Main Manuscript for

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

Lillian McGill[1]*, Toni Sleugh[1], Colleen Petrik[1], Ken Schiff[2], Karen McLaughlin[2], Lihini Aluwihare[1], Brice Semmens[1]

[1]Scripps Institution of Oceanography, University of California, San Diego, La Jolla, CA, USA
[2]Southern California Coastal Water Research Project, Costa Mesa, CA, USA

*Lillian McGill
Email: lmmcgill@ucsd.edu

Author Contributions: Paste the author contributions here.
Competing Interest Statement: Disclose any competing interests here.
Classification: Paste the major and minor classification here. Dual classifications are permitted, but cannot be within the same major classification.
Keywords: Paste the keywords here. There should be at least three and no more than five.

This PDF file includes:
      Main Text
      Figures 1 to 4
      Table 1

Abstract

Globally, ocean dumping of chemical waste is a common method of disposal and relies on the assumption that dilution, diffusion, and dispersion at ocean scales will mitigate human exposure and ecosystem impacts. In Southern California, extensive dumping of agrochemical waste, particularly chlorinated hydrocarbon contaminants such as dichlorodiphenyltrichloroethane (DDT), via sewage outfalls and permitted offshore barging occurred for most of the last century. This study compiled a comprehensive database of existing sediment and fish DDT measurements to examine how this unique legacy of regional ocean dumping translates into the contemporary contamination of the coastal ocean. We used spatiotemporal modeling to derive continuous estimates of sediment DDT contamination and show that the spatial signature of dumping is highly conserved in sediments. Moreover, we demonstrate that the proximity of fish to areas of high sediment loadings (historic dumping sites) explained nearly half of the variation in fish DDT concentrations. The relationship between sediment and fish contamination was mediated by ecological predictors (e.g., species, trophic ecology, habitat use), and the relative influence of each predictor was context-dependent, with habitat exhibiting greater importance in heavily contaminated areas. Thus, despite more than half a century since the cessation of industrial dumping in the region, local ecosystem contamination continues to mirror the spatial legacy of dumping, suggesting that sediment can serve as a robust predictor of fish contamination, and general ecological characteristics offer a predictive framework for unmeasured species or locations.

Significance Statement

In our study, we build and use a uniquely comprehensive database of DDX measurements for fish and marine sediments, spanning multiple decades and 10,000s of km2 of coastal ocean, to show that despite more than a half-century since the cessation of industrial dumping, the spatial footprint of DDX disposal is remarkably well conserved in sediments throughout the region. In addition, our study finds compelling evidence for context dependent controls on DDX bioaccumulation in biota, and clearly illustrates how the major drivers of bioaccumulation differ across a gradient of sediment contamination. Our findings provide a generalizable mechanistic framework for predicting DDX burdens in fisheries species as a function of space and ecology. Our findings also support a cautionary approach to future ocean dumping of chemicals, where place-base impacts of dumping dominate the prediction of toxin burden in fisheries, decades, if not centuries, into the future.

1. Introduction

Concerns about synthetic chemicals gave rise to the modern environmental movement in the early 1960s (Carson 1962), yet the transport, storage, and fate of these contaminants within ecological communities remains poorly understood (Bernhardt et al. 2017). For example, legacy chlorinated hydrocarbon contaminants, including organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT), continue to pose health risks to humans and wildlife despite a ban on their use in the United States half a century prior (Wilson et al. 2001, Randhawa et al. 2015, Mackintosh et al. 2016). The persistence of DDT and its breakdown products, cumulatively referred to as DDX, in the environment is due to its stability and resistance to degradation (Blais 2005). Similar to other organic contaminants, low water solubility, high partitioning to organic matter and lipids, and resistance to biodegradation result in the accumulation of DDX in sediments, bioaccumulation in organisms, and biomagnification through food webs (Kidd et al. 1998, 2001, Wang and Wang 2005). The presence of organic contaminants like DDX in fish tissues poses human and ecosystem health risks, and resource managers would benefit from tools to characterize relative risks in under sampled regions or species.

Bioaccumulation mechanisms of DDX by marine fishes are well understood and reflect a combination of uptake and elimination processes. Generally the concentration of contaminants in the organism's diet, associated with habitat or trophic position over appropriate temporal and spatial scales, represents the primary route of exposure to higher trophic level consumers such as fish (Clark et al. 1990, Borgå et al. 2004, Wu et al. 2019). Individual or species level differences in growth efficiency, foraging ecology, reproductive offloading, and the partitioning capacity of the organism for the contaminant, often associated with the whole-body lipid content of the organisms, can also be important (Madenjian et al. 1994, Hebert and Keenleyside 1995, Pastor et al. 1996, Paterson et al. 2006, Conard et al. 2022). Although individually the factors that impact bioaccumulation are well understood, there is limited knowledge of how complex, multilayer interactions affect organic contaminant transfers across species and habitats (Bonito et al. 2016). Particularly lacking is an understanding of how spatial variability in sediment contaminants interact with ecological processes to drive overall bioaccumulation. Understanding these factors, and the relative importance of their roles in bioaccumulation, is essential for predicting where risks of exposure may be high for humans and wildlife.

In heavily industrialized or agricultural regions, such as coastal areas of the Southern California Bight (SCB), concentrations of contaminants can be particularly high. Repositories of contaminants stored in marine sediments are remobilized through bioturbation and resuspension of sediments, acting as chronic sources of these compounds (Smokler et al. 1979, Zeng and Venkatesan 1999, Bay et al. 2003, Blasius and Goodmanlowe 2008). Within the SCB from 1947-1971, the world's largest producer of the pesticide DDX, Montrose Chemical Corporation, discharged its industrial waste both through the Los Angeles County wastewater treatment plant, which was deposited nearshore on the Palos Verdes Shelf (PVS), and via ships that transported and dumped barrels or bulk waste in deeper waters (Venkatesan et al. 1996, Kivenson et al. 2019). The PVS was designated as an EPA Superfund Site, and extensive monitoring of contaminants in fish tissue and sediments has occurred. However, the offshore dumpsites were largely ignored until recently, when visual confirmation of offshore dumping of containerized waste was provided by Remotely Operated Vehicle (ROV) footage (Kivenson et al. 2019). This has ignited an urgency to understand the extent and impact of the waste that was released within the region. For DDX, this legacy of pollution has resulted in a gradient of sediment contamination spanning several orders of magnitude across the SCB (Schiff et al. 2016).

Due to the long history of anthropogenic impacts in the region, contaminant monitoring efforts for sediment and fish in the SCB have been ongoing for more than two decades, resulting in a wealth of available data. We compiled a comprehensive, spatially-explicit database of existing sediment and fish DDX measurements to examine how this unique legacy of regional ocean dumping translates into the contemporary contamination of the coastal ocean. The SCB presents a unique opportunity to study fish bioaccumulation due to the large and complex gradients of DDX contamination across 10,000s of km^2 of coastal ocean and the legacy of economically and culturally valuable fisheries in the region. Through a synthesis and analysis of these data, we aim to answer the following questions:

1. How is DDX distributed in sediments throughout the SCB?
2. What is the relationship between fish and sediment DDX, and how do ecological factors mediate this relationship?

To date, toxin-related health advisories for seafood consumption have generally focused on taxonomy alone to determine which species are safe to eat, and which bear risk. By answering the questions above,

112   we aim to lay the groundwork for predictive models of exposure that are spatially explicit, but generalizable

113   across taxa with shared ecologies. Moreover, our findings will link the long legacy of regional ocean

114   dumping to contemporary ocean ecosystem processes and fisheries of southern California.

115

116   **2. Materials and Methods**

117   **2.1 Datasets**

118   The area of focus of our study is the Southern California Bight (SCB; Figure 1). Extending more

119   than 600 km from the United States – Mexico border northwards to Point Conception, California, the SCB

120   is a dynamic and productive region where the cold, southward flowing California Current mixes with the

121   warm, northward flowing Davidson Countercurrent (Hickey 1992, Bray et al. 1999). The SCB is among the

122   most biodiverse of all Northern Hemisphere temperate coastal ecosystems and one of the most densely

123   populated coastal regions in the country, home to the nation's largest commercial port, one of the largest

124   US Naval complexes, and over 20 million people. Recreational marine fisheries in the region comprise a

125   multibillion-dollar industry that is economically, socially, and culturally important (Pitchon and Norman 2012,

126   Schnaker et al. 2015).

127   We obtained the sediment and fish data used in this study from nine primary sources collected

128   between 1998 and 2021 (SI Appendix, Table S1). All laboratories that analyzed the sediment and fish data

129   were subject to a common set of rigorous quality assurance and quality control (QA/QC) guidelines to

130   ensure comparability (Du et al. 2020, McLaughlin et al. 2020). Sediment samples were collected via grab

131   samples of the top 5 cm of sediment at embayment sites and the top 2 cm at offshore sites between July 1

132   and September 30 in 2003, 2008, 2013, and 2018 (SI Appendix, Figure S1; Schiff et al. 2016, Du et al.

133   2020). Sites where concentrations were below detection limits were reported as zero values. For fish tissue,

134   samples were collected off piers and boats and saved for analysis as either muscle tissue filet with the skin

135   off (for large species) or whole fish without the head, tail, or internal organs (for small species). Generally,

136   fish samples were composites of 5-10 specimens, depending on the species and monitoring program. We

137   included only single-species composites in our analysis and subset available data to include only sediment

138   and fish samples that explicitly measured 2,4′-DDE, 4,4′-DDE, 2,4′-DDD, 4,4′-DDD, 2,4′-DDT, and 4,4′-

139 DDT, collectively referred to as DDX, and lipid content for fish samples, to ensure consistency among

140 measurements.

141   We assigned each species represented in our aggregated dataset diet and habitat classifications

142 according to their adult life history characteristics (SI Appendix, Table S3). Diet categories included

143 herbivores, primary carnivores, secondary carnivores, and tertiary carnivores and habitat categories

144 included pelagic, midwater, and demersal with the demersal group sub-divided again into benthic (species

145 that rest on the ocean floor) and benthopelagic (species found in the water just above the ocean floor).

