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Comparisons of field and laboratory estimates of risk of DDTs from contaminated sediments to humans that consume fish in Palos Verdes, California, USA

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Abstract

Calculating risk from seafood exposure to persistent organic pollutants continues to be problematic as estimates of exposure from diet require extensive monitoring of fish species and limited assessments of bioavailability from sediments where the contaminants tend to reside. Previous studies in our laboratory utilized a laboratory-based isotope dilution method (IDM) to estimate the bioavailability of DDT [1,1,1-trichloro-2, 2-bis(*p*-chloro-phenyl)ethane] and its metabolites from sediment to biota from a superfund site on the shelf of the Palos Verdes (PVS) Peninsula in California (USA). Using a biota-sediment accumulation factor (BSAF) derived from IDM and biomagnification factors (BMF) calculated from previous studies as well as seafood-consumption data specific to anglers in the PVS area, we estimated cancer and non-cancer risks for anglers and nursing infants representing sensitive groups. Predicted cancer risks from consumption of White Croaker (*Genyonemus lineatus*) to the 50th and 95th percentile to all shore mode anglers were, respectively, 2×10^{-7} and 7×10^{-7}, which were similar to field studies using fish concentrations of all DDT isomers and their environmental degradates (ΣDDT) from collected animals. The calculated non-cancer hazard quotient values for the 50th and 95th percentile shore mode anglers consuming White croaker from this study (0.008 and 0.023, respectively) were also of similar magnitude as those obtained from studies based on samples obtained solely from fish. For nursing infants, similar results were also observed. These results indicate that estimates of bioavailability using IDM from sediment could be used accurately to determine risk to ΣDDT in humans from fish consumption.

Graphical abstract

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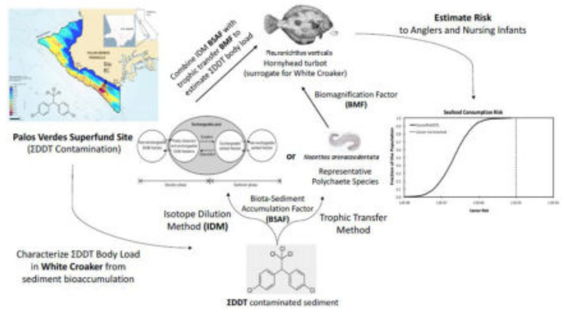

1. Introduction

From the 1950's to the 1970's, approximately 1,000 metric tons of technical grade 1,1,1-trichloro-2, 2-bis (*p*-chloro-phenyl) ethane (DDT; Chemical Abstracts Service Registry Number 50-29-3) was discharged to the Palos Verdes Shelf (PVS) in Los Angeles County, California (contamination map depicted in graphical abstract, source: CH2M Hill, 2007). The mixture consisted of ~80% p,p'- and 20% o,p'-isomers. In the marine environment, p,p'-DDT is rapidly dehydrochlorinated to p,p'-DDE during early stages of digenesis and as such p,p'-DDE represents the majority (~69%) of DDT isomers and environmental transformation products (ΣDDTs) currently in the PVS (Eganhouse and Pontolillo, 2000). DDT is an organochlorine pesticide whose effects include neurotoxicity and endocrine disruption (alteration in reproduction and development) as well as carcinogenicity (US Agency for Toxic Substances and Disease Registry, 2002). Due to the persistent and bio-accumulative nature of DDT, 40 km^2 of aquatic sediment is now designated the Palos Verdes Superfund Site (Stull et al., 1996). Strong hydrophobicity and a high affinity for organic carbon allows DDT (K_{OW}= 6.91 for p,p'-DDT) and other hydrophobic organic contaminants (HOCs) to preferentially deposit in bed sediments of aquatic systems and potentially bioaccumulate (Pontolillo and Eganhouse, 2001).

Accumulation potential to benthic organisms in sediments occurs as a function of bioavailability in addition to bulk chemical concentrations, which complicates estimates of accumulation to biota from sediments (McGroddy et al., 1996; Melwani et al., 2009). Bioaccumulation in fish represents a significant route of exposure through dietary consumption by subsistence anglers in this area, particularly of White Croaker (*Genyonemus lineatus*), which has historically been the most contaminated fish species from the PVS, (Alexander, 2000; LACSD, 2014a). Several studies have shown that dietary exposure via fish may present increased cancer and non-cancer risks in sensitive populations such as anglers and nursing infants (Klasing et la. 2009; SAIC 1999; Wilson et al. 2001).

To evaluate dietary exposure to DDT and its metabolites (ΣDDTs), a greater emphasis for decision-making has been placed on results from measurements made in fish versus physicochemically-based modeled studies due to the specific behavior of these chemicals *in vivo* (USEPA 1998). However, due to high variability in field-caught fish studies, high sample numbers are needed in order to obtain accurate estimates (USEPA 1998). Therefore, reliable data from alternative techniques, including laboratory and modeled bioaccumulation studies may provide equivalent estimates of accumulation without the need for expensive sampling in affected habitats. Moreover, passive sampling techniques have been encouraged as a scientifically robust tool for assessing exposure at United States Environmental Protection Agency (USEPA) superfund sites (USEPA 2012). A previous comparison of paired laboratory/field-based bioaccumulation factor estimates revealed that for oligochaetes, these values generally fall within a factor of 2 (Burkhard et al., 2012). However, few studies have compared risk assesment values calculated from fish ΣDDT tissue concentrations estimated from sediment bioavailability derived from passive sampling methods.

