

## **Supporting Information for**

# The persistent DDT footprint of ocean disposal, and ecological controls on bioaccumulation in fishes

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#### **Supporting text**

**Fish Model Fit.** The relationship between [DDX<sub>sed</sub>] and [DDX<sub>fish</sub>] varied by both diet and habitat (Table 1 and SI Appendix, Fig. S6). For habitat classifications, the slope-only model and the model that included both slope and intercept effects were comparable and more supported than the intercept-only model. In the slope and intercept model, slopes varied substantially between baseline (pelagic) (0.10 [0.03,0.17], mean estimate [highest posterior density 80% credible interval]), midwater (0.27 [0.19, 0.35]), benthopelagic (0.42 [0.35, 0.50]), and benthic (0.56 [0.47, 0.64]) groupings. Intercept estimates showed less variability. Baseline (pelagic), benthopelagic, and benthic groupings had similar estimates (1.72 [1.54, 1.90], 0.10 [0.03, 0.17], 0.16 [0.03, 0.36], respectively), whereas the intercept estimate for the midwater group was slightly larger (0.31 [0.07, 0.53]).

For diet classifications, the most supported model included both slope and intercept effects. Diet-model coefficients exhibit a somewhat inverse pattern to the habitat-model. Intercept estimates varied across baseline (herbivore), primary consumer, secondary consumer, and tertiary consumer groups (1.21 [0.81, 1.58], 0.25 [-0.15, 0.66], 0.66 [0.28, 1.06], 0.78 [0.39, 1.20], respectively). Slope estimates were close to estimates from the null model for primary consumer (0.54 [0.38,0.70]), secondary consumer (0.68 [0.53,0.83]), and tertiary consumer (0.67 [0.52,0.83]) groups, however, the baseline (herbivore) group had a much lower slope estimate (-0.13 [-0.29, 0.02]).

We performed typical diagnostic analyses to confirm goodness of fit, identify residual patterns, and to evaluate overdispersion for our final best-fit model. We used a simulation approach and simulated data from the fitted model conditional on the fitted random effects. We modified functions in the DHARMa R package (1) and the DHARMa.helpers R package (2) to perform calculations on our censored "brms" model output wherein the simulated values were censored to zero if predicted concentrations fell below the minimum MDL for a given fish sample. The best-fitting model indicated no diagnostic issues (SI Appendix, Figure S8). The quantile plot indicated no systematic deviation away from the assumed distribution (Kolmogrov-Smirnoff p-value = 0.26). To test for overdispersion, we compared the observed variance of the residuals to simulated variance of the residuals, as implemented in the DHARMa and DHARMa.helpers packages. We found no evidence of overdispersion of the data from the fitted model (p  $= 0.358$ ).

**Sensitivity of [DDXsed] fishing zone-averages to increasing MDLs.** The Method Detection Limits (MDLs) for 2,4′-DDE, 4,4′-DDE, 2,4′-DDD, 4,4′-DDD, 2,4′-DDT, and 4,4′-DDT in sediment samples within the Southern California Bight (SCB) generally increased through time (SI Appendix, Fig. S9). The increased detection limits are reflected in the binomial encounter model year coefficients and spatiotemporal random fields, wherein encounter is more likely given low MDLs, as everything below the MDL is treated as a zero. We conducted a sensitivity analysis to determine whether changing MDLs affected zone-wide averages for [DDXsed] that were used in subsequent fish models.

As MDLs were variable across analytes, for the sensitivity analysis we assumed a universal MDL of 1 ng g<sup>-1</sup> (or zero on the log-scale) and censored all [DDX<sub>sed</sub>] values below this threshold. This resulted in the conversion of 155 values from positive estimates to zeroes ( $N_{2003} = 5$ ,  $N_{2008} = 33$ ,  $N_{2013} = 58$ ,  $N_{2018} = 59$ ). We then ran the same sediment best-fit model (Model 8 in SI Appendix, Table S4 and SI Appendix, Fig. S10) and used output to predict the average [DDXsed] for each fishing zone corresponding to the four sampling time periods. We compared zone-wide averages for our original data to the censored data (SI Appendix, Fig. S11).