146 Each fish composite was assigned to one of 27 spatially explicit fishing zones following McLaughlin et al.

147 (2021), however, composites from Jarvis et al. (2007) were assigned to one of 25 spatially explicit California

148 Department of Fish and Game 256 km^2 fishing blocks. Fishing zones and fishing blocks were combined

149 into a common spatial array hereafter referred to as fishing zones. Assignment to fishing zones allowed us

150 to approximate fish contaminant exposure when the exact capture location was unknown.

151 **2.2 DDX in sediment modeling**

152   For each fishing zone, we generated a time varying value for average sediment DDX

153 concentrations, [DDX$_{sed}$]. The goal with this analysis was not to determine environmental factors driving

154 sediment DDX concentrations, but rather to generate zone-averages to be used as covariates in a model

155 of fish bioaccumulation. To generate continuous spatial estimates for [DDX$_{sed}$] we fit spatiotemporal

156 regression models to sediment DDX measurements. The underlying statistical model was a spatiotemporal

157 generalized linear mixed-effects model with Gaussian random fields to model spatiotemporal processes.

158 Because concentrations were positive, skewed, and with frequent zero observations, we assume the data

159 were observed with a delta-gamma distribution (Lo et al. 1992, Stefansson 1996, Martin et al. 2005). The

160 delta-gamma, or zero-modified gamma, distribution is the mixture of a gamma distribution with a positive

161 probability mass at zero that separately models the probability of having non-zero values and positive

162 values for each sampling event. It consists of a binomial presence-absence model and a model for positive

163 values only with a gamma observation distribution and a log link.

164   We compared eight alternative models. Candidate models included a combination of spatial effects,

165 spatiotemporal effects (first-order autoregressive spatiotemporal random fields to account for correlation

166 from one time step to the next), year effects (as factors), and depth (modeled as a smooth function). We

167 used a flexible spatial modeling approach as we expected [DDX$_{sed}$] to be patchy due to an uneven legacy

168 of dumping and the distribution of organic material within sediment, which covaries with depth within the

169 SCB (SI Appendix, Figure S2; Schiff et al. 2016). We fit our models using the 'sdmTMB' R package

170 (Anderson et al. 2022), which makes use of INLA (Rue et al. 2009) to generate stochastic partial differential

171 equation matrices and TMB (Kristensen et al. 2016) to calculate the log likelihood, gradient of the log

172 likelihood and implementation of the Laplace approximation. Candidate models were compared via ΔAIC

173 (Akaike information criteria) and five-fold-cross-validation (SI Appendix, Table S1). The best-fit model was

174 subsequently used to predict average [DDX$_{sed}$] for each fishing zone corresponding to the four time periods

175 captured by both sediment and fish data: 1998-2005, 2005-2010, 2010-2015, and 2015-2020.

176 **2.3 DDX in fish modeling**

177 To understand the extent of coupling between sediment and fish DDX concentrations, we fit 11

178 candidate Bayesian linear and linear mixed-effects models to [DDX$_{sed}$] and lipid-normalized fish DDX

179 concentrations, [DDX$_{fish}$] (Table 1). We chose to use lipid normalized concentrations due to the well

180 documented, positive relationship between lipid content and organic contaminant concentration in fish

181 tissue (Hebert and Keenleyside 1995). Prior to analysis, fish and sediment values were transformed as

182 $log_{10}(x+1)$, where x is the DDX concentration, to account for right-skewness and zero values. In all models,

183 [DDX$_{sed}$] was treated as a continuous variable assuming normal errors. [DDX$_{fish}$] was treated as a

184 continuous, left censored variable where non-detect values were constrained to fall between zero and the

185 Method Detection Limit (MDL). As MDLs were variable across analytes and monitoring programs, the

186 minimum MDL across all six DDX analytes was selected for each fish composite.

187 First, to understand the mean relationship between [DDX$_{sed}$] and [DDX$_{fish}$], we included [DDX$_{sed}$] as

188 a fixed effect with no hierarchical groupings. Next, to understand if and how fish ecological traits mediate

189 this relationship, six additional models were fit to the same dataset. The first set examined the impact of

190 fish diet on [DDX$_{fish}$] values and consisted of three individual models. The first included the interaction

191 between [DDX$_{sed}$] and fish diet as a categorical variable (slope and intercept estimated for each group), the

192 second included fish diet as a fixed effect (intercept only), and the third included only the interaction between

193 [DDX$_{sed}$] and fish diet (slope only). The second set of models took the same form but examined the

194 relationship between [DDX$_{sed}$] and fish habitat (as a categorical variable). This formulation allowed the

estimation of unique slopes and intercepts between [DDX$_{sed}$] and [DDX$_{fish}$] across different diet or habitat classifications. Lastly, we fit four additional models to understand whether species specific variation and time further influenced [DDX$_{fish}$] values. Models included both diet and habitat as interaction terms for [DDX$_{sed}$], species as a random effect, and year as a continuous, centered variable.

Parameter estimates were obtained using the 'brms' R package (Bürkner 2017), which makes use of Stan (Stan Development Team 2023) to implement a Hamiltonian Monte Carlo Sampler and its extension the No-U-Turn Sampler (NUTS; Hoffman and Gelman 2014). Weakly informative priors were used for random effects (normal distribution with a mean of 0 and variance of 2) and fixed effects (Student's t-distribution with a mean of 0 and standard deviation of 0.5). Candidate models were compared via approximate leave-one-out-cross-validation (LOOIC) using the loo package (Vehtari et al. 2017), where the lowest value indicates the best model, and via the difference between theoretical expected pointwise predictive density (elpd$_{loo}$) for each model compared to elpd$_{loo}$ of the best model (Table 1). Diagnostics for candidate models can be found in the SI Appendix, Supplementary Text.

3. Results

3.1 General data description

Our final dataset consisted of 1271 sediment samples that measured DDX across the SCB, with 75% showing detectable concentrations. We obtained 1103 fish tissue composites from 61 species that measured DDX, with 93% of samples showing detectable concentrations. Fish diet and habitat classifications are partially crossed. Under our classification scheme, the pelagic habitat group contains 161 composites ($N_{Pelagic}$ = 161), of which 90 are primary consumers ($N_{PrimaryConsumer}$ = 90), 68 are secondary consumers ($N_{SecondaryConsumer}$ = 68), and 3 are tertiary consumers ($N_{TertiaryConsumer}$ =3). For the remaining habitat groups, $N_{Midwater}$ = 206 ($N_{Herbivore}$=7, $N_{PrimaryConsumer}$ = 64, $N_{SecondaryConsumer}$ = 134, $N_{TertiaryConsumer}$ = 1), $N_{Benthopelagic}$ = 601 ($N_{Herbivore}$ = 24, $N_{PrimaryConsumer}$ = 73, $N_{SecondaryConsumer}$ = 368, $N_{TertiaryConsumer}$ = 136), and $N_{Benthic}$ = 135 ($N_{SecondaryConsumer}$ = 88, $N_{TertiaryConsumer}$ = 47).

3.2 DDX in sediment modeling

Observed concentrations in [DDX$_{sed}$] ranged from 0.0 to 5182.5 ng^{-1}g^{-1} dry weight (56.72 ± 331.4 µg^{-1}g^{-1} dw, mean ± SD). The proportion of sediment samples with detectable DDX increased through time

223   (Figure 2; Figure S1). DDX was detected in 56%, 72%, 76%, and 90% of samples from 2003, 2008, 2013,

224   and 2018, respectively. Modeling results reflect this, as the year coefficient for the binomial encounter model

225   increased steadily through time (Figure 2; SI Appendix, Table S1) from -0.88 ± 0.87 (mean ± SE) in 2003

226   to 3.15 ± 0.90, 3.76 ± 0.90, and 5.57 ± 0.94 in 2008, 2013, and 2018, respectively. DDX concentrations, on

227   the other hand, were broadly similar across years. Year coefficients for the gamma concentration model

228   were estimated as 1.28 ± 0.50, 1.94 ± 0.50, 1.18 ± 0.50, and 1.62 ± 0.49 for 2003, 2008, 2013, and 2018,

229   respectively.

230   [DDX$_{sed}$] patterns generally showed expected gradients, where concentrations were highest directly

231   off the Palos Verdes Shelf (Figure 2). Los Angeles Harbor and Santa Monica Bay tended to have higher

232   DDX concentrations as well, with concentrations declining moving northward of the Palos Verdes Shelf.

233   The lowest values were found offshore of San Diego. Spatial patterns for the encounter and concentration

234   models were similar, with areas of higher concentrations having larger probabilities of encounter.

235   Uncertainty was highest in deep, offshore locations (Figure 2). These locations were less densely

236   sampled, and therefore prediction confidence was low as spatial interpolation occurred over a broad extent.

237   The largest residuals occurred in ports, bays, and marinas, particularly San Diego Bay (Figure S3; Figure

238   S4). These regions were intensively sampled and spatially heterogeneous, with very high DDX values

239   occurring in close proximity to non-detect samples.

240   **3.3 DDX in fish modeling**

241   All tissue results are reported in lipid-normalized wet weights. Observed concentrations in [DDX$_{fish}$]

242   ranged from 0.0 to 5583.3 ng^{-1}g lipid weight (16.9 ± 460.2 ng^{-1}g lipid weight, mean ± SD). There was a

243   positive linear association between transformed [DDX$_{sed}$] and [DDX$_{fish}$] in the null model with a mean slope

244   of 0.52 and a highest posterior density 80% credible interval of [0.50, 0.54] and an intercept of 1.78 [1.71,

245   1.85] (Figure S8; Figure S9). In all cases, including unique effects by diet, habitat, or species improved

246   model performance compared to applying a single slope and intercept between sediment and fish DDX

247   (Table 1).