Previously, Bao et al., (2013) developed a method to estimate the bioavailable fractions of total organochlorine concentrations from sediments to biota using stable isotope labeled hydrophobic organic contaminant standards. This method, termed the Isotope Dilution Method (IDM) relies on the premise that when a small amount of an isotopic analogue is added into a solid-water system, it will rapidly and exclusively distribute itself into the accessible pool with the native hydrophobic organic contaminant (Celis and Koskinen 1999; Hamon et al. 2002). Following equilibrium with the sediment-water mixture (a short mixing period), phase separation (centrifugation and filtration) and analysis of the solution using gas-chromatography-mass-spectrometry or liquid chromatography-mass-spectrometry, one may accurately estimate the bioavailable fraction (Delgado-Moreno et al., 2013). The bioavailability of hydrophobic organic contaminants as determined by the IDM is an effective parameter for evaluating the effectiveness of remediation practices, such as activated carbon treatment (Millward et al., 2005; Zimmerman et al., 2004). An advantage of this method over previously developed passive-sampling laboratory techniques is the assumption that the added isotope labeled analogue will behave in the same way as its non-labeled counterpart, thus estimating first order rate transfer coefficients and thereby estimate biota-sediment accumulation factor based on equilibrium partitioning prior to equilibrium being reached (Delgado-Moreno et al., 2013). As such, this method can reduce the required sample analysis time from 9 days to 9 hours (Bao et al., 2013).

Using IDM, Jia et. al. (2014) assessed bioavailable ΣDDT in PVS sediment (site 8C; [33° 41.91' N; 118° 20.14' W]) and validated these results with laboratory uptake studies in a marine polychaete (*Neanthes arenaceodentata)*. Subsequent studies also using a laboratory trophic system exposing *N. arenaceodentata* and then feeding the worms to Hornyhead turbot (*Pleuronichthys verticalis*) calculated estimates of biota-sediment accumulation factors (BSAF) and biomagnification factors (BMF) (Crago et al. 2016). The estimates were comparable to ΣDDT concentrations measured by the Los Angeles County Sanitation Districts (LACSD) in Hornyhead turbot (*Pleuronichthys verticalis*) collected from the locations where sediments were sampled (LACSD 2014). The purpose of the current study was to compare laboratory estimates of bioavailability and accumulation derived from IDM in PVS sediments with estimates derived from measurements made in animals collected in the field and calculate risk from the estimates to sensitive groups (e.g. adult anglers and nursing infants of adult anglers) consuming fish in the Southern California bight. The risk values, which are based on either laboratory or field-based techniques, were then compared to previously conducted seafood risk assessments for the PVS to determine their usefulness and relative magnitude in estimating risk (SAIC, 1999; Santa Monica Bay Restoration Project, 1994; Wilson et al., 2001).

2.0 Methods

To assess bioavailability from sediments to benthic organisms, Biota-Sediment-Accumulation Factors (BSAF) were calculated using the IDM method (Jia et Al. 2014) and compared with values measured from a field study (Zeng and Tran 2002) as well as a laboratory-based biological method which used Hornyhead turbot (Crago et al. 2016). In the case of Zeng and Tran (2002) a BSAF value was not directly reported by the authors, in which case it was calculated using data reported in their publications (see Supporting Information; Table S2, Equation S1).

2.2 Biomagnification Factor (BMF)

To estimate trophic transfer from benthic infauna to fish, Biomagnification Factor (BMF) for Hornyhead turbot was calculated from a field-based study (Zeng and Tran 2002) and was compared to a laboratory-based trophic transfer study (Crago et al. 2016) (Equation S2). Since BMF values were not reported directly from the field studies, the appropriate data was used from these publications to calculate a BMF value for Hornyhead Turbot/polychaete (see Supporting Information; Table S1). In the case of Crago et. al (2016), muscle tissue ΣDDT was not available, so an estimation was made based on the liver tissue values and the calculated ratio of Hornyhead turbot liver/muscle in the field (see Supporting Information) (LACSD, 2014b). Hornyhead turbot was used as a surrogate species for White Croaker based upon feeding strategy (LACSD, 2014b; Ning-Chao, 1995). The BMF values, along with their respective BSAF values were then used to estimate White Croaker ΣDDT content for human exposures (Equation 1).

2.3 Estimation of Seafood Consumption

2.3.1 Adult Anglers—Data for the angler consumption habits of seafood from the PVS was collected from OEHHA (2000) and RecFIN (2017). The central tendency and 95th percentile values for fish consumption of Palos Verdes boat anglers used in this study are 30.5 and 85.2 grams/day, respectively (OEHHA, 2000). Scenarios calculated herein assumed that an angler would eat a variety of fish species as surveyed by the Santa Monica Bay Restoration Project (1994), with the consumption rate for White croaker calculated by multiplying the species diet fraction for pier and dock anglers by the overall fish consumption rate for all boat anglers in the area based on recent consumption rate data (RecFIN, 2017). White croaker represents 1.19% (or 0.44 and 1.22 g/day; median and 95th percentile, respectively) of fish consumed by boat anglers (OEHHA, 2000; RecFIN, 2017).