Results indicated that censoring data using a higher MDL did not substantially alter zone-wide average sediment DDX concentrations, as most zones fell on or near the 1:1 line. Fishing zones in the southern region of the SCB (e.g., San Diego Shelf) showed the largest differences between censored and uncensored data, likely due to greater censoring of data from within this region due to lower overall [DDXsed] values. As differences were small and general patterns preserved, we elected to use uncensored data within this paper.

**Proportional analysis of DDX**. We examined how the major DDX compounds, DDD, DDE, and DDT, were distributed across sediment and fish samples. For data with detectable DDX concentrations, DDE was the primary DDX compound present in both sediment and fish samples. For fish samples, an average of 97.1% of DDX was DDE, followed by DDD at 2.4% and DDT at 0.5% (SI Appendix, Figure S2). The trend is the same with sediment data, however, DDD and DDT were slightly more common. For sediments, the average DDE contribution to total DDX was 88.6%, followed by 7.9% for DDD and 3.5% for DDT (SI Appendix, Figure S2). Sediment data used in this analysis agrees with recent studies of the region, which have found that DDE is the primary DDX compound present in sediment on the PVS (3), whereas offshore stations between the PVS and Santa Catalina Island exhibit higher but variable proportions of DDT and DDD, especially within the most highly contaminated strata associated with offshore disposal (4).

We focused our analysis on total DDX, as statistically predicting contaminant compositions in fish based on sediment is challenging, and generally process-based models have been used to this end (5). Bioconcentration and biomagnification are influenced by several factors, with the octanol-water partition coefficient (log-Kow) being a key determinant. Higher log-Kow values generally lead to greater bioconcentration and biomagnification within the observed DDX ranges (6, 7). Taylor et al. (8), however, found an inverse relationship between bioaccessibility and log-Kow for DDX compounds and that bioavailability was impacted by organic matter content within sediments, as organic matter was considered to be the primary domain for binding hydrophobic contaminants (9). The relative importance of these two processes —enhanced bioaccumulation potential versus reduced bioavailability— cannot be easily resolved in a statistical framework. Our methods demonstrate that for bulk DDX, the value most often used to develop consumption advisories, a statistical relationship is sufficient to predict DDX concentrations in fish. Future work should leverage the compositional nature of the data, as summary metrics like "total DDX" encompass many tracers that, while correlated, contain unique information. Additionally, future studies could integrate fish and sediment data within a process-based modeling framework to address the complexities of contaminant uptake, although such work is beyond the scope of this study.

### **Supporting Information Figures**



**Fig. S1**. Map of the Palos Verdes Shelf superfund site (orange polygon) and 13 known deep ocean disposal locations within our study area (purple polygons).



**Fig. S2**. Maps of the relative fraction of DDE in sediment samples (A) and ternary plots of the relative fraction of DDT, DDD, and DDE in sediment samples (B), separated out by year. Ternary plot of the relative fraction of DDT, DDD, and DDE in fish composites (C). For (B) and (C), symbol color represents the log concentration of total DDX.



**Fig. S3**. Empirical data for sediment DDX presence (A) and log total DDX concentration (B), separated out by year.



**Fig. S4**. Standardized quantile residuals of DDX concentrations from the most supported model for sediment DDX presence (A) and log total DDX concentration (B), separated out by year.



**Fig. S5**. Observed vs. simulated log total DDX concentrations for sampling sites, broken apart by stratum. Residuals are generally largest in the Ports, Marinas, and Bays stratum. This may be due to high physical heterogeneity across short distances (i.e., sharp increases or decreases in depth and organic material) and numerous localized inputs of organic contaminants (i.e., runoff, rivers, proximity to industry), which are not well represented by the model. Cross validation statistics are presented in Table S4.



**Fig. S6.** Model estimated posterior distributions from the null-model (A), diet-model (B) and habitat-model (C; Table 1). The baseline category for the diet model is herbivore and the baseline category for the habitat model is pelagic. Points are the mean estimated parameter, colored boxes represent the 80% credible interval, and the black lines are the maximum and minimum for each parameter distribution. Model fits are shown in Table 1.



**Fig. S7**. Correlations between total DDX, total organic carbon (TOC), and depth across all years and stations. The top panels display the absolute value of the correlation between variables, the diagonal panels show the distribution of each variable, and the bottom panels display bivariate scatter plots and a fitted line. All values were transformed as log(x+ 1) prior to analysis.



