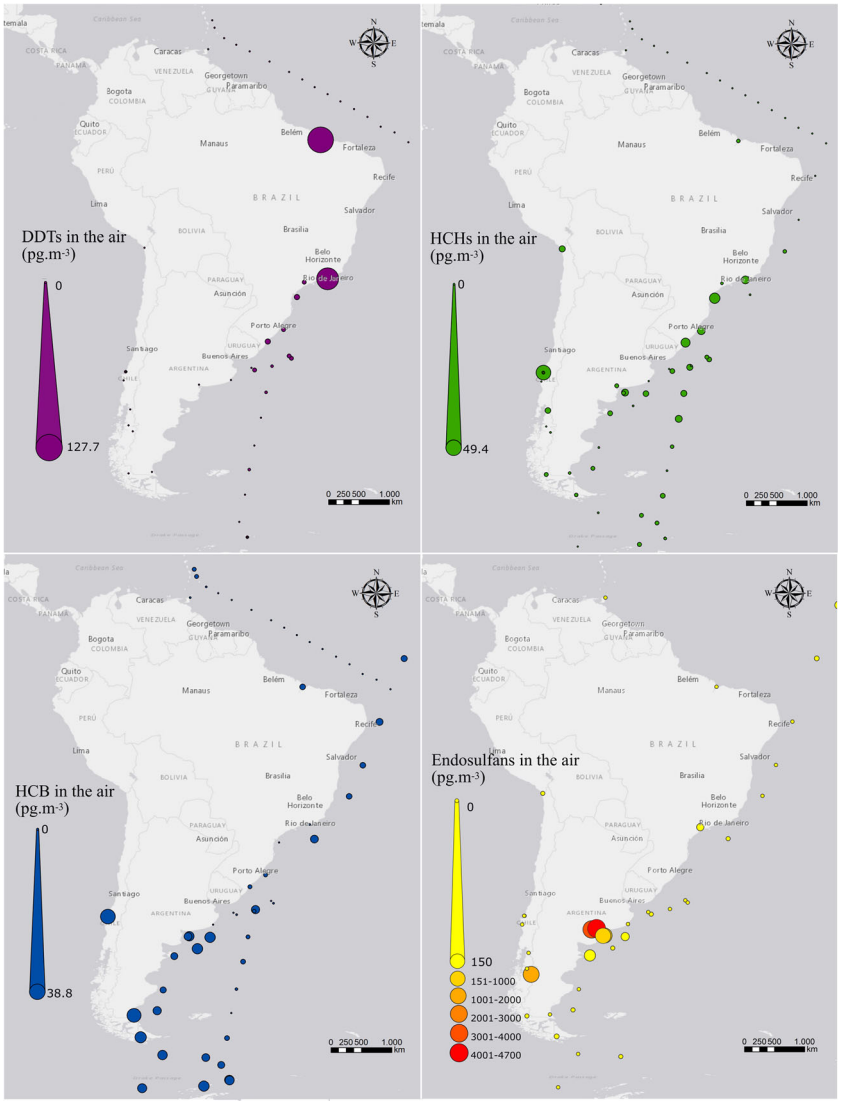

Fig. 3 Spatial distribution of DDT, HCH, endosulfan, and HCB levels in the air of South American marine areas

concentrations of all the compounds were found in the east of Buenos Aires Province and Uruguay, an extensive region with intense agricultural activity, excepting HCB, which was found in similar concentrations throughout the southern Atlantic Ocean, with average 11.61 pg m3^{-1} and SD 8.11 (Appendix C, Supplementary material and Fig. 3).

Sediments

Sediments were used as an environmental monitor in 20 of the reviewed articles [39–58]. In terms of individual OCPs, heptachlor, drin, and chlordane levels were generally <DL or lower, with a mean of 0.01, 0.01, and 0.02 ng g^{-1} dry weight (dw), respectively. However, some places showed relatively high levels for the three groups of pesticides. For example, heptachlor, drin, and chlordane levels were 3.61, 4.52, and 0.15 ng g^{-1} dw, respectively, in Guanabara Bay [51–54], and 3.74, 0.62, and 1.19 ng g^{-1} dw, respectively, in the Bahía Blanca Estuary [39–45, 46•, 47, 48].