2.3.2 Calculation of Dosage of ΣDDT from Ingested White Croaker—*ΣDDT* contaminant exposure via White croaker ingestion was estimated based on the concentration calculated in the fish using BMF/BSAF values and the seafood consumption rate. Using the calculated BMF and BSAF values from each respective method, with Hornyhead turbot as a model species, the concentration of ΣDDT in White croaker (C_f) may be calculated using equation 1.

$$C_f = V_f \times {}^{C_s}/_{TOO} \times BSAF \times BMF \quad \text{Equation 1}$$

Equation 2 was then used to calculate dose via ingestion of contaminated White croaker (OEHHA, 2000):

$$ADD_{White\ Croaker} = (C_f \times Ifish \times GI \times EF \times F_{White\ Croaker}) \quad \text{Equation 2}$$

Where $ADD_{White\ Croaker}$ = dose of contaminant via ingestion of White croaker for adults (mg/kg BW-day), *Ifish* = sport fish ingestion rate of all fish from adults (g/kg BW-day), *GI* = gastrointestinal absorption fraction, unitless [default =1], *EF* = exposure frequency (days/365 days) [The exposure frequency (EF) is set at 350 days per year (i.e., per 365 days) to allow for a two-week period away from home (US EPA (1991), $F_{White\ Croaker}$ is the fraction of White croaker in the diet of pier and dock anglers in the Palos Verdes shelf (0.0119) (RecFIN, 2017).

2.3.3 Nursing Infant Exposure—Exposure to ΣDDTs through seafood in the PVS may be separated into three distinct pathways: maternal transfer during the third trimester of pregnancy, breast milk consumption during the first year of life and consumption of fish from 1–70 years of life. Estimating dose for the fetus during the third trimester of pregnancy is not straightforward, and varies for each based on toxicokinetics. While this data is not available for DDT, OEHHA (2012) suggests that a reasonable approximation of the dose during the third trimester may be made by assuming the dose (mg/kg-body weight) is the same as the mother's dose (mg/kg-body weight), with 70 kg representing the default body weight for an adult female. Based on the values described in OEHHA (2000) this yields a mean and high-end time-weight averaged "ingestion rate" of 0.44 and 1.22 g/day-kg-body weight, which compares to 0.306 and 1.53 g/day-kg-body weight based on the SAIC (1999) values.

A high-end estimate of exposure to DDT in this life stage may be reasonably calculated by considering breast milk consumption as the primary source of this toxicant (OEHHA 2012). OEHHA (2012) suggests calculating exposure from mother's milk pathway only during the first year of the 0–2-year age group, with a daily exposure frequency. To estimate the breast milk lipid concentration of ΣDDT, a method based on the observed relationship between daily intake of ΣDDT and body fat concentration in humans was used (Mariën and Laflamme, 1995). This equation relies on the observed relationship by Durham et al. (1965) from human data in which the ratio of ΣDDT concentration in adipose tissue and breast milk lipid concentration is 1:1, thus deriving a linear algorithm (equation 3) for breast milk lipid concentration of ΣDDT.

$$ln\ BMLC\ (mg/kg-milk)=0.7\ ln\ (ADD\ (mg/kg-day-BW)\times BW_{adult}(kg))+3$$ Equation

3

Where BMLC is the breast milk lipid concentration in mg/kg-lipid, ADD is the average maternal daily dose of ΣDDT in mg/kg-day-BW (calculated above), BW_{adult} is the default weight of an adult woman (70 kg) (USEPA 2014). The infant daily intake (IDI; mg/kg-day) may then be approximated using equation 4.

$$IDI(\text{mg/kg- day- BW})=BMLC(\text{mg/kg- milk})\times BMI_{bw}(\text{kg- milk/kg- BW- day})\times PMF(\%)$$

Equation 4

Where BMI_{bw} is the daily breast-milk ingestion rate of a "fully-breast-fed" infant (kg-milk/kg BW/day; 0.101 and 0.139 median and 95th percentile, respectively) (OEHHA, 2012) and *PMF* is percent milk fat in breast milk (4%; unitless) (Mariën and Laflamme, 1995). Thus, based on the calculated concentration of ΣDDT in White croaker using the Bao et al (2013) IDM method (1.3x10^{-5} mg/g in fish muscle tissue), the infant daily intake would be 3.4 x10^{-4} and 9.8 x10^{-4} mg/kg-day-BW for the median and 95th percentile, respectively.

2.5 Calculation of Non-cancer hazard quotient and Cancer Risk

Non-cancer hazard quotient was evaluated for infants and adults using the USEPA non-cancer hazard quotient equation (equation 5) and the ΣDDT reference dose of 5x10^{-4} mg/kg-day-BW (USEPA 2000a).

$$\text{Hazard Quotient}=\frac{ADD_{White\ Croaker}\left(\frac{mg}{kg}-day-BW\right)}{\text{Reference Dose}\left(\frac{\text{mg}}{\text{kg}}-\text{day}-\text{BW}\right)}$$ Equation 5

According to OEHHA (2009), the potency of carcinogens, and thus cancer risk, varies based on the life stage at exposure. To address this concern, OEHHA suggests applying a weighting factor to early life exposures, termed the Age Sensitivity Factor (ASF). Cancer risk is multiplied by an ASF of ten to weight lifetime risk from exposures occurring from the third trimester of pregnancy to less than two years of age. Similarly, an ASF of three is applied for ages two-sixteen. Accounting for effects of early-in life exposure requires accounting for both the increased potency of early life exposure to carcinogens as well as the greater exposure on a per kg body weight that occurs early in life due to behavioral and physiological differences between infants, children and adults. When considering the worst-case scenario of exposure from *in utero* to adulthood, all listed terms in equation 6 are added. When only considering adult exposure, the final term (ages 16–70) is used and the rest are omitted.

