



A decadal trend study (1998–2008) of POPs in marine sediments at the south of the Southern California Bight



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HIGHLIGHTS

- No concentration differences for DDTs were found after a decade.
- Subtle indications exist of changes occurring in the composition of DDT.
- A parameter indicator of recent vs old DDT suggest temporal changes.
- Larger number of station exceeded DDTs ERLs in 1998 than in 2008.

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ABSTRACT

In this study we present a temporal analysis of two groups of persistent organic pollutants. We compare dichlorodiphenyltrichloroethane (DDT) collected in coastal sediment samples during 1998 and 2008 at the southern end of the Southern California Bight. Other group of organochlorine compounds (OCs) compared in this decadal analysis is the polychlorinated biphenyls (PCBs). For DDTs, the most abundant isomer was dichlorodiphenyldichloroethylene DDE followed by DDT. Although no statistically significant differences in total concentration were noticeable, composition-wise some differences were still observable. The fraction parameter $FDDTe = p,p'-DDT / (p,p'-DDT + p,p'-DDE)$ used as a measure of freshness of DDT use, is utilized here to show changes in composition. These changes are due to natural degradation of p,p' -DDT under mostly oxic conditions. These changes indicate a slow transformation of DDT residues to DDE. In addition, during 1998, several stations (12 stations) showed concentrations above Effect Range Low (ERL) for the sum of DDTs while only six showed exceedance during 2008. The number of extreme values was also less frequently found in 2008 samples. For PCBs, we detected statistically significant changes, however, in both years the most abundant congeners were mostly heavy congeners ($>PCB \# 77$) which may indicate old residues. PCBs concentrations were found in very low concentrations and do not appear to represent a danger to ecosystems. Possible explanations are offered as to the lack of observable temporal changes in concentration for DDTs in this important region.

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1. Introduction

Persistent organic pollutants (POPs) include a group of organochlorine (OCs) chemicals widely distributed in the environment. These substances were highly employed from the 1950s till 1990 (and even up to the 2000 in Mexico, in particular dichlorodiphenyltrichloroethanes DDTs for malaria control) mostly for agriculture pest control, to reduce damage to agricultural harvest and also for PCBs, in the electric sector among other uses. In Mexico, spraying of DDT started around 1944 (Stapleton, 1998) for malaria control. Agricultural use started just a few years later in the late 1940 including Baja California (Pérez-

Maldonado et al., 2012). Use of DDT in Mexico was completely phased-out in the year 2000. Once released in the environment, due to their persistency, these substances are able to travel to other environmental compartments. It is well recognized that most of marine pollutants come from land (www.gpa.unep.org). In particular these manmade OCs substances are expected to arrive, via different routes into the coastal environments. Agricultural, industrial waste disposal as well as urban uses of these chemicals represents the brunt of the original source into the ocean. Coastal environment such as that of the Southern California Bight (SCB), a highly populated (about 20 million people) coastal feature that extends from Point Conception in Southern California and ends south of Ensenada, Baja California, Mexico, is a large marine ecosystem with a history of sediment pollution due to OCs (Dodder et al., 2011). These compounds because of their physicochemical properties, including its persistence, toxicity, and volatility can travel long distance before decomposition. Due to their hydrophobicity, they

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can become a threat to aquatic resources and accumulate in marine sediments and in the lipids of organisms. POPs pose great risks to ecosystems and human health. As persistent semivolatile compounds, POPs are capable of traveling long distances through the atmosphere as gases or aerosols, eventually accumulating in low-temperature regions following condensation and deposition (Wania and Mackay, 1993, 1996).

Globally, temporal trend for OCs have been abundantly described as showing a clear declining trend after the 1980s (see for example Rigét et al., 2010; Kim and Yoon, 2014). The temporal decline of OCs has been based on measures such as the elimination, phasing-out and substitution of several of these substances. Our concern was based on the fact that some of the most elevated concentrations of DDTs as well as PCBs have been reported in marine sediments to the north of our study area (Zeng and Venkatesan, 1999; Zeng et al., 2002). Several studies at the north of the SCB have shown a strong presence of these pollutants (Environmental Protection Agency Palos Verdes Shelf, 2000) including in bivalves, fishes, pinnipeds and seabirds of several POPs of the Stockholm Convention. The concentrations reported for DDTs, PCBs, PBDEs and other chemicals do not limit their presence to the north of the SCB. It was stated by Jarvis et al. (2007) that as much as 82% of the total surface area of the continental shelf of SCB is tainted with OC pollution mostly PCBs and DDTs. Advection processes in suspended particle flow may carry these OCs as well as other chemicals into the southern end of the Southern California Bight (SSCB). Another concern is that about one third of the Southern California Bight is located on the Mexican side and it has been less well studied than the US part. If one wishes to understand the behavior of OCs in the SCB, one cannot ignore the Mexican portion of the Bight. In addition, we are also aware that Mexicali Valley was one of the most extensive areas in Mexico where DDTs and other OCs were used. It is expected that events such as the so-called Santa Ana winds (Hu and Liu, 2003) may transport these and other chemicals to the sea. Additionally, the urban use of chemicals for gardens, pest control and other uses may be another potential source of OCs to the marine coastal area.