On the one hand, DDTs were widely studied in this matrix, and their levels were considerably higher than the ones of other OCPs. The highest DDT levels were found on the Brazilian coasts (mean 7.22 ng g^{-1} dw, SD 15.51); Guanabara Bay was the most polluted site [51], followed by Babitonga Bay [52] and Guajara Bay in the Amazon Estuary [50•], with a mean of 53.9 ng g^{-1} dw, 7.14 ng g^{-1} dw, and 5.93 ng g^{-1} dw, respectively. Sediments from the Peruvian coast also presented high DDT levels, with a general mean of 5.23 ng g^{-1} dw and a maximum of 16.89 ng g^{-1} dw in the Chillón River front, near Lima [53]. However, the reported levels corresponded to sediments sampled in 1998, a few years after DDT had been banned. On the other hand, sediments from Argentina showed low DDT levels, with an average of 0.7 ng g^{-1} dw and a maximum value of 2.55 ng g^{-1} dw in the Bahía Blanca Estuary, south of the Pampas [57].

According to the NEQCS, DDT levels in most marine sediments in South America could not generate toxic effects on biota, except in Guanabara Bay in Rio de Janeiro, Brazil [51], and in the Chillón River front, near Lima, Peru [53], where it could cause damage to the structure and function of the community in the long term. However, the DDT/DDE ratio at both sites was less than 1; that is, the input would be historical. In other sites, this proportion was higher than 1 (recent input), but the levels represented a low ecotoxicological risk there.

In addition, HCH levels in sediments were lower than DDT ones. The general average of HCH levels in Argentina was 0.66 ng g^{-1} dw (SD 0.85) whereas in Brazil it was 0.37 ng g^{-1} dw (SD 0.59). The levels were highly variable in nearby sites; the highest levels were found in the Amazon River Estuary in Brazil and on the coasts of the Pampas in Argentina, an area of intense agricultural activity. As indicated above, the NEQCS sets environmental guidance levels for lindane in sediment, not for HCHs, so this congener was used to assess its possible effects on the biota of each site. Nonetheless, lindane levels are not shown in all the studies; consequently, the associated ecological risk could not be included in some sites analyzed for HCHs. It was observed that the lindane levels of several sites could cause damage to the structure and function of the ecological community in the short term (Fig. 4) in Mar Chiquita coastal lagoon in the Pampas, Argentina [49], and in Guanabara Bay in Rio de Janeiro state, Brazil [54]. Moreover, to the southwest of Buenos Aires Province [39, 48, 49, 57] and several sites in the southeast of Brazil, such as the Santos Estuary [40] and Ilha Grande [51], lindane levels in marine sediments could generate damage to biota in the long term.

Besides, endosulfan levels in sediment in the South American coast matched the levels found in the air and other environmental compartments (Fig. 3). The highest endosulfan levels in sediments were found in the Bahía Blanca Estuary [39, 48], Guanabara Bay [54], and the Rio Negro Estuary [47], where they could harm ecological communities in short periods. Moreover, near the Pampas in Argentina, there are several sites where sediment endosulfan levels could damage marine ecological communities in the long term (Fig. 4).

Finally, the HCB levels in the coastal sediment of South America were low, which would not pose a risk to biota in any of the evaluated sites.

Fish

Eleven studies on fish were found in the literature [41, 53, 59–67]. *Micropogonias furnieri* and *Mugil* spp. were the most common analyzed species, and muscle was the most used tissue. Dias et al. [61] reported OCP levels only in fish liver whereas Ferreira et al. [62] reported OCP levels in lipid-based muscle without indicating the lipid percentage; thus, they could not be compared with the data from other scientific research or with the NEQCS values.