$$\text{Excess Lifetime Cancer Risk}=$$
$$\frac{ADD_{third\ trimester}\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})\times CSF\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})^{-1}\times ASF(10)\times 0.3\ \mathrm{years}}{70\ \mathrm{years}}$$
$$\frac{IDI\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})\times CSF(\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})^{-1}\times ASF(10)\times 1\mathrm{year}}{70\ \mathrm{years}}+$$
$$\frac{ADD_{1-2\ year}\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})\times CSF(\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})^{-1}\times ASF(10)\times 1\ \mathrm{year}}{70\ \mathrm{years}}+$$
$$\frac{ADD_{2-16\ years}\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})\times CSF(\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})^{-1}\times ASF(3)\times 14\ \mathrm{years}}{70\ \mathrm{years}}+$$
$$\frac{ADD_{16-70\ years}\ (\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})\times CSF(\frac{\mathrm{mg}}{\mathrm{kg}}-\mathrm{day}-\mathrm{BW})^{-1}\times ASF(1)\times 54\ \mathrm{years}}{70\ \mathrm{years}}$$

Equation 6

Where $ADD_{1\text{-}2yrs}$ and $ADD_{2\text{-}16\ years}$ are the same as adult exposure in mg/kg-day-bw, adjusted for the time-averaged body weight of each age group (9.7 and 37.0 kg, respectively) (OEHHA 2012). OEHHA (2012) suggests assuming the daily dose in the third trimester is the same as the maternal daily dose ($ADD_{16\text{-}70\ years}$ in mg/kg-day-BW). *CSF* is the cancer slope factor for DDT and p,p'-DDE (0.34 mg/kg-day-BW)$^{-1}$ (USEPA, 2000a).

2.5.1 Stochastic Analysis—While threshold-based, deterministic risk assessment methods have often been used in the past, the U.S.EPA (2004) has focused in recent years to use probabilistic methods to account for and represent uncertainty in its risk assessments. The linkage between sediment-associated bioaccumulative compounds (DDT, PCBs) and their ability to cause deleterious effects to wildlife and potential health risks to humans has been well-established (Alava et al., 2012; Anderson et al., 1975; Beyer et al., 2014; Hesslein et al., 2011; Huang et al., 2008; Kidd et al., 1995; Schaeffer et al., 2006; Wiener and Suchanek, 2008). As such, the state of California has developed a draft framework to assess whether sediment at a site or water body meets the Sediment Quality Objective (SQO), meaning that "pollutants shall not be present in sediments at levels that will bioaccumulate in aquatic life to levels that are harmful to human health" (California State Water Resources Control Board, 2009; Greenfield et al., 2015). This framework follows a 3-tiered approach dependent on two indicators: consumption risk to humans and sediment linkage (Greenfield et al. 2015).

In order to evaluate hazard and sediment linkage, a Decision Support Tool spreadsheet model was used in which monitored fish species are divided into 1 of 8 guilds and the user inputs data for specific attributes, such as total lipid content in fish, portion of specific fish guild in total fish diet, and contaminant concentration within each guild (Greenfield et al. 2015). Other relevant site-specific attributes, including sediment contaminant concentration, total organic carbon, site size, and fish consumption data were also added (Table S2) (LACSD, 2014b). The Monte Carlo simulation was then used to generate probabilistic distributions of cancer risk, non-cancer hazard, and sediment linkage to the seafood contamination per the instructions that accompany the excel spreadsheet from Greenfield et al. (2015).

Since our risk assessment focused on a specific fish guild based on comparable feeding strategies (White croaker/Hornyhead turbot), and did not consider other fish guilds or trophic transfer models. Consumption data for White croaker, as discussed earlier, was used (30.5 and 85.2 g/day, 50th and 95th percentile, respectively, with White croaker representing 1.19% of total seafood diet for pier and dock anglers) (OEHHA, 2000; Recreational Fisheries Information Network (RecFIN), 2017). An additional risk assessment was evaluated using SQO DST, which utilized PVS-specific field tissue contaminant loads (NOAA, 2007) for a variety of commonly consumed species for a mixed-species dietary comparison. The Monte Carlo simulation was set to run 5,000 times to obtain a stable cumulative distribution function.

3. Results

Biota-sediment accumulation factors for sediment collected from site 8C PVS are compared in Table 1. The BSAF calculated from values obtained from the laboratory-based IDM (0.0105 ± 0.0026; N=9) (mean ± one standard error of the mean; N= number of replicates) (Jia et Al. 2014) was about 3-fold greater than the value based on the laboratory exposure of 8C PVS sediment to polychaetes (*Neanthes arenaceodentata)* (0.0031 ± 0.0001; N=5) (Crago et al. 2016). IDM BSAF was less than a field-collected tissue-based study (1.1 ± 1.3; N=2) (Zeng and Tran 2002).

BMFs for polychaete to Hornyhead turbot from site 8C PVS are also compared in Table 1. The calculated BMF value from the laboratory trophic transfer study (Crago et al. 2016) (1.49 ± 0.47; N=5) is consistent with a previous field-based BMF study (Zeng and Tran, 2002) (1.49 ± 1.2; N=2).

Using the deterministic risk assessment equations discussed in the methods section as well as BSAF and BMF values obtained from each respective study, comparisons of deterministic cancer and non-cancer hazard quotients for adult anglers and nursing infants were made between various methods and are displayed in Tables 2 and 4 and stochastic assessments of risk for adults and infants are compared in Tables 3 and 4. These values are compared to risk assessment values for Palos Verdes subsistence fishermen obtained from other studies (SAIC 1999, Wilson et Al. 2001, Santa Monica Bay Restoration Project 1994).

