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

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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 km² 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,

we aim to lay the groundwork for predictive models of exposure that are spatially explicit, but generalizable across taxa with shared ecologies. Moreover, our findings will link the long legacy of regional ocean dumping to contemporary ocean ecosystem processes and fisheries of southern California.

2. Materials and Methods

2.1 Datasets

The area of focus of our study is the Southern California Bight (SCB; Figure 1). Extending more than 600 km from the United States – Mexico border northwards to Point Conception, California, the SCB is a dynamic and productive region where the cold, southward flowing California Current mixes with the warm, northward flowing Davidson Countercurrent (Hickey 1992, Bray et al. 1999). The SCB is among the most biodiverse of all Northern Hemisphere temperate coastal ecosystems and one of the most densely populated coastal regions in the country, home to the nation's largest commercial port, one of the largest US Naval complexes, and over 20 million people. Recreational marine fisheries in the region comprise a multibillion-dollar industry that is economically, socially, and culturally important (Pitchon and Norman 2012, Schnaker et al. 2015).

We obtained the sediment and fish data used in this study from nine primary sources collected between 1998 and 2021 (SI Appendix, Table S1). All laboratories that analyzed the sediment and fish data were subject to a common set of rigorous quality assurance and quality control (QA/QC) guidelines to ensure comparability (Du et al. 2020, McLaughlin et al. 2020). Sediment samples were collected via grab samples of the top 5 cm of sediment at embayment sites and the top 2 cm at offshore sites between July 1 and September 30 in 2003, 2008, 2013, and 2018 (SI Appendix, Figure S1; Schiff et al. 2016, Du et al. 2020). Sites where concentrations were below detection limits were reported as zero values. For fish tissue, samples were collected off piers and boats and saved for analysis as either muscle tissue filet with the skin off (for large species) or whole fish without the head, tail, or internal organs (for small species). Generally, fish samples were composites of 5-10 specimens, depending on the species and monitoring program. We included only single-species composites in our analysis and subset available data to include only sediment and fish samples that explicitly measured 2,4'-DDE, 4,4'-DDE, 2,4'-DDD, 4,4'-DDD, 2,4'-DDT, and 4,4'-

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

We assigned each species represented in our aggregated dataset diet and habitat classifications according to their adult life history characteristics (SI Appendix, Table S3). Diet categories included herbivores, primary carnivores, secondary carnivores, and tertiary carnivores and habitat categories included pelagic, midwater, and demersal with the demersal group sub-divided again into benthic (species that rest on the ocean floor) and benthopelagic (species found in the water just above the ocean floor). Each fish composite was assigned to one of 27 spatially explicit fishing zones following McLaughlin et al. (2021), however, composites from Jarvis et al. (2007) were assigned to one of 25 spatially explicit California Department of Fish and Game 256 km² fishing blocks. Fishing zones and fishing blocks were combined into a common spatial array hereafter referred to as fishing zones. Assignment to fishing zones allowed us to approximate fish contaminant exposure when the exact capture location was unknown.

2.2 DDX in sediment modeling

For each fishing zone, we generated a time varying value for average sediment DDX concentrations, [DDX_{sed}]. The goal with this analysis was not to determine environmental factors driving sediment DDX concentrations, but rather to generate zone-averages to be used as covariates in a model of fish bioaccumulation. To generate continuous spatial estimates for [DDX_{sed}] we fit spatiotemporal regression models to sediment DDX measurements. The underlying statistical model was a spatiotemporal generalized linear mixed-effects model with Gaussian random fields to model spatiotemporal processes. Because concentrations were positive, skewed, and with frequent zero observations, we assume the data were observed with a delta-gamma distribution (Lo et al. 1992, Stefansson 1996, Martin et al. 2005). The delta-gamma, or zero-modified gamma, distribution is the mixture of a gamma distribution with a positive probability mass at zero that separately models the probability of having non-zero values and positive values for each sampling event. It consists of a binomial presence-absence model and a model for positive values only with a gamma observation distribution and a log link.