In brief, OCP levels in fish from some South American coastal sites pose a concern. For example, the DDT levels in fish from the inner Rio de la Plata Estuary, Argentina (304 ng g^{-1} wet weight (ww)) [60], Paranaguá Bay, Brazil (15.2 ng g^{-1} ww) [64], and the Cañete coast, Peru (10.15 ng g^{-1} ww) [41] were high and thus, according to the NEQCS values, were related to a high ecotoxicological risk. The remaining sites showed moderate to low ecotoxicological risk for DDT levels, with values of less than 10 ng g^{-1} ww. Fortunately, DDT/DDE ratios were less than 1 in all the sites, which indicates historical inputs. The only exception was the fish (*Menticirrhus elongates*) collected in the Cañete coast, Peru, where the DDT/DDE ratio was greater than 7.6 and the p,p′-DDT level was 6.14 ng g^{-1} ww [41].

On the other hand, the ecotoxicological risk associated with the HCH levels in fish was low in all the sites excepting the inner Rio de la Plata Estuary, where the risk level in sabalo fish (*Prochilodus lineatus*) was high (Class IV) because the HCHs level was 8.8 ng g^{-1} ww [60]. The other OCPs were rarely analyzed, and the concentrations found were, in almost all cases, very low (less than 1 ng g^{-1} ww), except for some OCPs in the Río de la Plata Estuary, where Aizpun et al. [59] found 8.75 ng g ww of endosulfan and 8.55 ng g ww of chlordanes in *Leporinus* spp. and *Prochilodus lineatus*, and Colombo et al. [60] found worrisome chlordane levels in sabalo fish (248 ng g^{-1} ww). Unfortunately, there are no international guideline values for these OCPs in wild fish.

Bivalves

Ten studies have used bivalves as biomonitors of OCPs in South America [45, 51, 56, 64, 68–73]. The most commonly used bivalves were mussels, Fam. Mytilidae, and oysters, Fam. Ostreidae. According to the revised data, the Callao coast in Peru presented the highest ecotoxicological risk of the South American coasts (Classes IV and III of the NEQCS, depending on the species) due to the high DDT levels in bivalves: 5.14 ng g^{-1} ww, 6.18 ng g^{-1} ww, and 10.61 ng g^{-1} ww in *Semimytilus algosus*, *Argopecten purpuratus*, and *Aulacomya ater*, respectively [73]. Paranaguá Bay, Brazil [64, 71], and Ushuaia Bay, Argentina [69], showed moderate ecotoxicological risk (Class III) for DDTs, with values ranging between 5 and 10 ng g^{-1} ww in bivalves. The remaining studied sites presented low ecotoxicological risk associated with DDTs. In all the sites, the DDT/DDE ratio indicates historical input except in Guanabara Bay, Ilha Grande Bay, and Sepetiba Bay in the Rio de Janeiro state, Brazil [51, 70], and San Vicente, Chile [72], where the input would be recent.

The ecotoxicological risk associated with HCHs in bivalves was low or nonexistent in all the sites excepting Paranaguá Bay, Brazil, where the risk was moderate with mean HCH levels in oysters higher than 3 ng g^{-1} ww [71].