The deterministic White croaker-only adult angler cancer risk values calculated from the laboratory-derived IDM BSAF (Jia et al., 2014) coupled with the laboratory trophic-transfer BMF value (Crago et al., 2016) were approximately one to two-fold higher than the laboratory based sediment/polychaete/turbot trophic transfer derived BSAF and BMF (Crago et al., 2016). The most conservative cancer risk values were obtained using the Zeng and Tran (2002) BSAF/BMF values, and were similar to the SAIC (1999) and Santa Monica Bay Restoration Project (1994) values. The stochastically-generated (SQO DST) mixed-species cancer risk assesment generated using field-based tissue values from NOAA (2007) were less conservative than the SAIC (1999) values but more conservative than the Wilson et. al (2001) values.

4. Discussion

The goal of this study was to estimate and compare risk values to sensitive human groups generated from sediment concentrations of ΣDDT. While previous risk assessments of ΣDDT from contaminated sediments to fish consuming humans in Palos Verdes have used tissue-based field values to estimate exposure (SAIC, 1999; Southern California Coastal Water Research Project & MBC Applied Environmental, 1994; Wilson et al., 2001), this study utilized a value based on an isotope-dilution method (IDM) for estimating bioavailability coupled with a laboratory trophic transfer model to calculate exposure and risk to subsistence fisherman and their offspring. The advantages of this approach may include reduced costs; quicker results and a lower impact to the site in question (which may be of greater concern in an ecologically threatened area). Overall, there was a consensus between values; however, estimates of risk made using mixed species as opposed to a single species (White croaker) were more variable and higher.

4.1 Biota-Sediment Accumulation Factor

Biota-sediment accumulation factors are necessary for the parameterization of trophic transfer models. The laboratory-based IDM BSAF was obtained using field-derived sediment samples equilibrated *ex-situ* with a polydimethylsiloxane fiber (Jia et Al. 2014). ^{13}C-Labeled or deuterated performance reference compounds were impregnated into the fiber prior to use, then used as an isotropic measure of desorption and adsorption of DDTs from sediment following mixing (Jia et Al. 2014). Using this technique, the freely (biologically) available amount of ΣDDT in the sediment was estimated based on the adsorption/desorption of isotopically-labeled ΣDDT. By relying on the desorption of the pre-loaded isotope-labeled analogue from the polyethylene device, the bioavailability of the compound may be estimated from the desorption rate after a short or flexible sampling time (Bao et al., 2013; Jia et al., 2014). To confirm the precision of this technique in estimating bioavailability, a laboratory-based biological BSAF value was calculated by exposing 40 polychaetes (*Neanthes arenaceodentata*) to PVS site 8C sediment (either low DDT sediment, high DDT sediment or control sediment) for four days with subsequent evaluation of DDT residues in the worms (Crago et al. 2016). Most values were in the same order of magnitude compared to the LACSD (2014) study. However, few replicates (N=2) and thus a high degree of variability were likely responsible for greater differences with another field-based BSAF value from Zeng and Tran (2002).

4.2 Biomagnification Factor

In addition to biota-sediment accumulation factor, biomagnification factors are also necessary for the parameterization of trophic transfer models. Hornyhead turbot (*Pleuronichthys verticalis*) was used as a surrogate model for White croaker (*Genyonemus lineatus*) due to their similarities in feeding habits and abundance in the Palos Verdes Shelf. Specifically, White croaker and Hornyhead turbot are known to consume primarily polychaetes as adults and are found in habitats with high polychaete density and sediment total organic carbon (Ahr et al., 2015; Allen and Collection, 1982; Cooper, 1993; Love et al., 1984; Malins et al., 1987; Ware, 1979). The calculated mean lipid-normalized concentration of ΣDDT in White croaker from this study (611 μg/kg) is within the range of field-collected

White croaker muscle tissue from LACSD between 2012 and 2013 (307.3–926.9 μg/kg, respectively; zones 1–3) (LACSD, 2014a).

The calculated BMF value from the laboratory trophic transfer study (Crago et al. 2016) was similar to values from field-collected samples (Zeng and Tran, 2002). These studies utilized the same benthic demersal fish as a model for biomagnification, Hornyhead turbot (*Pleuronichthys verticalis*). The higher variability associated with the BMF value from the field-based value (Zeng and Tran, 2002) compared with the laboratory trophic transfer study (Crago et al. 2016) is likely due to the lower number of replicates.

4.2 Estimation of Seafood Consumption

The most comprehensive survey on seafood consumption in the PVS was conducted in 1994 by the Southern California Coastal Water Research Project (SCCWRP) and MBC Applied Environmental Sciences. From September 1991 to August 1992, 1,243 anglers were interviewed at 29 sites, including piers, jetties, private boats, party boats and beach and consumption estimates for 8 common fish species was recorded (Southern California Coastal Water Research Project & MBC Applied Environmental, 1994). This survey asked participants to recall consumption from the past 28 days in order to calculate a daily rate. However, angler bias has called into question some of the values from this study. To address this bias, Wilson et. al (2001), used a Monte-Carlo microexposure event modeling evaluation. SAIC (1999) and Santa Monica Bay Restoration Project (1994) risk assessments used consumption values of 21.4 and 107.1 g/day for the mean and 95th percentile, respectively. However, these values have been criticized as being overestimates of consumption due to avidity bias, because the survey was designed as an intercept survey, and thus over-sampled frequent anglers (OEHHA, 2000; Price et al., 1994; UESPA, 1997). To correct the values for avidity a Monte-Carlo simulation was employed, resulting in 30.5 and 85.2 g/day consumption rates for mean and 95th percentile (OEHHA, 2000). The Wilson et. al (2001) study claimed that the listed mean of 21.4 g/day of the Santa Monica Bay Restoration Project is beyond their calculated 99.9th percentile of the long-term estimate. While the basis of this claim of overestimation of seafood consumption is consistent with the critique discussed in OEHAA (2000), fish consumption values for all risk calculations performed in our study are based on the re-calculated values reported in OEHHA (2000) due to their more conservative nature. Thus, the difference in total fish consumption rate for PVS anglers as well as the use of a single-species diet (White croaker; 1.19% of diet) largely accounts for the discrepancies between risk assessment values calculated using the IDM/ trophic transfer method and values calculated in previous risk assessments (SAIC 1999, Santa Monica Bay Restoration Project 1994, Wilson et. Al 2001).