We compared eight alternative models. Candidate models included a combination of spatial effects, spatiotemporal effects (first-order autoregressive spatiotemporal random fields to account for correlation from one time step to the next), year effects (as factors), and depth (modeled as a smooth function). We

used a flexible spatial modeling approach as we expected $[DDX_{sed}]$ to be patchy due to an uneven legacy of dumping and the distribution of organic material within sediment, which covaries with depth within the SCB (SI Appendix, Figure S2; Schiff et al. 2016). We fit our models using the 'sdmTMB' R package (Anderson et al. 2022), which makes use of INLA (Rue et al. 2009) to generate stochastic partial differential equation matrices and TMB (Kristensen et al. 2016) to calculate the log likelihood, gradient of the log likelihood and implementation of the Laplace approximation. Candidate models were compared via ΔAIC (Akaike information criteria) and five-fold-cross-validation (SI Appendix, Table S1). The best-fit model was subsequently used to predict average $[DDX_{sed}]$ for each fishing zone corresponding to the four time periods captured by both sediment and fish data: 1998-2005, 2005-2010, 2010-2015, and 2015-2020.

2.3 DDX in fish modeling

To understand the extent of coupling between sediment and fish DDX concentrations, we fit 11 candidate Bayesian linear and linear mixed-effects models to $[DDX_{sed}]$ and lipid-normalized fish DDX concentrations, $[DDX_{fish}]$ (Table 1). We chose to use lipid normalized concentrations due to the well documented, positive relationship between lipid content and organic contaminant concentration in fish tissue (Hebert and Keenleyside 1995). Prior to analysis, fish and sediment values were transformed as $\log_{10}(x+1)$, where x is the DDX concentration, to account for right-skewness and zero values. In all models, $[DDX_{sed}]$ was treated as a continuous variable assuming normal errors. $[DDX_{fish}]$ was treated as a continuous, left censored variable where non-detect values were constrained to fall between zero and the Method Detection Limit (MDL). As MDLs were variable across analytes and monitoring programs, the minimum MDL across all six DDX analytes was selected for each fish composite.

First, to understand the mean relationship between $[DDX_{sed}]$ and $[DDX_{fish}]$, we included $[DDX_{sed}]$ as a fixed effect with no hierarchical groupings. Next, to understand if and how fish ecological traits mediate this relationship, six additional models were fit to the same dataset. The first set examined the impact of fish diet on $[DDX_{fish}]$ values and consisted of three individual models. The first included the interaction between $[DDX_{sed}]$ and fish diet as a categorical variable (slope and intercept estimated for each group), the second included fish diet as a fixed effect (intercept only), and the third included only the interaction between $[DDX_{sed}]$ and fish diet (slope only). The second set of models took the same form but examined the 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 (Cauchy distribution with a mean of 0 and variance of 2) and fixed effects (Cauchy 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 ($56.72 \pm 331.4 \mu g^{-1}g^{-1}$ dw, mean \pm SD). The proportion of sediment samples with detectable DDX increased through time

(Figure 2; Figure S1). DDX was detected in 56%, 72%, 76%, and 90% of samples from 2003, 2008, 2013, and 2018, respectively. Modeling results reflect this, as the year coefficient for the binomial encounter model increased steadily through time (Figure 2; SI Appendix, Table S1) from -0.88 ± 0.87 (mean \pm SE) in 2003 to 3.15 ± 0.90 , 3.76 ± 0.90 , and 5.57 ± 0.94 in 2008, 2013, and 2018, respectively. DDX concentrations, on the other hand, were broadly similar across years. Year coefficients for the gamma concentration model 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, respectively.

[DDX_{sed}] patterns generally showed expected gradients, where concentrations were highest directly off the Palos Verdes Shelf (Figure 2). Los Angeles Harbor and Santa Monica Bay tended to have higher DDX concentrations as well, with concentrations declining moving northward of the Palos Verdes Shelf. The lowest values were found offshore of San Diego. Spatial patterns for the encounter and concentration models were similar, with areas of higher concentrations having larger probabilities of encounter.

Uncertainty was highest in deep, offshore locations (Figure 2). These locations were less densely sampled, and therefore prediction confidence was low as spatial interpolation occurred over a broad extent. The largest residuals occurred in ports, bays, and marinas, particularly San Diego Bay (Figure S3; Figure S4). These regions were intensively sampled and spatially heterogeneous, with very high DDX values occurring in close proximity to non-detect samples.