248   The relationship between [DDX$_{sed}$] and [DDX$_{fish}$] varied by both diet and habitat (Figure 3; Table 1).

249   The best performing model of [DDX$_{fish}$] included diet, habitat, year, and a random intercept for species

250   (Figure 3; Table 1). Slopes varied substantially between midwater (0.27 [0.19, 0.34]), benthopelagic (0.43

251 [0.36, 0.49]), and benthic (0.52 [0.44, 0.61]) groupings, but were generally less certain and more similar

252 across primary consumer, secondary consumer, and tertiary consumer groups (0.52 [0.39, 0.66], 0.49

253 [0.36, 0.63], 0.42 [0.28, 0.56], respectively). Intercept estimates, on the other hand, varied across all groups.

254 Estimates increased across primary, secondary, and tertiary carnivores (0.14 [-0.30, 0.60], 0.53 [0.07, 1.03],

255 0.89 [0.33, 1.46], respectively) and decreased across midwater, benthopelagic, and benthic groups (0.08

256 [-0.22, 0.41], -0.13 [-0.43, 0.17], -0.13 [-0.51, 0.23], respectively). Based on the inclusion of species as a

257 random effect, the data support that species-specific effects not represented by diet or habitat classification

258 impact fish DDX bioaccumulation. Random intercept estimates ranged from -1.42 [-1.79, -1.07] for market

259 squid to 1.17 [0.59, 1.75] for starry rockfish, but were generally small and near zero (Figure 3). The

260 coefficient on year was negative (-0.056 [-0.06, -0.05]), indicating a decrease in [DDX$_{fish}$] through time that

261 was not explained by changing [DDX$_{sed}$] values.

262

263 **4. Discussion**

264 Despite more than half a century since the cessation of industrial dumping in the SCB, local

265 ecosystem contamination continues to mirror the spatial legacy of contaminant disposal. Thus, the impacts

266 of ocean dumping on marine ecosystems and fisheries resources remain place based, despite the long-

267 held belief that the vastness of the ocean holds nearly limitless capacity for contaminant mitigation via

268 dilution and advection (Lumsdaine 1976). That said, our findings clearly demonstrate that ecological

269 characteristics mediate the strength of linkages between sediment DDX concentrations and the DDX

270 burdens of fishes. Based on our findings, we propose a conceptual model of DDX transport in the coastal

271 ecosystem (Figure 4) where the magnitudes of sediment signatures are highly conserved across space,

272 and trophic ecology and habitat use mediate the vertical transport, lateral mixing, and biomagnification of

273 DDX through the food web. Overarching all these factors, the magnitude of DDX in the SCB ecosystem as

274 represented by fishes decreased over our study period, suggesting that the deleterious effects of these

275 legacy pollutants in the SCB will continue to diminish over time as sediment DDX becomes less

276 bioavailable.

277 **4.1 Sediment DDX is the primary control on DDX burdens for fishes**

278   A linear relationship between transformed [DDX$_{sed}$] and [DDX$_{fish}$] exists within the SCB across two

279   decades and over 60 species of fishes. We provide evidence that local [DDX$_{sed}$] is the primary control on

280   [DDX$_{fish}$] once lipid content of the fish composite has been accounted for and explains nearly half of the

281   total variation in [DDX$_{fish}$]. Results mirror previous, more targeted, studies from the region that found that

282   contaminant concentrations of flatfish were highest near Palos Verdes where sediment concentrations

283   maxima are located (Mearns et al. 1991, McLaughlin et al. 2021) and SCB-wide relationships between

284   sediment contaminant concentrations and flatfish tissue concentrations were highly correlated for both DDX

285   and PCBs (Schiff and Allen 2000, Allen et al. 2002, 2004). This research expands upon previous work and

286   suggests that sediment can serve as a robust predictor of fish organic contamination across the full range

287   of recreationally fished species.

288   The spatial distribution of DDX in sediments reflects historical dumping and ocean transport. The

289   highest concentrations of DDX occurred on the Palos Verdes Shelf (PVS) and immediately upcoast,

290   presumably due to the discharge of roughly 870-1450 tons of DDX onto the PVS between 1947 and 1971

291   (Eganhouse and Pontolillo 2008) and the subsequent transport of these contaminants by ocean currents

292   flowing northward at the discharge depth (Allen et al. 2006). The year coefficient of our encounter model

293   systematically increased through time, likely due to a decrease in MDLs as analytical methods improved

294   (Du et al. 2020), yet we observed no systematic changes in non-zero [DDX$_{sed}$] values through the sampling

295   period. The lack of an observable trend in non-zero [DDX$_{sed}$] values can be attributed to sediment sampling

296   procedures, where a single grab sample consisted of the top 2-5 cm of the sediment column. Sedimentation

297   rates within the region have been recorded at 1-5 mm/ year (Ferré et al. 2010, Kivenson et al. 2019) and

298   natural degradation of DDX is extremely slow, with reported half-lives generally >20 years (Taylor et al.

299   2019, Kurek et al. 2019). Observed [DDX$_{sed}$] values, therefore, represent a time-integrated average that

300   would not be expected to change substantially over a 15-year sample period (Young et al. 1977).

301   Interestingly, although [DDX$_{sed}$] showed limited change through time, [DDX$_{fish}$] showed a marked decrease

302   over the past 20 years, as evidenced by the negative year coefficient (Figure 3). The most likely cause of

303   this discrepancy is a decrease in the bioavailability of DDX to fish and benthic organisms not reflected in

304   bulk [DDX$_{sed}$] values, either due to downward migration of DDX in the sediment column or reduced

305   bioaccessibility due to contaminant aging (Taylor et al. 2019). Although it is possible that changing fish

population characteristics (e.g., fishing down populations to younger individuals) or diets (e.g., where or what fish are eating) resulted in the observed trends, it is unlikely given the consistent declines observed across sampled organisms.

4.2 Ecological traits mediate the relationship between sediment and fish DDX concentrations

Although it's commonly assumed that ecological traits play a consistent role in mediating organism contaminant exposure and bioaccumulation (Windsor et al. 2019), our findings provide evidence that critical factors influencing [DDX_{fish}] are context dependent and contingent on [DDX_{sed}] (Figure 3). In our data, habitat emerged as a more important predictor of [DDX_{fish}] and primarily impacted the slope term in linear model fits, whereas diet primarily modified the intercept term. Small differences in slope result in large differences in [DDX_{fish}] at higher [DDX_{sed}] values, whereas intercept term impacts are highest in low [DDX_{sed}] environments. Therefore, in relatively pristine (low contaminant) environments, the dominant force controlling fish bioaccumulation is trophic magnification. In highly contaminated areas, however, habitat becomes a more important predictor of DDX burdens (Figure 4; Buckman et al. 2021). We posit this occurs due to the relative balance of DDX transport from sediments to fishes (vertical transport), the spatial extent of contamination an organism integrates over (lateral mixing), and biomagnification due to trophic interactions (biomagnification) at a given sediment concentration.

DDX bioaccumulation increased faster with sediment concentrations for demersal species than for midwater and pelagic species residing higher in the water column, as evidenced by the larger [DDX_{sed}] slope term for benthic and benthopelagic fishes. Close correspondence between [DDX_{sed}] and demersal species [DDX_{fish}] can be explained by the hydrophobic nature of DDX. Due to its high octanol water partition coefficient, DDX does not readily dissolve in water and instead binds to suspended particles, organic matter, and sediments (Blais 2005), resulting in sediment concentrations that can be orders of magnitude above water column concentrations (Zeng and Venkatesan 1999, Zeng et al. 2005). Demersal organisms exposed to these higher concentrations may bioaccumulate DDX to a greater extent through direct bioconcentration of DDX (Prince et al. 2021), incidental ingestion of contaminated sediment while feeding (Arnot and Gobas 2004), or consumption of benthic food resources that are high in DDX due to the aforementioned processes (SI Appendix, Table S3). For fish the most likely route of exposure is through diet (Borgå et al. 2004, Wu et

333 al. 2019), and varying correlations of [DDX$_{sed}$] with tissue concentrations across habitat groups is therefore

334 likely reflective of differences in dietary pathways.

335   In addition to diet, the extent of lateral mixing further impacts fish DDX bioaccumulation. Demersal

336 fishes typically occupy a smaller range than pelagic or midwater species, and thus their DDX concentrations

337 are more reflective of local conditions (SI Appendix, Table S3). For this reason, benthic fishes show both

338 the largest slope term and smallest intercept term of any habitat group within our model, as they closely

339 track local sediment conditions. For pelagic species that are characterized by seasonal migrations and

340 extensive spatial movement (Mais 1974, Konno and Wolf 2001, Wolf et al. 2001), the region of capture may

341 not be representative of where they forage and observed DDX concentrations may reflect a region-wide

342 average of water-column DDX concentrations rather than local sediment concentrations.

343   Finally, biomagnification, a central process in ecotoxicology (Walters et al. 2016), was evidenced

344 in our system by larger intercept estimates for higher trophic level organisms (Figure 3). Consistent slope

345 terms across primary, secondary, and tertiary consumers indicates similar rates of biomagnification across

346 [DDX$_{sed}$] gradients. Notably, patterns in biomagnification became evident only after accounting for

347 background sediment contamination. There is a long history of using trophic position as a predictor of

348 organism contamination (Kidd et al. 1995, Jardine et al. 2006, Corsolini and Sarà 2017). Although terrestrial

349 taxa show clear patterns of biomagnification (Kelly et al. 2007), patterns among marine fishes are less clear,

350 particularly when looking over a broad region or multiple species (Gray 2002, Bonito et al. 2016). More

351 localized studies tend to find biomagnification for DDX chemicals, however, exact rates can vary with

352 latitude, chemical constituent and subset of the food web sampled (Walters et al. 2016, Hoondert et al.

353 2020). Recent modeling studies and reviews have examined the impact of variable contaminant exposure

354 on bioaccumulation (Borgå et al. 2012, Kim et al. 2016, Mackay et al. 2016), however, empirical evidence

355 for this process has been limited to date. Our study illustrates the importance of accounting for spatially

356 variable contaminant exposures using empirical data, and results suggest that pooling samples across

357 regions with different background contamination is ill-advised.