**Fig. S8.** Diagnostic plots of best-fitting model predicting [DDXfish] as a function of [DDXsed], diet, habitat, species, and year. Left panel depicts expected (theoretical) versus observed scaled residuals, where each point is an observation, and the red line indicates observed = expected. Right panel depicts the scaled residuals as a function of rank-transformed model prediction. The horizontal line at 0.5 is the expectation, the curved lines are quantile fits for observed data.



**Fig. S9.** Log-method detection limits through time for 2,4′-DDE, 4,4′-DDE, 2,4′-DDD, 4,4′-DDD, 2,4′-DDT, and 4,4′-DDT in sediment samples within the SCB. For the sensitivity analysis, we assumed a MDL of 1 ng g<sup>-1</sup> (or zero on the log-scale, dashed line) and censored all [DDX<sub>sed</sub>] values below this threshold.



**Fig. S10.** Sediment spatiotemporal model results for censored data by year showing the probability of detection via the binomial presence-absence model (A), log total DDX estimates from the log-link gamma model (B), total estimated DDX concentrations from both models in ng  $g^{-1}$  dw (C), and the coefficient of variation on predictions (D).



Fig. S11. Zone-wide averages for [DDXsed] using original data (x-axis) and censored data (y-axis). Points are colored by fishing-zone region to better visualize spatial differences, and the dashed line indicates 1:1 correspondence.

## **Supporting Information Tables**

**Table S1.** Inventory of data used in this study. The Southern California Bight (SCB) is defined as Point Conception, CA to US-Mexico border.





**Table S2.** Inventory of fish species used in this study. Fish photos are from the California Department of Fish and Wildlife [\(https://marinespecies.wildlife.ca.gov/\)](https://marinespecies.wildlife.ca.gov/). Much of the literature on rockfish and bottom species summarized below was collated by the Pacific Coast Groundfish Fishery Management Plan (18). Fish species were classified into four categories for both diet (herbivore, primary consumer, secondary consumer, tertiary consumer) and habitat (benthic, benthopelagic, midwater, pelagic). Diet categories represent coarse approximations of trophic level for each species. This classification scheme builds off of diet classifications developed by subject matter experts for the Southern California Bight (19) that were available for 30 of the 61 species included in our analysis. Whenever possible, we deferred to this scheme for trophic classifications, and when unavailable, we used a combination of empirical diet studies from the SCB and California Current and FishBase estimated trophic level to assign diet categories.































**Table S3.** The number of fish composites within each of our habitat and diet classifications.

**Table S4.** Model selection using ΔAIC (Akaike information criteria) and 5-fold-cross-validation (CV) for alternative models predicting [DDXsed]. To conduct cross-validation we split the data into training sets (80% of the data) and testing sets (20% of the data) using unstratified random sampling. We then re-ran each spatiotemporal model using only the training data. We simulated DDX concentrations for the testing data by simulating 500 draws from the joint precision matrix, taking the median estimated value, and calculating the mean  $R^2$  and mean squared error (MSE) as a measure of out-of-sample predictive ability. Metrics were calculated on log(x+ 1) transformed data, where x is the sediment DDX concentration. Simulating data from the joint precision matrix, rather than using the expected value for a point, more accurately captured the expected distribution of zeroes and allowed us to report a single value for both models.





**Table S5**. Estimated coefficients and 90% Confidence Intervals from the beset-fit model of sediment DDX concentrations.

**Table S6**. Fish Advisory Tissue Levels (ATLs) developed by the California EPA's Office of Environmental Health Hazard Assessment (OEHHA) for DDX based on carcinogenic or non-carcinogenic risk using an 8-ounce serving size per week prior to cooking (164). Values are in ng  $g<sup>-1</sup>$  wet weight. Fish Contaminant Goals (FCGs) are estimates of contaminant levels in fish that pose no significant health risk to individuals consuming sport fish at a standard consumption rate of eight ounces per week (32 g/day), prior to cooking, over a lifetime and can provide a starting point for OEHHA to assist other agencies that wish to develop fish tissue-based criteria with a goal toward pollution mitigation or elimination.



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