Since 1994, efforts have been made to inform Los Angeles county anglers of the risks associated with consuming contaminated White croaker (Jonick et al., 2010; OEHHA, 2009b). Due to the implementation of these social marketing campaigns, an accurate estimate of ΣDDT exposure through White croaker should utilize the most up-to-date consumption rates for Palos Verdes shelf anglers. As such, catch number and mass data from 2004–2014 for 66 species was compiled from the Pacific States Marine Fisheries Commission's Recreation Fishing Information Network database (RecFin.org). A total of

53,162 anglers were interviewed between 2004–2014 on piers, docks, jetty's, beaches, party boats and charter boats accounting for a total of 42,462 kg estimated catch of total fish (Recreational Fisheries Information Network (RecFIN), 2017). Analysis of this data reveals a persevering popularity of White croaker by anglers, as it represents 1.18% of the total estimated mass of caught species by pier/dock anglers and 2.8% of total caught species by charter boat anglers (RecFin.org 2016). This popularity has fluctuated between 2004 and 2014, ranging between 4.97% of the total mass of fish caught by all anglers in 2005 to 0.36% in 2013 (Recfin.org 2017). An apparent reduction in the popularity of this species was observed between 2010 and 2014, possibly due to the implementation of social marketing campaigns (Jonick et al., 2010; OEHHA, 2009b). However, a resurgence in popularity in 2014 seems to have occurred, as 2.2% of total caught species by mass were White croaker (all fishing modes) (Recfin.org 2017). Graphical representation of this data is available in supplementary information (Figure S1 and Figure S2).

4.3 Deterministic Cancer Risk and Non-Cancer Hazard Quotient

The cancer risk and non-cancer hazard quotient values for anglers listed in Table 2 were derived in this study using equations described above, except for the SAIC (1999), Santa Monica Bay Restoration Project (1994) and the Wilson et al (2001) values, which were taken directly from their respective publications and serve as external comparisons. The methods for cancer risk and non-cancer hazard quotient calculations used herein were based on those used by the USEPA and the State of California (OEHHA, 2012; USEPA, 2009). Differences in the values represented in this table to external comparisons may be in part explained by the older USEPA methodology (1995) used by the SAIC (1999) and Santa Monica Bay Restoration Project (1994), which was based on a high-end exposure scenario (>90th percentile).

The median cancer risk and non-cancer hazard quotient values for White croaker obtained from the laboratory-based biological investigation (Crago et al. 2016) and IDM study (Jia et Al. 2014) are smaller than a previous risk assessment's field-derived values for White croaker (SAIC 1999). This difference in risk may be explained by the previous assessment's use of higher daily fish consumption values based on the afore-mentioned un-adjusted survey values (Southern California Coastal Water Research Project & MBC Applied Environmental, 1994).

The cancer risk and non-cancer hazard quotient values for anglers and infants (Tables 2 and 3) obtained using the BSAF value from the isotope-dilution method (Jia et Al. 2014) and the BMF from the laboratory-based trophic transfer study (Crago et al. 2016) were acceptable estimations of bioavailability of ΣDDT in sediment based on their similarity to other risk assessment values based on field, lab and/or probabilistic methods.

4.4 Stochastic Application of Sediment quality objectives for California

Median cancer risk and non-cancer hazard quotient values calculated using a stochastic risk assessment approach (SQO DST) with White croaker ΣDDT tissue estimates derived from the BSAF value from IDM (Jia et Al. 2014) coupled with the BMF value from laboratory-based biological trophic transfer (Crago et al 2016) were comparable to other risk assessment values generated for this area using stochastic and deterministic methods (Table 2).

Comparing the White croaker-only diet values with mixed-species diet assessments is difficult due to the small contribution of ΣDDTs from White croaker to a mixed-species diet (1.19%). Thus, to evaluate how this SQO model compares with other mixed-species diet risk assessments, the model was also run using field-based tissue data of 20 different fish caught in the Palos Verdes Shelf from a previous survey (NOAA, 2007). Specifically, tissue sample data was used from fish caught in the "PV12-13" site as described in the NOAA survey (2007) as this collection site was nearest to the Palos Verdes outfall (site of highest DDT contamination). The results from this parameterization are more conservative than a recent Monte-Carlo risk assessment for the area (Wilson et al. 2001), likely due to smaller consumption rates employed by the Wilson et. al study. However, this SQO model is less conservative than the SAIC (1999) Monte Carlo Simulation risk assessment, likely due to the higher fish consumption rate data used in the 1999 study.

4.5 Risk to Nursing Infants

In the first year of life, breast-fed infants may be exposed to significantly higher levels of ΣDDT on a weight per body-weight basis due to the lipophilic nature of the compound and its tendency to become concentrated in breast milk (OEHHA 2012). Considering this higher exposure, as well as the increased susceptibility to toxic effects at this early life stage, it is no surprise that the estimates of cancer risk for a full lifetime exposure (Table 4) are approximately one to two orders of magnitude greater than adult-only exposure (Tables 2–3) and as such, under the conservative assumptions of this study, the site still poses a potential health hazard to infants. Despite the increased risk of cancer, OEHHA (2012) suggests that the benefits of breastfeeding generally outweigh the risks to the infant exposed to toxicants through this pathway.