358   Recognition of context dependent controls on bioaccumulation is a substantial step forward in

359 understanding of this issue, as previous results examining the relative importance of ecological factors on

360 contaminant bioaccumulation have been mixed. Some studies found that foraging habitat was a stronger

361   predictor of tissue contaminant concentrations, and that the subsequent biomagnification was secondary

362   (Goutte et al. 2015, Yoshino et al. 2020, Carreira et al. 2023) while others found that age and trophic

363   position, as opposed to habitat and carbon source, dominated bioaccumulation (Vander Zanden and

364   Rasmussen 1996, Mcintyre and Beauchamp 2007). Similar to our results, Dromard et al. (2018) posited

365   that biomagnification became a less important pathway of bioaccumulation in more contaminated areas

366   and Fonseca et al. (2019) found that rates of bioaccumulation were similar across sites with variations in

367   biota concentrations reflecting baseline differences in site environmental levels.

368   **4.3 Species-specific characteristics play an observable, if limited, role**

369   [DDX_{sed}], diet, and habitat explained most variation in [DDX_{fish}], however, species-specific factors,

370   reflected in random effects, further impacted [DDX_{fish}] (Table 1). Deviations from group means (i.e., species

371   random effects) could reflect unmodeled aspects of life history such as growth, lifespan, or habitat

372   utilization. For instance, species that grow quickly often exhibit low contaminant concentrations due to

373   growth dilution, wherein fish with high growth efficiency add more tissue per unit contaminant consumed in

374   prey, diluting contaminants in a larger biomass (Karimi et al. 2007, Madenjian et al. 2016). Growth dilution

375   is perhaps evidenced by the large negative anomaly for Market squid (*Doryteuthis opalescens*), a pelagic

376   species that lives only 9 months (Yaremko 2001), indicating lower than expected lipid-normalized DDX

377   concentrations. Our modeling framework also implicitly assumes that fish composites reflect the distribution

378   of sediment contaminants within their zone of capture; however, certain species preferentially use particular

379   habitats within a given fishing zone, decoupling an individual's exposure from the average benthic

380   conditions (Wolfe and Lowe 2015, Burns et al. 2021). For example, due to the documented positive

381   relationship between depth, organic content of sediments, and [DDX_{sed}] within the SCB (SI Appendix, Figure

382   S2), deeper dwelling fish that reside in organic-rich sediments, such as Starry rockfish (*Sebastes

383   constellatus*), may accumulate more contaminants. Proximity to point sources of contamination, such as

384   rivers, estuaries, and sewer outflows, may also increase contaminant burdens of individuals (Ahr et al.

385   2015). Future research could use more specific location-of-capture information in conjunction with

386   movement patterns to optimize sediment exposure calculations (Melwani et al. 2009).

387   Our use of composites prohibited us from discerning how individual-level variation in sex (Johnston

388   et al. 2002, Lyons et al. 2014), diet (Allen 1982, Smith and Smith 2020), or age (Arnot and Gobas 2004)

389 impacted fish DDX bioaccumulation. However, our classification scheme provides a generalizable and

390 accurate framework for predicting DDX burdens in fisheries species as a function of space and ecology.

391 Understanding the interplay between life history, movement, and feeding patterns of an organism can help

392 determine which exposure routes and stressors are likely to be most important to that particular species

393 and should be the focus of future studies.

394 **4.4 Implications of predicting DDX in coastal fisheries and ecosystems**

395   Though bioaccumulation and sediment contamination are closely linked, they are typically

396 assessed separately (Schäfer et al. 2015). Our use of spatially and temporally distributed data from regional

397 monitoring efforts offered a unique opportunity to assess the interplay among local sediment contamination,

398 habitat-use, and biomagnification in driving fish tissue contamination. Our work indicates that sediment can

399 serve as a robust predictor of fish contamination, and general ecological characteristics offer a predictive

400 framework for unmeasured species or locations. Closer integration of ecotoxicology and ecology is

401 essential for predicting contaminant impacts on biological communities and ecosystems (Chapman 2002,

402 Gessner and Tlili 2016). Synthetic chemicals are increasing as or more rapidly than other agents of global

403 change such as biodiversity or habitat loss (Bernhardt et al. 2017), and methods used here could be

404 extended to other hydrophobic, bioaccumulative emerging and legacy pollutants to better inform animal

405 and human exposure. Improved understanding of how regional contamination impacts bioaccumulation in

406 marine organisms provides a framework for assessing potential impacts of the recently discovered DDX

407 dump site, and results can be used to refine consumption advisories or improve monitoring efforts for

408 organisms sensitive to bioaccumulation, such as marine mammals (Randhawa et al. 2015, Gulland et al.

409 2020). Results could also be integrated with California's Sediment Quality Objectives, a series of regulatory

410 targets intended to protect sediment quality in enclosed bays and estuaries, to promote aquatic ecosystems

411 and human health (Bay and Weisberg 2012, Beegan and Bay 2012).

412   Accurate and accessible consumption advisories are particularly pressing for vulnerable

413 communities. Urban anglers in the SCB are a heterogenous and often socioeconomically vulnerable

414 community (Stevenson et al. 2012) who supplement their access to food via coastal fishing in a

415 predominantly urban context (Quimby et al. 2020, Marjadi et al. 2021). The close association of sediment

416 and fish DDX concentrations evident in our results implies that sediment could be used as a first-order

417   proxy for fish contamination in regions without a current monitoring program. Modeling results also indicate

418   that using generalized ecological classifications produces results that are comparable to more detailed,

419   species-specific models (Table 3). Most consumption advice to consumers generally occurs on a species-

420   by-species basis (Oken et al. 2012), and consumption recommendations for unmeasured species can be

421   absent or confusing; a more generalized framework would allow consumption advisories to be extended

422   out to unmeasured species. Lastly, differing patterns across habitat groups have implications for how

423   advisories are created. For example, a region-wide advisory for mobile pelagic species may be sufficient,

424   whereas advisories for benthic fishes need to consider highly localized conditions. This integrated approach

425   enhances our ability to address the complex dynamics of contaminant exposure and bioaccumulation,

426   ultimately contributing to more informed environmental and public health policies.

427   Environmental contaminants are both increasing and diversifying through time, posing substantial

428   threats to human and ecosystem health (Bernhardt et al. 2017). Despite more than half a century since the

429   cessation of industrial dumping in the SCB, the impacts remain prominent. DDX concentrations in marine

430   mammals frequenting the region are among the highest worldwide (Mackintosh et al. 2016) and critically

431   endangered species, such as coastal populations of the California condor, are exposed to levels capable

432   of impacting reproduction (Stack et al. 2022). Our findings, however, suggest spatial and ecological nuance

433   to the DDX ecosystem pathways of toxin transport in the region. Leveraging this nuance should be an

434   important part of efforts to safeguarding both people and the environment against the legacy of ocean

435   dumping.

436

Ocean Monitoring Program for providing the sediment and fish chemistry data used in our analyses. We

445	also thank Max Titcomb for his help in crafting Figure 4 of this manuscript.

References

Ahr, B., M. Farris, and C. G. Lowe. 2015. Habitat selection and utilization of white croaker (Genyonemus lineatus) in the Los Angeles and Long Beach Harbors and the development of predictive habitat use models. Marine Environmental Research 108:1–13.

Allen, L. G., D. J. Pondella, and M. H. Horn, editors. 2006. Ecology of marine fishes: California and adjacent waters. University of California Press, Berkeley.

Allen, M. J. 1982. Functional Structure of Soft-bottom Fish Communities of the Southern California Shelf. University of California, San Diego.

Allen, M. J., A. K. Groce, and J. A. Noblet. 2004. Distribution of contamination above predator-risk guidelines in flatfishes on the Southern California Shelf in 1998. Southern California Coastal Water Research Project, Westminster, CA.

Allen, M. J., S. L. Moore, S. B. Weisberg, A. K. Groce, and M. K. Leecaster. 2002. Comparability of bioaccumulation within the sanddab guild in coastal Southern California. Marine Pollution Bulletin 44:452–458.

Anderson, S. C., E. J. Ward, P. A. English, and L. A. K. Barnett. 2022. sdmTMB: an R package for fast, flexible, and user-friendly generalized linear mixed effects models with spatial and spatiotemporal random fields. preprint, Ecology.

Arnot, J. A., and F. A. P. C. Gobas. 2004. A food web bioaccumulation model for organic chemicals in aquatic ecosystems. Environmental Toxicology and Chemistry 23:2343.

Bay, S. M., and S. B. Weisberg. 2012. Framework for interpreting sediment quality triad data. Integrated Environmental Assessment and Management 8:589–596.

Bay, S. M., E. Y. Zeng, T. D. Lorenson, K. Tran, and C. Alexander. 2003. Temporal and spatial distributions of contaminants in sediments of Santa Monica Bay, California. Marine Environmental Research 56:255–276.

Beegan, C., and S. M. Bay. 2012. Transitioning sediment quality assessment into regulations: Challenges and solutions in implementing California's sediment quality objectives. Integrated Environmental Assessment and Management 8:586–588.

Bernhardt, E. S., E. J. Rosi, and M. O. Gessner. 2017. Synthetic chemicals as agents of global change. Frontiers in Ecology and the Environment 15:84–90.

Blais, J. M. 2005. Biogeochemistry of persistent bioaccumulative toxicants: processes affecting the transport of contaminants to remote areas. Canadian Journal of Fisheries and Aquatic Sciences 62:236–243.

Blasius, M. E., and G. D. Goodmanlowe. 2008. Contaminants still high in top-level carnivores in the Southern California Bight: Levels of DDT and PCBs in resident and transient pinnipeds. Marine Pollution Bulletin 56:1973–1982.

Bonito, L. T., A. Hamdoun, and S. A. Sandin. 2016. Evaluation of the global impacts of mitigation on persistent, bioaccumulative and toxic pollutants in marine fish. PeerJ 4:e1573.