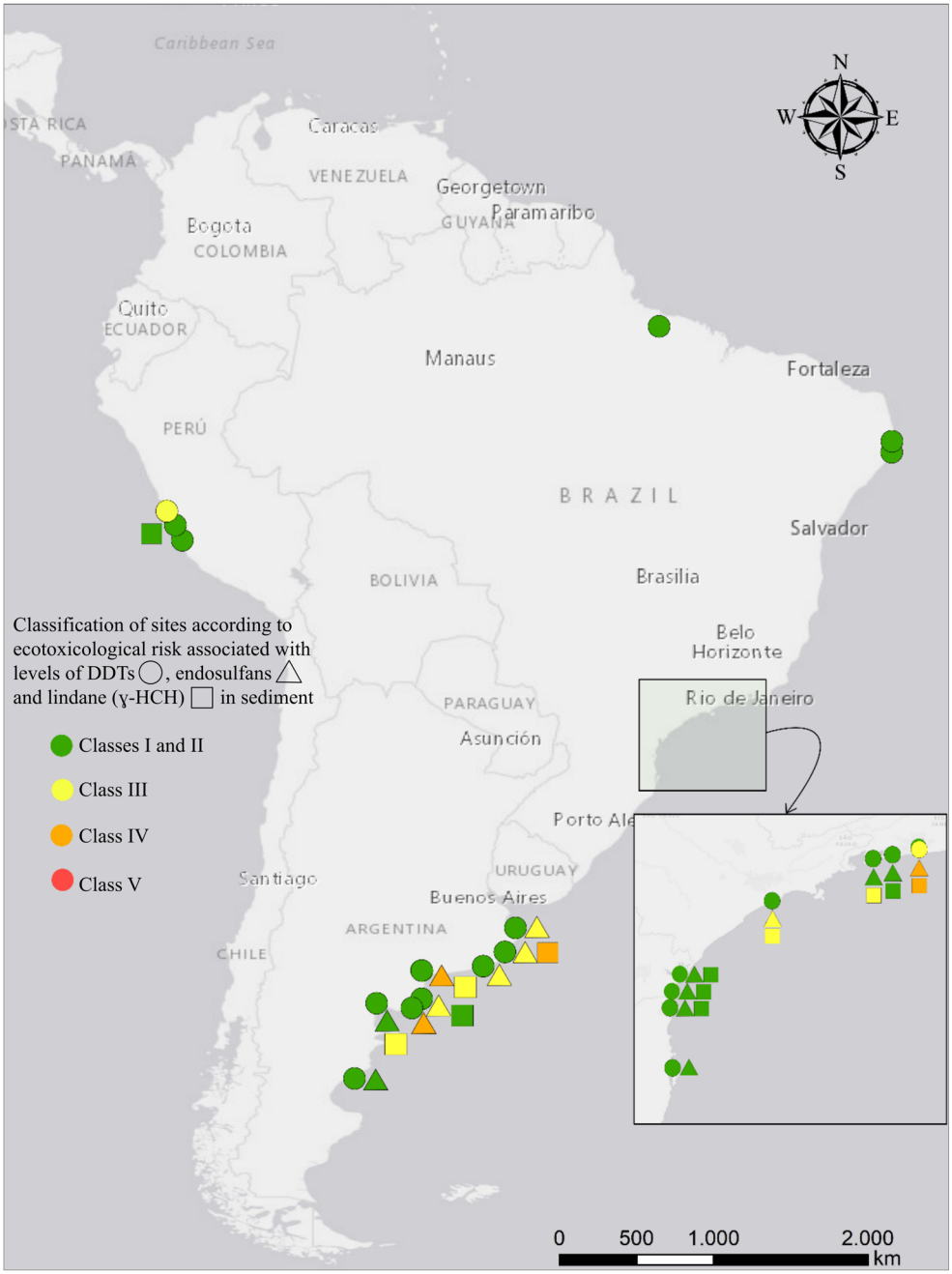

Fig. 4 Ecotoxicological risk associated with DDT ( ), endosulfan (▲), and lindane ( ) levels in sediments. Classes I and II, no toxic effects could be generated in the biota; Class III, long-term exposure could cause harmful effects on biota; Class IV, short-term exposure could damage biota; Class V, short-term exposure could produce more severe community effects

On the other hand, HCB was only analyzed in 40% of the selected sites, and the found levels represented low or very low ecotoxicological risk there.

The other OCPs analyzed in this review are not listed in the NEQCS for bivalves, so the ecotoxicological risk associated with their concentrations could not be determined; however, the spatial distribution of these compounds could be evaluated. Heptachlor levels were less than 0.13 ng g^{-1} ww in all the sites, excepting San Vicente, Chile, where the levels in chorus mussels were 1.17 ng g^{-1} ww [72]. Drins were found at very low levels, less than 0.07 ng g^{-1} ww in almost all the sites, except in the three Chilean sites (Yaldad, San Vicente, and Corral) [72] and Guanabara Bay in Brazil [51]. Dieldrin levels ranged between 0.26 and 0.52 ng g^{-1} ww in chorus mussels from Chile [72] and 0.21 ng g^{-1} in *Perna perna* mussels from Guanabara Bay [51]. Finally, the other OCPs such as endosulfans, chlordanes, and mirex were found at levels below 0.18 ng g^{-1} ww in all the sites where they were analyzed.