It is important to note that infants born to mothers 30 years of age or older possessing college degrees and having high income may have higher exposure rates to ΣDDT through this pathway as they are most likely to be fully breast-fed (OEHHA, 2012). The term "fully-breast-fed" applies to infants that receive breast milk as the primary, if not sole, source of milk for at least the first 6 months of life (OEHHA, 2012). Per a National Immunization Survey carried out by the Centers for Disease Control and Prevention in 2006, approximately 49–52% of infants in the general population of California would fall into this category (OEHHA, 2012).

5. Conclusion

Since sediments are more readily collected than biota, the use of the isotope-dilution method may be particularly advantageous for rapid assessment of risk to humans consuming fish that reside in sediments contaminated with hydrophobic contaminants. The cancer risk and non-cancer hazard quotient values calculated using the IDM method presented in this study validates the IDM method as a reliable tool to model bioavailability of organochlorines (specifically DDT and its metabolites) in marine sediment. The calculated cancer and non-cancer risk values for the laboratory-based IDM method (Jia et Al. 2014) have between a one to two-fold difference between a laboratory-based trophic transfer method (Crago et al. 2016) and are approximately two orders of magnitude less conservative than a previous field-based method that relied on older USEPA methodology for calculating risk and higher consumption rate values (SAIC 1999). While the IDM method coupled with a laboratory trophic transfer model may accurately estimate the risks to human health from White Croaker, the estimate may not be representative of all fish guilds, thereby underestimating risk. However, it may serve as a useful, relatively simple and reliable tool for screening sediments and co-occurring fish species for contamination.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Abbreviations

DDT	1,1,1-trichloro-2, 2-bis (*p*-chloro-phenyl) ethane
PVS	Palos Verdes Shelf
HOCs	hydrophobic organic contaminants
USEPA	United States Environmental Protection Agency
IDM	Isotope Dilution Method
LACSD	Los Angeles County Sanitation District
ΣDDT	total bioavailable DDT
BSAF	biota-sediment accumulation factor
BMF	biomagnification factor
C_f	concentration of ΣDDT in White Croaker
V_f	lipid fraction in the tissue of White Croaker
C_s	ΣDDT concentration in sediment
TOC	total organic carbon content
C_p	concentration of ΣDDT in polychaete
V_p	lipid fraction in polychaete tissue
OEHHA	California Environmental Protection Agency Office of Environmental Health Hazard Assessment
BLMC	breast milk lipid concentration of ΣDDT

ADD average daily dose of ΣDDT from fish

BMI_{bw} body weight-normalized breast milk ingestion rate during the first year of life

CAPCOA California Air Pollution Control Officers Association

I_{fish} sport fish ingestion rate

GI gastrointestinal absorption fraction

EF exposure frequency

ASF age slope factor

CSF cancer slope factor

SQO sediment quality objective

USACE US Army Corps of Engineers.

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HIGHLIGHTS

- Isotope dilution is used to estimate sediment to polychaete bioaccumulation and is comparable to field-based estimates.
- Non-cancer and cancer risk assessment values for DDT in the Palos Verdes Peninsula to boat angler and nursing infants in California (USA) using IDM and benthic flatfish as food source are similar to field based evaluations.
- Non-cancer and cancer risk estimates to fishermen and nursing infants based on mixed fish species diets tends to underestimate risk to populations in the Palos Verdes Peninsula.

Table 1

Comparisons of Biota-Sediment Accumulation Factor (BSAF) and Biomagnification Factor (BMF).

Study	BSAF Method	BMF Method	BSAF Value*	BMF Value*
Bao et. al (2013)	IDM (Lab)	N/A	0.0030 ± 0.0011 (9)	N/A
Crago et. al (2016)	Trophic Transfer (lab)	Trophic Transfer (lab)	0.0031 ± 0.0001 (5)	1.49 ± 0.47 (8)
Zeng and Tran (2002)	Field	Field	1.117 ± 1.28 (2)	1.49 ± 1.2 (3)

SE= one standard error of the mean value based on the reported standard deviation and sample size (N). All values were calculated using data reported in each respective publication using the equations described in the methods section (2.1 and 2.2).

*Mean ± SE (N)

Table 2

Deterministic cancer and non-cancer estimates for adult anglers in the Palos Verdes shelf are compared to values from previous risk assessments.

			Consumption rate (g/day)		Cancer Risk		Non-Cancer HQ	
Model	**BSAF**	**BMF**	**(mean, 95th)**	**White Croaker only Diet**	**Mean**	**95th**	**Mean**	**95th**
Lab	0.011	1.49	0.36, 1	**IDM** BSAF **Jia** *et. al* (2014) + **Trophic Transfer** BMF *Crago et. al* (2016)[A]	2×10^{-7}	7×10^{-7}	0.008	0.023
	0.003	1.49	0.36, 1	**Trophic Transfer** BSAF + BMF *Crago et. al* (2016)[A]	7×10^{-8}	2×10^{-7}	0.002	0.007
Field	1.117	1.49	0.36, 1	*Zeng and Tran* (2002)[A]	3×10^{-5}	7×10^{-5}	0.9	2.4
	N/A	N/A	0.48, 28	*SAIC* (1999)[B]	9×10^{-6}	2×10^{-3}	0.2	26
	N/A	N/A	30, N/A	*Santa Monica Bay Restoration Project (1997)*[C]	2×10^{-3}	N/A	9.9	N/A
				Mixed-species Diet				
Field	N/A	N/A	21.4, N/A	*SAIC* (1999)[B]	2×10^{-5}	N/A	0.30	N/A
	N/A	N/A	21, N/A	*Santa Monica Bay Restoration Project* (1997)[D]	2×10^{-4}	N/A	0.50	N/A

[A] Value calculated in this study. Assumes a total fish ingestion rate of 30.5 g/day mean and 85.2 g/day 95th percentile (OEHHA 2000), 1.2 percent of diet is White Croaker (Recreational Fisheries Information Network (RecFIN), 2017). Pier/dock fishing modes only.