Borgå, K., A. T. Fisk, P. F. Hoekstra, and D. C. G. Muir. 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in Arctic marine food webs. Environmental Toxicology and Chemistry 23:2367.

Borgå, K., K. A. Kidd, D. C. Muir, O. Berglund, J. M. Conder, F. A. Gobas, J. Kucklick, O. Malm, and D. E. Powell. 2012. Trophic magnification factors: Considerations of ecology, ecosystems, and study design. Integrated Environmental Assessment and Management 8:64–84.

Bray, N. A., A. Keyes, and W. M. L. Morawitz. 1999. The California Current system in the Southern California Bight and the Santa Barbara Channel. Journal of Geophysical Research: Oceans 104:7695–7714.

493   Buckman, K. L., R. P. Mason, E. Seelen, V. F. Taylor, P. H. Balcom, J. Chipman, and C. Y. Chen. 2021.
494   Patterns in forage fish mercury concentrations across Northeast US estuaries. Environmental Research
495   194:110629.
496   Bürkner, P.-C. 2017. **brms**: An *R* Package for Bayesian Multilevel Models Using *Stan*. Journal of
497   Statistical Software 80.
498   Burns, E. S., B. W. Wolfe, J. Armstrong, D. Tang, K. Sakamoto, and C. G. Lowe. 2021. Using acoustic
499   telemetry to quantify potential contaminant exposure of Vermilion Rockfish (Sebastes miniatus),
500   Hornyhead Turbot (Pleuronichthys verticalis), and White Croaker (Genyonemus lineatus) at wastewater
501   outfalls in southern California. Marine Environmental Research 170:105452.
502   Carreira, B. M., V. Kolář, E. Chmelová, J. Jan, J. Adašević, A. Landeira-Dabarca, L. Vebrová, M.
503   Poláková, P. Horká, Š. Otáhalová, Z. Musilová, J. Borovec, R. Tropek, and D. S. Boukal. 2023.
504   Bioaccumulation of chemical elements at post-industrial freshwater sites varies predictably between
505   habitats, elements and taxa: A power law approach. Science of The Total Environment 901:165794.
506   Carson, R. 1962. Silent spring. Houghton Mifflin Harcourt, Boston.
507   Chapman, P. M. 2002. Integrating toxicology and ecology: putting the "eco" into ecotoxicology. Marine
508   Pollution Bulletin 44:7–15.
509   Clark, K. E., F. A. P. C. Gobas, and D. Mackay. 1990. Model of organic chemical uptake and clearance
510   by fish from food and water. Environmental Science & Technology 24:1203–1213.
511   Conard, W. M., H. D. Whitehead, K. J. Harris, G. A. Lamberti, G. F. Peaslee, and A. A. Rand. 2022.
512   Maternal Offloading of Per- and Polyfluoroalkyl Substances to Eggs by Lake Michigan Salmonids.
513   Environmental Science & Technology Letters 9:937–942.
514   Corsolini, S., and G. Sarà. 2017. The trophic transfer of persistent pollutants (HCB, DDTs, PCBs) within
515   polar marine food webs. Chemosphere 177:189–199.
516   Dromard, C. R., Y. Bouchon-Navaro, S. Cordonnier, M. Guéné, M. Harmelin-Vivien, and C. Bouchon.
517   2018. Different transfer pathways of an organochlorine pesticide across marine tropical food webs
518   assessed with stable isotope analysis. PLOS ONE 13:e0191335.
519   Du, B., C. Wong, K. McLaughlin, and K. Schiff. 2020. Southern California Bight 2018 Regional Monitoring
520   Program: Volume II. Page Sediment Chemistry (Technical Report No. 1130). Southern California Coastal
521   Water Research Project Costa Mesa, CA.
522   Eganhouse, R. P., and J. Pontolillo. 2008. DDE in Sediments of the Palos Verdes Shelf, California: *In Situ*
523   Transformation Rates and Geochemical Fate. Environmental Science & Technology 42:6392–6398.
524   Ferré, B., C. R. Sherwood, and P. L. Wiberg. 2010. Sediment transport on the Palos Verdes shelf,
525   California. Continental Shelf Research 30:761–780.
526   Fonseca, V. F., S. França, B. Duarte, I. Caçador, H. N. Cabral, C. L. Mieiro, J. P. Coelho, E. Pereira, and
527   P. Reis-Santos. 2019. Spatial Variation in Mercury Bioaccumulation and Magnification in a Temperate
528   Estuarine Food Web. Frontiers in Marine Science 6:117.
529   Gessner, M. O., and A. Tlili. 2016. Fostering integration of freshwater ecology with ecotoxicology.
530   Freshwater Biology 61:1991–2001.
531   Goutte, A., Y. Cherel, C. Churlaud, J.-P. Ponthus, G. Massé, and P. Bustamante. 2015. Trace elements
532   in Antarctic fish species and the influence of foraging habitats and dietary habits on mercury levels.
533   Science of The Total Environment 538:743–749.
534   Gray, J. S. 2002. Biomagnification in marine systems: the perspective of an ecologist. Marine Pollution
535   Bulletin 45:46–52.
536   Gulland, F. M. D., A. J. Hall, G. M. Ylitalo, K. M. Colegrove, T. Norris, P. J. Duignan, B. Halaska, K.
537   Acevedo Whitehouse, L. J. Lowenstine, A. C. Deming, and T. K. Rowles. 2020. Persistent Contaminants
538   and Herpesvirus OtHV1 Are Positively Associated With Cancer in Wild California Sea Lions (Zalophus
539   californianus). Frontiers in Marine Science 7:602565.
540   Hebert, C. E., and K. A. Keenleyside. 1995. To normalize or not to normalize? Fat is the question.
541   Environmental Toxicology and Chemistry 14:801–807.

542 Hickey, B. M. 1992. Circulation over the Santa Monica-San Pedro Basin and Shelf. Progress in
543 Oceanography 30:37–115.
544 Hoffman, M. D., and A. Gelman. 2014. The No-U-Turn Sampler: Adaptively Setting Path Lengths in
545 Hamiltonian Monte Carlo. Journal of Machine Learning Research 15:1593–1623.
546 Hoondert, R. P. J., N. W. Van Den Brink, M. J. Van Den Heuvel-Greve, AdM. J. Ragas, and A. Jan
547 Hendriks. 2020. Implications of Trophic Variability for Modeling Biomagnification of POPs in Marine Food
548 Webs in the Svalbard Archipelago. Environmental Science & Technology 54:4026–4035.
549 Jardine, T. D., K. A. Kidd, and A. T. Fisk. 2006. Applications, Considerations, and Sources of Uncertainty
550 When Using Stable Isotope Analysis in Ecotoxicology. Environmental Science & Technology 40:7501–
551 7511.
552 Jarvis, E., K. Schiff, L. Sabin, and M. J. Allen. 2007. Chlorinated hydrocarbons in pelagic forage fishes
553 and squid of the Southern California Bight. Environmental Toxicology and Chemistry 26:2290.
554 Johnston, T. A., A. T. Fisk, D. M. Whittle, and D. C. G. Muir. 2002. Variation in Organochlorine
555 Bioaccumulation by a Predatory Fish; Gender, Geography, and Data Analysis Methods. Environmental
556 Science & Technology 36:4238–4244.
557 Karimi, R., C. Y. Chen, P. C. Pickhardt, N. S. Fisher, and C. L. Folt. 2007. Stoichiometric controls of
558 mercury dilution by growth. Proceedings of the National Academy of Sciences 104:7477–7482.
559 Kelly, B. C., M. G. Ikonomou, J. D. Blair, A. E. Morin, and F. A. P. C. Gobas. 2007. Food WebSpecific
560 Biomagnification of Persistent Organic Pollutants. Science 317:236–239.
561 Kidd, K. A., H. A. Bootsma, R. H. Hesslein, D. C. G. Muir, and R. E. Hecky. 2001. Biomagnification of
562 DDT through the Benthic and Pelagic Food Webs of Lake Malawi, East Africa: Importance of Trophic
563 Level and Carbon Source. Environmental Science & Technology 35:14–20.
564 Kidd, K. A., R. H. Hesslein, B. J. Ross, K. Koczanski, G. R. Stephens, and D. C. G. Muir. 1998.
565 Bioaccumulation of organochlorines through a remote freshwater food web in the Canadian Arctic.
566 Environmental Pollution 102:91–103.
567 Kidd, K. A., D. W. Schindler, D. C. G. Muir, W. L. Lockhart, and R. H. Hesslein. 1995. High
568 Concentrations of Toxaphene in Fishes from a Subarctic Lake. Science 269:240–242.
569 Kim, J., F. A. P. C. Gobas, J. A. Arnot, D. E. Powell, R. M. Seston, and K. B. Woodburn. 2016. Evaluating
570 the roles of biotransformation, spatial concentration differences, organism home range, and field sampling
571 design on trophic magnification factors. Science of The Total Environment 551–552:438–451.
572 Kivenson, V., K. L. Lemkau, O. Pizarro, D. R. Yoerger, C. Kaiser, R. K. Nelson, C. Carmichael, B. G.
573 Paul, C. M. Reddy, and D. L. Valentine. 2019. Ocean Dumping of Containerized DDT Waste Was a
574 Sloppy Process. Environmental Science & Technology 53:2971–2980.
575 Konno, E. S., and P. Wolf. 2001. Pacific mackerel. Pages 306–308 *in* W. S. Leet, C. M. Dewees, R.
576 Klingbeil, and E. J. Larson, editors. California's Living Marine Resources: A Status Report. California
577 Department of Fish and Game, Sacramento, CA.
578 Kristensen, K., A. Nielsen, C. W. Berg, H. Skaug, and B. M. Bell. 2016. **TMB** : Automatic Differentiation
579 and Laplace Approximation. Journal of Statistical Software 70.
580 Kurek, J., P. W. MacKeigan, S. Veinot, A. Mercer, and K. A. Kidd. 2019. Ecological Legacy of DDT
581 Archived in Lake Sediments from Eastern Canada. Environmental Science & Technology 53:7316–7325.
582 Lumsdaine, J. A. 1976. Ocean Dumping Regulation: An Overview. Ecology Law Quarterly 5:753–792.
583 Lyons, K., R. Lavado, D. Schlenk, and C. G. Lowe. 2014. Bioaccumulation of organochlorine
584 contaminants and ethoxyresorufin- *o* -deethylase activity in southern California round stingrays (*Urobatis*
585 *halleri*) exposed to planar aromatic compounds: Bioaccumulation of organochlorines and EROD in *U.*
586 *halleri*. Environmental Toxicology and Chemistry 33:1380–1390.
587 Mackay, D., A. K. D. Celsie, J. A. Arnot, and D. E. Powell. 2016. Processes influencing chemical
588 biomagnification and trophic magnification factors in aquatic ecosystems: Implications for chemical
589 hazard and risk assessment. Chemosphere 154:99–108.