3.3 DDX in fish modeling

All tissue results are reported in lipid-normalized wet weights. Observed concentrations in [DDX_{fish}] ranged from 0.0 to 5583.3 ng⁻¹g lipid weight (16.9 ± 460.2 ng⁻¹g lipid weight, mean \pm SD). There was a positive linear association between transformed [DDX_{sed}] and [DDX_{fish}] in the null model with a mean slope of 0.52 and a highest posterior density 80% credible interval of [0.50, 0.54] and an intercept of 1.78 [1.71, 1.85] (Figure S8; Figure S9). In all cases, including unique effects by diet, habitat, or species improved model performance compared to applying a single slope and intercept between sediment and fish DDX (Table 1).

The relationship between [DDX_{sed}] and [DDX_{fish}] varied by both diet and habitat (Figure 3; Table 1). The best performing model of [DDX_{fish}] included diet, habitat, year, and a random intercept for species (Figure 3; Table 1). Slopes varied substantially between midwater (0.27 [0.19, 0.34]), benthopelagic (0.43

[0.36, 0.49]), and benthic (0.52 [0.44, 0.61]) groupings, but were generally less certain and more similar across primary consumer, secondary consumer, and tertiary consumer groups (0.52 [0.39, 0.66], 0.49 [0.36, 0.63], 0.42 [0.28, 0.56], respectively). Intercept estimates, on the other hand, varied across all groups. Estimates increased across primary, secondary, and tertiary carnivores (0.14 [-0.30, 0.60], 0.53 [0.07, 1.03], 0.89 [0.33, 1.46], respectively) and decreased across midwater, benthopelagic, and benthic groups (0.08 [-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 random effect, the data support that species-specific effects not represented by diet or habitat classification impact fish DDX bioaccumulation. Random intercept estimates ranged from -1.42 [-1.79, -1.07] for market squid to 1.17 [0.59, 1.75] for starry rockfish, but were generally small and near zero (Figure 3). The coefficient on year was negative (-0.056 [-0.06, -0.05]), indicating a decrease in $[DDX_{fish}]$ through time that was not explained by changing $[DDX_{sed}]$ values.

4. Discussion

Despite more than half a century since the cessation of industrial dumping in the SCB, local ecosystem contamination continues to mirror the spatial legacy of contaminant disposal. Thus, the impacts of ocean dumping on marine ecosystems and fisheries resources remain place based, despite the long-held belief that the vastness of the ocean holds nearly limitless capacity for contaminant mitigation via dilution and advection (Lumsdaine 1976). That said, our findings clearly demonstrate that ecological characteristics mediate the strength of linkages between sediment DDX concentrations and the DDX burdens of fishes. Based on our findings, we propose a conceptual model of DDX transport in the coastal ecosystem (Figure 4) where the magnitudes of sediment signatures are highly conserved across space, and trophic ecology and habitat use mediate the vertical transport, lateral mixing, and biomagnification of DDX through the food web. Overarching all these factors, the magnitude of DDX in the SCB ecosystem as represented by fishes decreased over our study period, suggesting that the deleterious effects of these legacy pollutants in the SCB will continue to diminish over time as sediment DDX becomes less bioavailable.

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

A linear relationship between transformed $[DDX_{sed}]$ and $[DDX_{fish}]$ exists within the SCB across two decades and over 60 species of fishes. We provide evidence that local $[DDX_{sed}]$ is the primary control on $[DDX_{fish}]$ once lipid content of the fish composite has been accounted for and explains nearly half of the total variation in $[DDX_{fish}]$. Results mirror previous, more targeted, studies from the region that found that contaminant concentrations of flatfish were highest near Palos Verdes where sediment concentrations maxima are located (Mearns et al. 1991, McLaughlin et al. 2021) and SCB-wide relationships between sediment contaminant concentrations and flatfish tissue concentrations were highly correlated for both DDX and PCBs (Schiff and Allen 2000, Allen et al. 2002, 2004). This research expands upon previous work and suggests that sediment can serve as a robust predictor of fish organic contamination across the full range of recreationally fished species.