[B] Value calculated in respective publication. Mean tissue concentrations from PV shelf, boaters only. Mean and 95th percentile values based on central tendency exposure and reasonable maximum exposure scenarios, respectively.

[C] Value calculated in this study. Fish consumption rate based on OEHHA (2000). Mean tissue concentrations of mixed-species fish based on NOAA (2007).

[D] Value calculated in respective publication. Assumes total fish ingestion rate of 1.9 and 7.4 g/day mean and 95th percentile, 2.6% of diet is White Croaker.

N/A - Information not calculated in respective publication, or not applicable.

Table 3

Stochastic cancer and non-cancer estimates for adult anglers in the Palos Verdes shelf are compared to values from previous risk assessments.

			Consumption rate (g/day)		Cancer Risk		Non-Cancer HQ	
Model	**BSAF**	**BMF**	**(mean, 95^{th})**	**White Croaker only Diet**	**Mean**	**95^{th}**	**Mean**	**95^{th}**
Lab	0.011	1.49	0.36,	**IDM** BSAF **Jia** *et. al* (2014) + **Trophic Transfer** BMF *Crago et. al* (2016)[A]	7×10^{-7}	2×10^{-6}	0.009	0.03
Field	N/A	N/A	21.4, 53.0	*SAIC* (1999)[B]	6×10^{-6}	1×10^{-3}	0.2	17
				Mixed-species Diet				
Field	N/A	N/A	21.4, 107.1	*SAIC* (1999)[B]	2×10^{-4}	9×10^{-4}	0.3	26
	N/A	N/A	30.5, 85.2	**SQO DST:** *NOAA*(2007)[C]	1×10^{-5}	5×10^{-5}	0.2	0.7
	N/A	N/A	1.9, 7.4	*Wilson et Al.* (2001)[D]	7×10^{-8}	3×10^{-7}	0.003	0.01

[A] Value calculated in this study. Assumes a total fish ingestion rate of 30.5 g/day mean and 85.2 g/day 95^{th} percentile (OEHHA 2000), 1.2 percent of diet is White Croaker (Recreational Fisheries Information Network (RecFIN), 2017). Pier/dock fishing modes only.

[B] Value calculated in respective publication. Mean tissue concentrations from PV shelf, boaters only. Mean and 95^{th} percentile values based on central tendency exposure and reasonable maximum exposure scenarios, respectively.

[C] Value calculated in respective publication. Mean tissue concentrations from PV Shelf, all fishing modes.

[D] Value calculated in respective publication. Assumes a total ingestion rate of 21 g/day, 7 percent of diet is White Croaker, mean tissue concentrations from PV Shelf, all fishing modes. N/A - Information not calculated in respective publication, or not applicable.

Table 4

Comparison of Risk Results for Nursing Infants of Palos Verdes Anglers with Previous Assessments.

			Consumption rate (g/day)		Cancer Risk		Non-Cancer HQ	
Model	**BSAF**	**BMF**	**(mean, 95th)**	**Deterministic; White Croaker only Diet**	**Mean**	**95th**	**Mean**	**95th**
Lab	0.011	1.49	0.36, 1	**IDM** BSAF **Jia** *et. al* (2014) + **Trophic Transfer** BMF *Crago et. al* (2016)[A]	2×10^{-5}	6×10^{-5}	0.7	2.0
	0.003	1.49	0.36, 1	**Trophic Transfer** BSAF + BMF *Crago et. al* (2016)[A]	2×10^{-5}	5×10^{-5}	0.5	1.5
Field	1.12	1.49	0.36, 1	*Zeng and Tran* (2002)[A]	1×10^{-3}	3×10^{-3}	19.6	55.2
	N/A	N/A	0.48, 28	*SAIC* (1999)[B]	6×10^{-6}	7×10^{-3}	N/A	220
				Stochastic; White Croaker only Diet				
Lab	0.011	1.49	0.36, 1	**SQO DST: IDM** BSAF **Jia** *et. al* (2014) + **BMF** *Crago et. al* (2016)[A]	1×10^{-5}	5×10^{-5}	0.6	1.8
				Stochastic; Mixed-species Diet				
Field	N/A	N/A	30.5, 85.2	**SQO DST:** *NOAA*(2007)[C]	2×10^{-5}	7×10^{-5}	6.3	15.0

[A] Value calculated in this study. Assumes a total fish ingestion rate of 30.5 g/day mean and 85.2 g/day 95th percentile (OEHHA 2000), 1.2 percent of diet is White Croaker (Recreational Fisheries Information Network (RecFIN), 2017). Pier/dock fishing modes only.

[B] Value calculated in respective publication. Mean tissue concentrations from PVS shelf, boaters only. Based on maternal consumption of one 150-gram meal of White Croaker per month, with ΣDDT concentrations as high as 0.8 mg/kg.

[C] Value calculated in this study. Fish consumption rate based on OEHHA (2000). Mean tissue concentrations of mixed-species fish from the PVS based on NOAA (2007). Pier/dock fishing modes only.