Mackintosh, S. A., N. G. Dodder, N. J. Shaul, L. I. Aluwihare, K. A. Maruya, S. J. Chivers, K. Danil, D. W. Weller, and E. Hoh. 2016. Newly Identified DDT-Related Compounds Accumulating in Southern California Bottlenose Dolphins. Environmental Science & Technology 50:12129–12137.

Madenjian, C. P., S. R. Carpenter, and P. S. Rand. 1994. Why Are the PCB Concentrations of Salmonine Individuals from the Same Lake So Highly Variable? Canadian Journal of Fisheries and Aquatic Sciences 51:800–807.

Madenjian, C. P., O. P. Jensen, R. R. Rediske, J. P. O'Keefe, A. R. Vastano, and S. A. Pothoven. 2016. Differences in Energy Expenditures and Growth Dilution Explain Higher PCB Concentrations in Male Summer Flounder. PLOS ONE 11:e0147223.

Mais, K. F. 1974. Pelagic Fish Surveys In The California Current. California Department of Fish and Game, Sacramento, CA.

Marjadi, M. N., L. Drakopulos, L. W. Guo, J. Z. Koehn, S. V. Panchang, and D. Robertson. 2021. Negative socio-environmental feedback loop may foster inequality for urban marine subsistence fishers. Environmental Science & Policy 121:68–77.

Mcintyre, J., and D. Beauchamp. 2007. Age and trophic position dominate bioaccumulation of mercury and organochlorines in the food web of Lake Washington. Science of The Total Environment 372:571–584.

McLaughlin, K., J. Davis, A. Bonnema, B. Du, G. Ichikawa, W. Jakl, W. Heim, and K. Schiff. 2021. Regional assessment of contaminant bioaccumulation in sport fish tissue in the Southern California Bight, USA. Marine Pollution Bulletin 172:112798.

McLaughlin, K., K. Schiff, B. Du, J. Davis, A. Bonnema, G. Ichikawa, B. Jakl, and W. Heim. 2020. Southern California Bight 2018 Regional Monitoring Program: Volume V. Contaminant Bioaccumulation in Edible Sport Fish Tissue. Technical Report 1155. Southern California Coastal Water Research Project ….

Mearns, A. J., M. Matta, G. Shigenaka, D. MacDonald, M. Buchman, H. Harris, J. Golas, and G. Lauenstein. 1991. Contaminant Trends in the Southern California Bight: Inventory and Assessment.

Melwani, A. R., B. K. Greenfield, and E. R. Byron. 2009. Empirical Estimation of Biota Exposure Range for Calculation of Bioaccumulation Parameters. Integrated Environmental Assessment and Management 5:138.

Oken, E., A. L. Choi, M. R. Karagas, K. Mariën, C. M. Rheinberger, R. Schoeny, E. Sunderland, and S. Korrick. 2012. Which Fish Should I Eat? Perspectives Influencing Fish Consumption Choices. Environmental Health Perspectives 120:790–798.

Pastor, D., J. Boix, V. Fernández, and J. Albaigés. 1996. Bioaccumulation of organochlorinated contaminants in three estuarine fish species (Mullus barbatus, Mugil cephalus and Dicentrarcus labrax). Marine Pollution Bulletin 32:257–262.

Paterson, G., K. G. Drouillard, and G. D. Haffner. 2006. An evaluation of stable nitrogen isotopes and polychlorinated biphenyls as bioenergetic tracers in aquatic systems. Canadian Journal of Fisheries and Aquatic Sciences 63:628–641.

Pitchon, A., and K. Norman. 2012. Fishing off the Dock and Under the Radar in Los Angeles County: Demographics and Risks. Bulletin, Southern California Academy of Sciences 111:141–152.

Prince, K. D., S. M. Crotty, A. Cetta, J. J. Delfino, T. M. Palmer, N. D. Denslow, and C. Angelini. 2021. Mussels drive polychlorinated biphenyl (PCB) biomagnification in a coastal food web. Scientific Reports 11:9180.

Quimby, B., S. Es. Crook, K. M. Miller, J. Ruiz, and D. Lopez-Carr. 2020. Identifying, defining and exploring angling as urban subsistence: Pier fishing in Santa Barbara, California. Marine Policy 121:104197.

Randhawa, N., F. Gulland, G. M. Ylitalo, R. DeLong, and J. A. K. Mazet. 2015. Sentinel California sea lions provide insight into legacy organochlorine exposure trends and their association with cancer and infectious disease. One Health 1:37–43.

638   Rue, H., S. Martino, and N. Chopin. 2009. Approximate Bayesian inference for latent Gaussian models by
639   using integrated nested Laplace approximations. Journal of the Royal Statistical Society: Series B
640   (Statistical Methodology) 71:319–392.
641   Schäfer, S., C. Antoni, C. Möhlenkamp, E. Claus, G. Reifferscheid, P. Heininger, and P. Mayer. 2015.
642   Equilibrium sampling of polychlorinated biphenyls in River Elbe sediments – Linking bioaccumulation in
643   fish to sediment contamination. Chemosphere 138:856–862.
644   Schiff, K., and M. J. Allen. 2000. Chlorinated hydrocarbons in flatfishes from the Southern California,
645   USA, Bight. Environmental Toxicology and Chemistry 19:1559–1565.
646   Schiff, K., D. Greenstein, N. Dodder, and D. J. Gillett. 2016. Southern California Bight regional monitoring.
647   Regional Studies in Marine Science 4:34–46.
648   Schnaker, Z., J. Anderson, and J. Hilger. 2015. Recreational Fisheries on the West Coast. NOAA
649   Fisheries.
650   Smith, B. E., and L. A. Smith. 2020. Multispecies functional responses reveal reduced predation at high
651   prey densities and varied responses among and within trophic groups. Fish and Fisheries 21:891–905.
652   Smokler, P. E., D. R. Young, and K. L. Gard. 1979. DDTs in marine fishes following termination of
653   dominant California input: 1970–77. Marine Pollution Bulletin 10:331–334.
654   Stack, M. E., J. M. Cossaboon, C. W. Tubbs, L. I. Vilchis, R. G. Felton, J. L. Johnson, K. Danil, G. Heckel,
655   E. Hoh, and N. G. Dodder. 2022. Assessing Marine Endocrine-Disrupting Chemicals in the Critically
656   Endangered California Condor: Implications for Reintroduction to Coastal Environments. Environmental
657   Science & Technology 56:7800–7809.
658   Stan Development Team. 2023. RStan: the R interface to Stan.
659   Stevenson, C., S. A. Sikich, and M. Gold. 2012. Engaging Los Angeles County subsistence anglers in the
660   California marine protected area planning process. Marine Policy 36:559–563.
661   Taylor, A. R., J. Wang, C. Liao, D. Schlenk, and J. Gan. 2019. Effect of aging on bioaccessibility of DDTs
662   and PCBs in marine sediment. Environmental Pollution 245:582–589.
663   Vander Zanden, M. J., and J. B. Rasmussen. 1996. A Trophic Position Model of Pelagic Food Webs:
664   Impact on Contaminant Bioaccumulation in Lake Trout. Ecological Monographs 66:451–477.
665   Vehtari, A., A. Gelman, and J. Gabry. 2017. Practical Bayesian model evaluation using leave-one-out
666   cross-validation and WAIC. Statistics and Computing 27:1413–1432.
667   Venkatesan, M. I., G. E. Greene, E. Ruth, and A. B. Chartrand. 1996. DDTs and dumpsite in the Santa
668   Monica Basin, California. Science of The Total Environment 179:61–71.
669   Walters, D. M., T. D. Jardine, B. S. Cade, K. A. Kidd, D. C. G. Muir, and P. Leipzig-Scott. 2016. Trophic
670   Magnification of Organic Chemicals: A Global Synthesis. Environmental Science & Technology 50:4650–
671   4658.
672   Wang, X., and W.-X. Wang. 2005. Uptake, absorption efficiency and elimination of DDT in marine
673   phytoplankton, copepods and fish. Environmental Pollution 136:453–464.
674   Wilson, N. D., P. S. Price, and D. J. Paustenbach. 2001. An event-by-event probabilistic methodology for
675   assessing the health risks of persistent chemicals in fish: a case study at the Palos Verdes Shelf. Journal
676   of Toxicology and Environmental Health, Part A 62:595–642.
677   Windsor, F. M., M. G. Pereira, C. R. Tyler, and S. J. Ormerod. 2019. Biological Traits and the Transfer of
678   Persistent Organic Pollutants through River Food Webs. Environmental Science & Technology 53:13246–
679   13256.
680   Wolf, P., P. E. Smith, and D. R. Bergen. 2001. Pacific sardine. Pages 299–302 *in* W. S. Leet, C. M.
681   Dewees, R. Klingbeil, and E. J. Larson, editors. California's Living Marine Resources: A Status Report.
682   California Department of Fish and Game, Sacramento, CA.
683   Wolfe, B. W., and C. G. Lowe. 2015. Movement patterns, habitat use and site fidelity of the white croaker
684   (Genyonemus lineatus) in the Palos Verdes Superfund Site, Los Angeles, California. Marine
685   Environmental Research 109:69–80.