Marine Mammals

There are no international guidelines for ecotoxicological risk associated with OCP levels in marine mammals. However, these animals are excellent sentinels to analyze the

environmental fate and dynamics of OCPs, such as bioaccumulation, biomagnification, sources, and transport because they usually have a wide and known distribution and dispersion, they are long-lived animals, they have large lipid reserves, and they occupy high trophic levels. Moreover, this environmental compartment was selected because there are no exhaustive reviews about OCP levels in South American marine mammals.

Marine mammals were used as biomonitor in 13 of the reviewed publications [74–86], nine of them carried out in Brazil [74–83], two in Argentina [84, 85], and one in Chile [86]. All the analyzed marine mammals were cetaceans, most of them odontocetes (e.g., dolphins) and a few mysticetes (e.g., whales).

In marine mammals, variables such as age, sex, and species could influence OCP concentrations. The analysis of OCP levels in individuals of the same site and species but of different sex and sexual maturity indicated that, in general, mature males had the highest OCP levels and mature females had lower levels than immature females (Fig. 5). This trend could be explained because female mammals could transfer part of their OCPs to their young during lactation and pregnancy [87]. On the other hand, the analysis of pesticide levels in individuals of the same site, sex, and sexual maturity but of different species indicated that interspecific variation is also large; for example, DDT levels in five mature males of different species from Sao Paulo and the Paraná coast, Brazil, ranged between 1.8 and 150 μg g^{-1} lipid weight (lw) [75], and DDT levels in five adult females of different species from the Rio de Janeiro coast ranged between 0.15 and 125.6 μg g^{-1} lw [77]. Therefore, data from organisms with similar ages, same sex, and same species were used to compare the OCP levels in South American sites. For this comparison, we used mature males of *Sotalia guianensis*, which was the most used species in South America. The result indicated that the dolphins (*S. guianensis*) of the southwest of Brazil showed higher DDT levels than those from the northeast; the maximum DDT levels were found in mature males from the Cananeia Estuary [83], the Sao Paulo and Parana coasts [75], and Baixada Santista [74], with DDT values of 93.95 μg g^{-1} lw, 52 μg g^{-1} lw, and 43.2 μg g^{-1} lw, respectively. On the other hand, HCH levels in *S. guianensis* were lower than those of DDTs, but they were similar in the northeast and southeast of Brazil (ranging between 0.011 and 0.037 μg g^{-1} lw). On the other hand, HCB levels were analyzed in all the sites where *S. guianensis* was used as a biomonitor of OCPs. In general, HCB levels in southeastern Brazil were higher than in the northeast, being on average 0.004 μg g^{-1} lw and 0.048 μg g^{-1} lw, respectively. The maximum HCB levels in *S. guianensis* were found in the Sao Paulo and Parana coasts, with values of 0.068 μg g^{-1} lw [75]. The rest of the pesticides were rarely analyzed; the only one studied in more than one article was chlordane in the Sao Paulo and Parana coasts. Its maximum level was 0.42 μg g^{-1} lw [75], and its general average was 0.157 μg g^{-1} lw, with concentrations higher than those of other pesticides such as HCB or HCHs but lower than those of DDTs.

Discussion

This review summarizes 63 studies on OCP levels in various environmental compartments in the South American coasts and seas. The studies were concentrated in Brazil and