In 1998, we carried-out a first work designed to measure OCs in the coastal region south of the international border of Mexico–USA. During 2008, the study was repeated to find out if the OCs had shown a declining tendency as shown elsewhere in the world (Zeng and Venkatesan, 1999). In addition, we wanted to provide a good database and a detailed record of the changes both, with a spatial as well as a temporal perspective. It was expected that a clear downward trend in most if not all of OCs concentrations would be found. The repeated presence of POPs at the north of the SCB makes the study of these compounds in the southern part of the Bight a necessity. In particular, there is interest in finding evidences of transport across borders in the ocean.

In particular, we wanted to establish a temporal comparison to determine if after a decade of coastal population and urban growth, followed by an increase in the wastewater treatment plant quality, these substances showed a specific temporal trend, that is if are declining in concentration or if on the contrary, they are becoming a larger problem due to coastal human development. In this work we compare concentrations of two groups of POPs measured in the same area in 1998 and again in 2008.

We summarize the results from 1998 and 2008 mainly for PCBs and DDTs and provide with possible explanations for these findings.

2. Material and methods

2.1. Study area

The SSCB is the south part of a large ocean feature originated from the abrupt change in direction of the western Coast in the North American Continent. It begins at Point Conception in Santa Barbara County, CA and ends at Cabo Colonett in Baja California, Mexico. It is characterized by a very large coastal population and with an even larger

number of touristic floating populations. Reduced rain regimes and dry weather also characterize this region. Due to the large number of people, there is a consequently large water consumption and wastewater discharge, through wastewater treatment plants into the ocean. There are several wastewater discharges of different qualities and volume discharging into the sea. There are officially reported eight wastewater treatment plants (WTP) within the study area. Starting from north, the city of Tijuana has two important ones; the first is the so-called binational wastewater treatment plant (BWTP) in the US side of the border wastewater from both Tijuana and San Diego discharging just at the border, about 1070 L/s. The second wastewater treatment plant is the Punta Banderas wastewater treatment plant (PBWTP) discharging volumes of about 1100 L/s in San Antonio de los Buenos Baja California. The other plants are the one at Playas de Rosarito, called Rosarito Norte (RNWTP) discharging 105 L/s and more to the south; there is a submarine discharge at the so-called Rosarito1 (R1WTP) with only 60 L/s. Further to the south, there is another small WTP at Puerto Nuevo (PNWTP) discharging 2 L/s. In the south, within the Todos Santos Bay, there are three WTP. The largest one is called el Naranjo WTP (NWTP) and it is discharging around 342 L/s, mostly into the ocean. The second in importance is the El Gallo wastewater treatment plant (GWTP) with a flow of 160 L/s, which is discharged into the Bay. Finally, the third plant is that of El Sauzal (SWTP) discharging 35 L/s.

2.2. Experimental procedure

2.2.1. Sampling designs

Sampling design for the whole SCB, including the SSCB was based on a randomly stratified design (Stevens, 1997). This probabilistic design has been previously described elsewhere (Zeng et al., 2005). The stratified randomized design insures that no under sampling or oversampling takes place within the strata. Three strata were preselected for our sampling design. The selection was based mainly on the human population distribution along the SSCB. The northern stratum was selected to include the most populated site that includes both the city of Tijuana and the city of Rosarito, Baja California (population for Tijuana in 2005 about 1.5 million). A second stratum was selected to include the second largest city, Ensenada Baja California that is located along the Todos Santos Bay. A third stratum was selected in between those two previous strata. It was originally conceived as a reference area given the reduced population that lives along this area and the lack of important wastewater works. Marine sediments were collected during 1998 and 2008 using similar field procedures established in the operation manual (Schiff et al., 2011) for 1998 and 2008. Briefly, from a randomly stratified sampling design site, the two top cm of sediments collected using a Van Veen drag of 0.1 m² capacity are collected in amber glass jars and kept frozen till analysis. The design has implication in so far as that every sample has an equal chance of selection, and that every sample represents an area for one of the strata. At the end, given the differences in area between strata, each sample has a proportionally different weight.

The sampling sites for 1998 and 2008 for the southern end of the SCB are shown in Fig. 1. It has been intentional within the sampling design, to keep a certain number of sites to be the same as before to be able to make direct comparisons. It has also been important to include new random sites to avoid missing possible important sites.

Statistical analysis included median comparisons to determine differences between years were a non-parametric Mann–Whitney U test and between strata was a Kruskal–Wallis ANOVA by Rank. To run these tests, we substituted all non-detected values by the limit of detection divide by the square root of two ($LOD/(2)^{1/2}$) as proposed by Croghan and Egeghy (2003) and by Glass and Gray (2001). This method produced less bias due to the substitution in particular for large number of non-detects.

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