686 Wu, P., M. J. Kainz, A. G. Bravo, S. Åkerblom, L. Sonesten, and K. Bishop. 2019. The importance of
687 bioconcentration into the pelagic food web base for methylmercury biomagnification: A meta-analysis.
688 Science of The Total Environment 646:357–367.
689 Yaremko, M. 2001. Market squid. Pages 295–298 *in* W. S. Leet, C. M. Dewees, R. Klingbeil, and E. J.
690 Larson, editors. California's Living Marine Resources: A Status Report. California Department of Fish and
691 Game, Sacramento, CA.
692 Yoshino, K., K. Mori, G. Kanaya, S. Kojima, Y. Henmi, A. Matsuyama, and M. Yamamoto. 2020. Food
693 sources are more important than biomagnification on mercury bioaccumulation in marine fishes.
694 Environmental Pollution 262:113982.
695 Young, D. R., D. McDermott-Ehrlich, and T. C. Heesen. 1977. Sediments as sources of DDT and PCB.
696 Marine Pollution Bulletin 8:254–257.
697 Zeng, E. Y., D. Tsukada, D. W. Diehl, J. Peng, K. Schiff, J. A. Noblet, and K. A. Maruya. 2005.
698 Distribution and Mass Inventory of Total Dichlorodiphenyldichloroethylene in the Water Column of the
699 Southern California Bight. Environmental Science & Technology 39:8170–8176.
700 Zeng, E. Y., and M. I. Venkatesan. 1999. Dispersion of sediment DDTs in the coastal ocean off southern
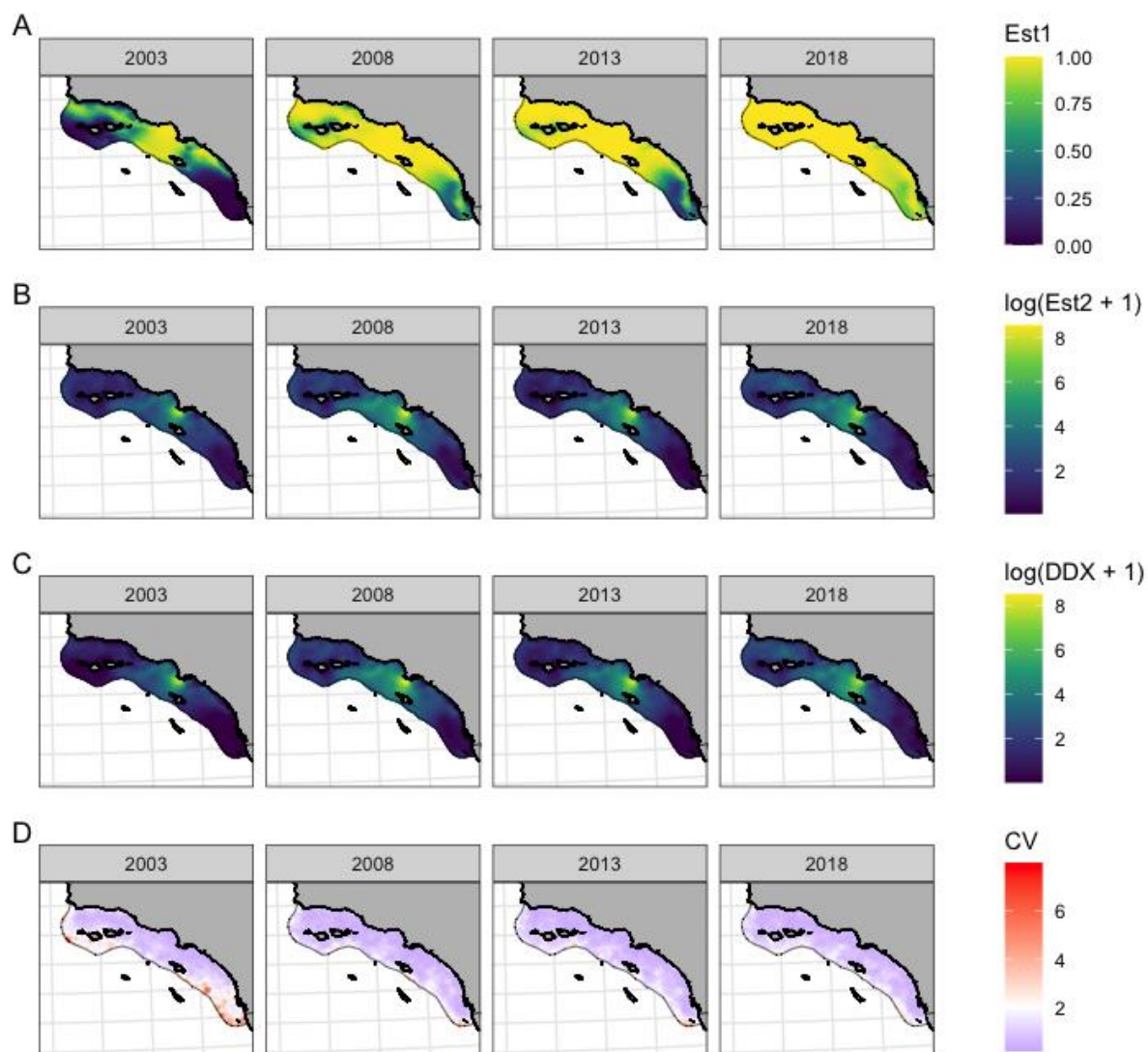701 California. Science of The Total Environment 229:195–208.
702

703
704
705
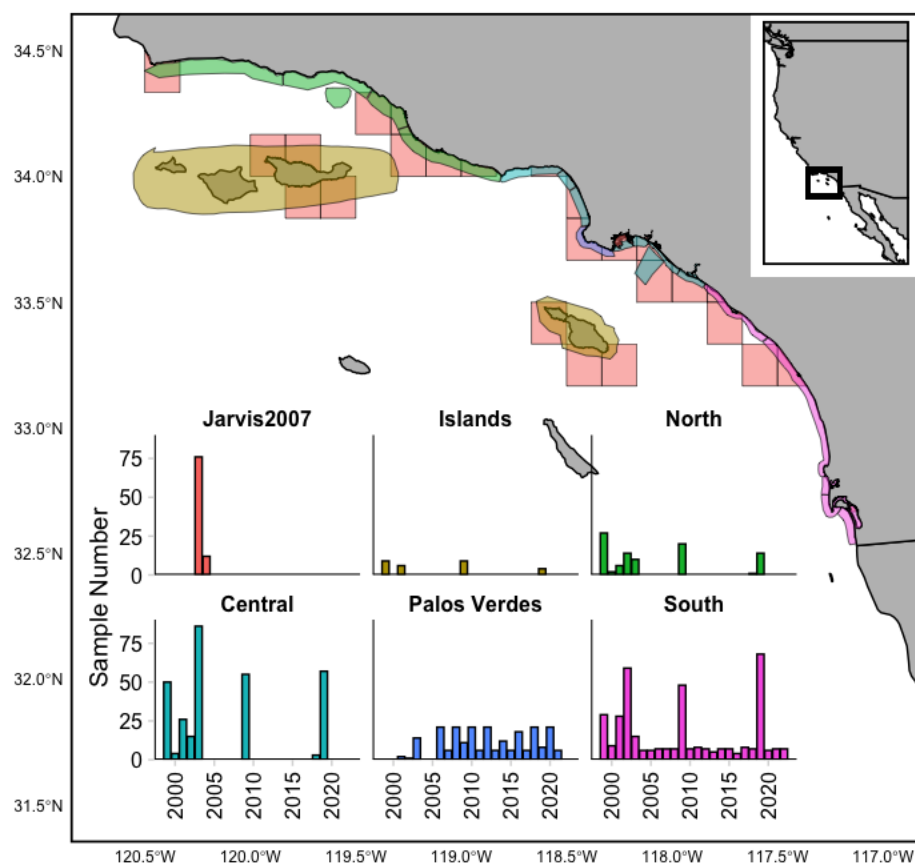
Figure 1. Map of fishing zones and the number of fish samples through time, by region (inset). Nearshore polygons are derived from McLaughlin et al. (2021) and pink blocks are California Department of Fish and Game 256 km² fishing blocks.