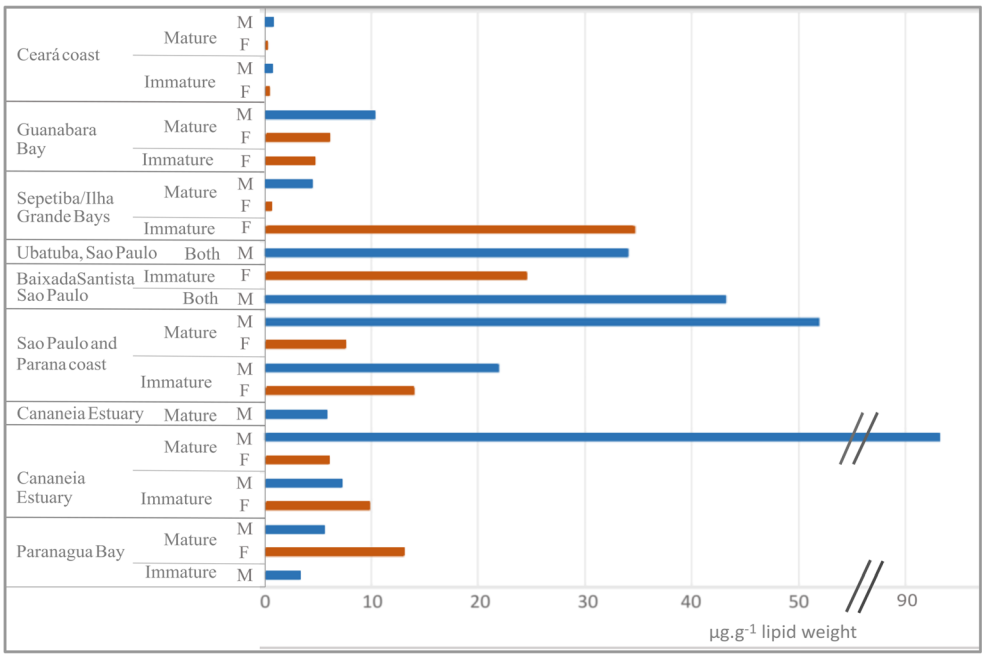

Fig. 5 DDT levels in dolphins of the genus *Sotalia* from the coasts of Brazil. Ordered according to the latitude of the sampling site, from north to south. Divided according to sexual maturity and sex: male (M) and female (F)

Argentina, in places close to large cities and in coastal landforms such as bays, estuaries, or coastal lagoons. The spatial analysis of the OCP levels performed in this review depended on the available data, and it is probably not representative of all the South American marine environments (Fig. 2). However, the sites where the studies were performed are likely to represent the most affected ones by OCPs pollution for several reasons. Among them, Argentina and Brazil were the largest consumers and producers of these pesticides in the region, and most of the large cities in South America are located in the areas of greatest agricultural production in each country [88]. Moreover, the most studied environments were those with the greatest potential to accumulate OCPs due to the high residence time of the water, where the process of loss of OCPs by export is limited [89]. They act as sinks for continental contributions from agricultural fields or large cities [14], which provides pesticides, nutrients, and organic matter to the system and increases the adsorption capacity of OCPs in environmental matrices [5].

When comparing the OCP levels in South American sites with different environmental conditions, we found that OCP levels were generally higher in semi-enclosed environments near highly anthropized areas, that is, with intensive agricultural, industrial, commercial, or urban activities (Figs. 2, 3, and 4). Many sites with similar conditions, however, have not been evaluated for OCPs, including many coastal sites in Venezuela, Suriname, French Guiana, Guyana, and Ecuador, because no publications were found on OCP levels in marine environments there. Furthermore, many sites have been evaluated for OCPs but in few matrices or for few analytes, then the information was not enough to understand the real problem of OCP levels in these environments. This problem was observed in Chile, Peru, Colombia, and the Brazilian northeast where the number of publications is low and only one environmental compartment was analyzed in each site.

Another issue to consider was that very few studies were conducted in remote and sensitive coastal environments, such as coral reefs, salt marshes, or mangroves. It would be interesting to study these environments, since they constitute highly productive sites and support a great biological diversity that could be affected by OCPs.

Regarding data analysis, most of the considered scientific articles use the same unit for the same matrix, which made it possible to compare the OCP levels between sites. However, data management for levels in fish and marine mammals was difficult because the concentrations had been reported based on wet, dry, or lipid weight. This problem could be solved in cases where the lipid or moisture content had been reported by converting the values and units; however, in some other cases, these data had not been reported, and the information could not be compared. Another drawback was that most articles had reported OCP levels as a sum of the congeners of each pesticide, so the spatial analysis of each congener was very limited. For this reason, we decided to analyze the distribution of the corresponding summations. However, when it was relevant, the levels of some pesticide congeners were used separately (e.g., to assess the age of DDT input into the environment and to evaluate the associated ecotoxicological risk with lindane levels in the sediment). Another problem related to this issue was that few studies show the ranges of OCP levels for each site; most of them report average values, so the ecotoxicological risk analysis was performed only with the average values. It would have been interesting to evaluate the ecotoxicological risk associated with maximum OCP levels at each site, since it would have allowed us to observe the most serious effect that pesticides could generate there.

Regarding the determination of the ecotoxicological risk in different compartments of South American marine environments, the NEQCS allowed classifying the sites according to the ecotoxicological risk associated with OCPs to make visible the importance of the problem that OCPs could generate in these environments. However, the NEQCS may not have been appropriate for the study area because it was developed for typical species of the Norwegian coastal environment (i.e., the biota of South America could have different sensitivity to that of Norway at the same OCP levels). Therefore, an ecotoxicological risk classification system for South American coastal environments, evaluating the effects of OCP levels (and other pollutants) in typical species of South America, should be provided.

Finally, in relation to the use, stock, and production of OCPs, all countries in South America have regulated the use and production of all OCPs through their own regulations and adhering to international treaties such as the Stockholm Convention on POPs and the Rotterdam Convention on hazardous chemicals and pesticides. South American countries have also developed National Implementation Plans (NIPs) of the Stockholm Convention on POPs to provide inventory and safely dispose of POPs stocks, among other objectives. However, most of the NIPs of South American countries show poor quality and limited information on the stock, import, production, and use of the OCPs in each country. It is thus imperative to have information on the real stock of OCPs in each country and to develop efficient actions to eliminate them safely. Moreover, it is necessary to give attention to the illegal use of some of the pesticides such as DDT, since it has been observed that in some places the DDT/DDE ratio indicates that the input to the environment was recent.

Conclusions

This review summarizes the information of 63 scientific articles published over the last 20 years on organochlorine pesticides in South American marine environments to evaluate their levels in water, sediment, bivalves, fish, and marine

mammals. The main conclusions of this review are the following:

- The highest OCP levels were found near large cities and in semi-enclosed coastal environments such as estuaries, bays, or coastal lagoons.
- In general, DDT levels were markedly higher than other OCPs in all environmental compartments, excepting the air. They were followed by HCH, heptachlor, mirex, and endosulfan levels, depending on the matrix and the region. Aldrin, dieldrin and endrin, HCB, methoxychlor, and chlordanes were found at very low levels in most matrices and sites, except for exceptional cases.
- In the air, the most abundant OCP was endosulfan, which has been recently banned in several countries, followed by HCB, a by-product of current industrial activity.

Moreover, the ecotoxicological risk associated with the levels of some of these pesticides in water, sediments, bivalves, and fish was analyzed according to the Norwegian Environmental Quality Classification System, and the conclusions are the following:

- The levels of some OCPs could cause damage to the ecological communities in several coastal places.
- DDT levels in sediments, mussels, and fish could cause damage to the structure and function of the community in the long term in Guanabara Bay, the coast of Lima, the Río de la Plata Estuary, and other sites.
- Lindane and endosulfan levels in sediments could cause long-term or short-term damage to biota in several coastal sites in southeastern Brazil and the coast of the Argentinean Pampas.

Recommendations

Based on the reviewed evidence, we list a set of recommended actions:

- State control over the illegal use of OCPs in agriculture crops and stockpiles should be improved.
- An updated monitoring should be carried out on the OCP levels in matrices with little temporal variability such as sediments or bivalves in South American coastal environments. Studies should address semi-closed environments such as coastal lagoons, estuaries, bays, or fjords near highly anthropized areas.
- Because of the gap in the knowledge regarding sensitive and biodiversity-rich coastal environments such as salt marshes, coral reef, and mangroves, this should be studied both to assess environmental pressure for OCPs and to understand their role in their distribution and degradation.
- New strategies and technologies for the remediation of coastal environments highly contaminated by these compounds should be researched and applied.

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