710
711

712
713 **Figure 2.** Sediment spatiotemporal model results by year showing the probability of detection via the
714 binomial presence-absence model (Est1) (A), total DDX estimates from the gamma positive-value model
715 (Est2) (B), total estimated DDX concentrations from both models in ng/g dw (C), and the coefficient of
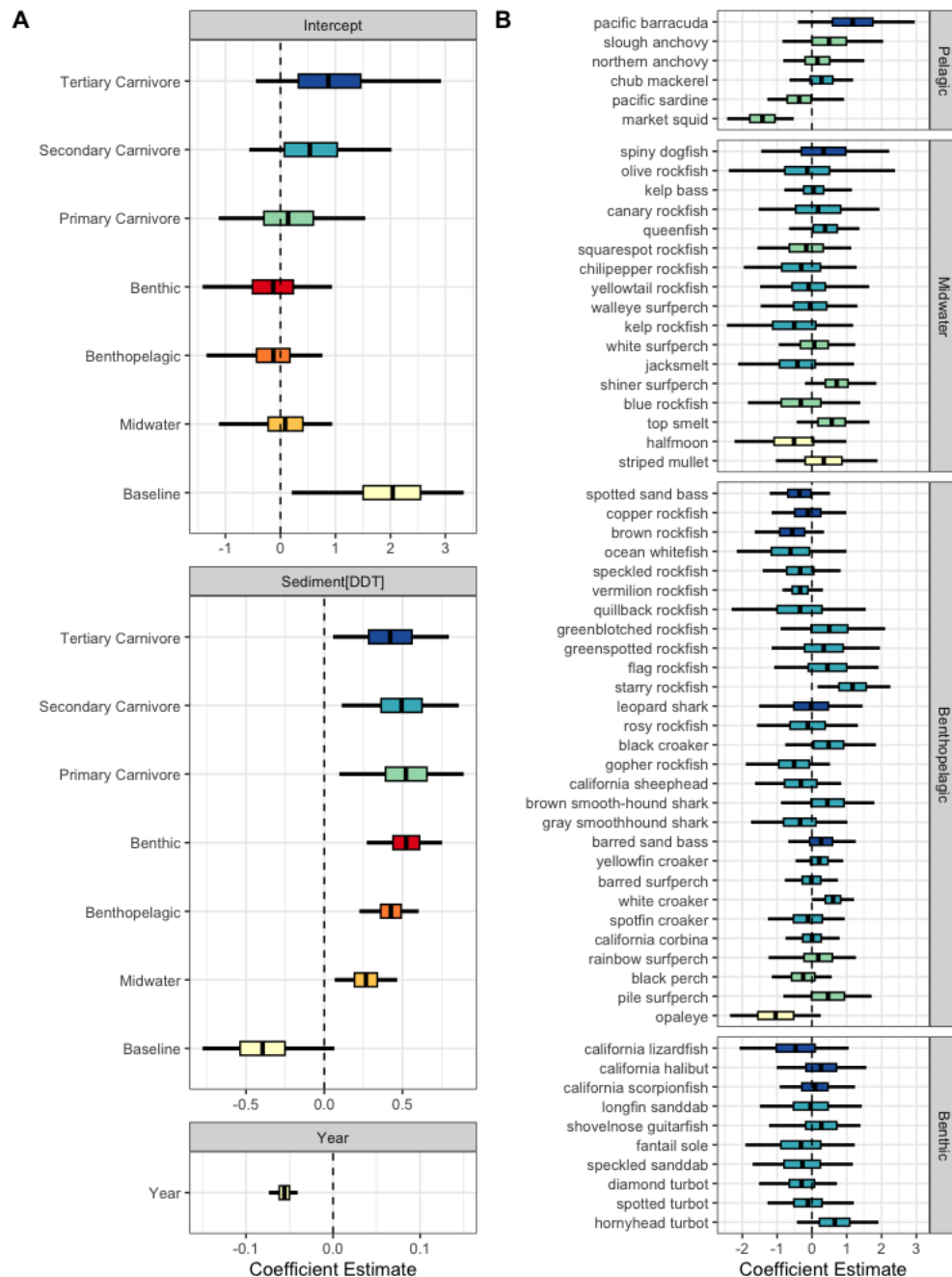716 variation on predictions (D).
717

Figure 3. Model estimated posterior distributions for fixed (A) and random (B) effects from the diet-habitat-species-year model (Table 1). The reference category is herbivore and pelagic. Center lines 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. Random effects (B) can be thought of as deviations from the group means (habitat x diet). Color indicates diet categorization, with darker colors indicating higher trophic levels, and horizontal lines denote habitat classification. Within each habitat category, species are arranged by the estimated FishBase trophic level.
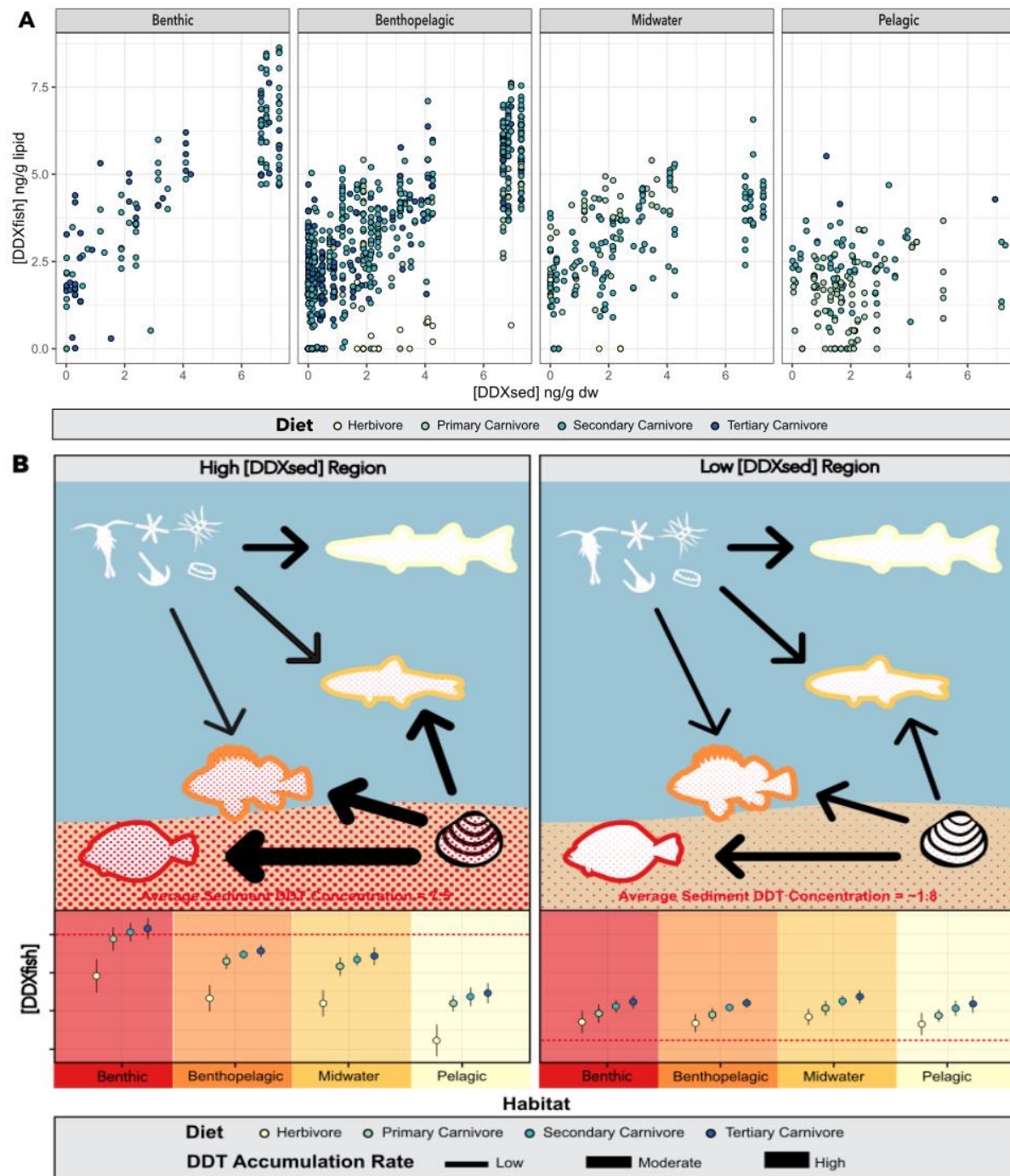
729
Figure 4. (A) Transformed [DDX$_{sed}$] data for each fishing zone versus [DDX$_{fish}$] values, separated by diet
(color) and habitat (position), and (B) a conceptual figure of bioaccumulation in fishes within the SCB,
illustrating that for high [DDX$_{sed}$] values habitat is the most important determinant, whereas at low
[DDX$_{sed}$] values diet becomes more important. Also shown is the 80% posterior predictive distribution for
the mean [DDX$_{sed}$] of a highly contaminated region ([DDX$_{sed}$] = 6.67 ng/g dw) and a pristine region
([DDX$_{sed}$] = 0.42 ng/g dw). The horizontal red line in (B) indicates the [DDX$_{sed}$] value used to generate
predictions.

738   **Table 1.** Results of model selection for fish and sediment linear and mixed-effects models, showing

739   model name, structure, LOOIC (leave-one-out information criterion), and elpd$_{loo}$. LOOIC describes the

740   support of each candidate model where lower values indicate better models and the difference elpd$_{loo}$ was

741   used to compare model predictive capacity relative to the best model. For all models, [DDX$_{fish}$] was

742   treated as a left-censored variable, where values were constrained to fall between zero and the

743   composite-specific MDL.

744

Question	Model Name	Fixed effects	Random effects	LOOIC (SE)	elpd$_{loo}$ (SE)	R^2 (Gelman et al. 2019)
1. What is the global relationship between [DDX$_{sed}$] and [DDX$_{fish}$]?	Null	[DDX$_{sed}$]		3642.4 (57.3)	-350.1 (25.5)	0.51
2. How do ecological factors mediate the relationship between [DDX$_{sed}$] and [DDX$_{fish}$]?	Diet (intercept only)	[DDX$_{sed}$] + Diet		3509.4 (55.6)	-283.6 (22.3)	0.57
	Diet (slope only)	[DDX$_{sed}$] + [DDX$_{sed}$] :Diet		3482.2 (54.9)	-270.0 (21.7)	0.57
	Diet	[DDX$_{sed}$] *Diet		3475.1 (55.3)	-266.5 (21.7)	0.58
	Habitat (intercept only)	[DDX$_{sed}$] + Habitat		3488.5 (57.8)	-273.2 (23.9)	0.57
	Habitat (slope only)	[DDX$_{sed}$] + [DDX$_{sed}$] :Habitat		3413.5 (58.7)	-235.7 (23.5)	0.59
	Habitat	[DDX$_{sed}$]*Habitat		3409.7 (58.9)	-233.7 (23.4)	0.59

3. Do additional factors improve prediction of [DDX$_{fish}$]?	Diet-Habitat	[DDX$_{sed}$] *Diet + [DDX$_{sed}$] *Habitat		3253.0 (55.3)	-155.4 (16.7)	0.65
	Diet-Habitat-Year	[DDX$_{sed}$] *Diet + [DDX$_{sed}$] *Habitat + Year		3118.6 (54.7)	-88.2 (13.7)	0.69
	Diet-Habitat-Species	[DDX$_{sed}$] *Diet + [DDX$_{sed}$] *Habitat	1 | Species	3099.7 (61.5)	- 78.8 (12.8)	0.71
	Diet-Habitat-Species-Year	[DDX$_{sed}$] *Diet + [DDX$_{sed}$] *Habitat + Year	1 | Species	2942.2 (58.4)	-	0.74

745