The spatial distribution of DDX in sediments reflects historical dumping and ocean transport. The highest concentrations of DDX occurred on the Palos Verdes Shelf (PVS) and immediately upcoast, presumably due to the discharge of roughly 870-1450 tons of DDX onto the PVS between 1947 and 1971 (Eganhouse and Pontolillo 2008) and the subsequent transport of these contaminants by ocean currents flowing northward at the discharge depth (Allen et al. 2006). The year coefficient of our encounter model systematically increased through time, likely due to a decrease in MDLs as analytical methods improved (Du et al. 2020), yet we observed no systematic changes in non-zero $[DDX_{sed}]$ values through the sampling period. The lack of an observable trend in non-zero $[DDX_{sed}]$ values can be attributed to sediment sampling procedures, where a single grab sample consisted of the top 2-5 cm of the sediment column. Sedimentation rates within the region have been recorded at 1-5 mm/ year (Ferré et al. 2010, Kivenson et al. 2019) and natural degradation of DDX is extremely slow, with reported half-lives generally >20 years (Taylor et al. 2019, Kurek et al. 2019). Observed $[DDX_{sed}]$ values, therefore, represent a time-integrated average that would not be expected to change substantially over a 15-year sample period (Young et al. 1977). Interestingly, although $[DDX_{sed}]$ showed limited change through time, $[DDX_{fish}]$ showed a marked decrease over the past 20 years, as evidenced by the negative year coefficient (Figure 3). The most likely cause of this discrepancy is a decrease in the bioavailability of DDX to fish and benthic organisms not reflected in bulk $[DDX_{sed}]$ values, either due to downward migration of DDX in the sediment column or reduced 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

al. 2019), and varying correlations of $[DDX_{sed}]$ with tissue concentrations across habitat groups is therefore likely reflective of differences in dietary pathways.

In addition to diet, the extent of lateral mixing further impacts fish DDX bioaccumulation. Demersal fishes typically occupy a smaller range than pelagic or midwater species, and thus their DDX concentrations are more reflective of local conditions (SI Appendix, Table S3). For this reason, benthic fishes show both the largest slope term and smallest intercept term of any habitat group within our model, as they closely track local sediment conditions. For pelagic species that are characterized by seasonal migrations and extensive spatial movement (Mais 1974, Konno and Wolf 2001, Wolf et al. 2001), the region of capture may not be representative of where they forage and observed DDX concentrations may reflect a region-wide average of water-column DDX concentrations rather than local sediment concentrations.

Finally, biomagnification, a central process in ecotoxicology (Walters et al. 2016), was evidenced in our system by larger intercept estimates for higher trophic level organisms (Figure 3). Consistent slope terms across primary, secondary, and tertiary consumers indicates similar rates of biomagnification across $[DDX_{sed}]$ gradients. Notably, patterns in biomagnification became evident only after accounting for background sediment contamination. There is a long history of using trophic position as a predictor of organism contamination (Kidd et al. 1995, Jardine et al. 2006, Corsolini and Sarà 2017). Although terrestrial taxa show clear patterns of biomagnification (Kelly et al. 2007), patterns among marine fishes are less clear, particularly when looking over a broad region or multiple species (Gray 2002, Bonito et al. 2016). More localized studies tend to find biomagnification for DDX chemicals, however, exact rates can vary with latitude, chemical constituent and subset of the food web sampled (Walters et al. 2016, Hoondert et al. 2020). Recent modeling studies and reviews have examined the impact of variable contaminant exposure on bioaccumulation (Borgå et al. 2012, Kim et al. 2016, Mackay et al. 2016), however, empirical evidence for this process has been limited to date. Our study illustrates the importance of accounting for spatially variable contaminant exposures using empirical data, and results suggest that pooling samples across regions with different background contamination is ill-advised.

Recognition of context dependent controls on bioaccumulation is a substantial step forward in understanding of this issue, as previous results examining the relative importance of ecological factors on contaminant bioaccumulation have been mixed. Some studies found that foraging habitat was a stronger

predictor of tissue contaminant concentrations, and that the subsequent biomagnification was secondary (Goutte et al. 2015, Yoshino et al. 2020, Carreira et al. 2023) while others found that age and trophic position, as opposed to habitat and carbon source, dominated bioaccumulation (Vander Zanden and Rasmussen 1996, McIntyre and Beauchamp 2007). Similar to our results, Dromard et al. (2018) posited that biomagnification became a less important pathway of bioaccumulation in more contaminated areas and Fonseca et al. (2019) found that rates of bioaccumulation were similar across sites with variations in biota concentrations reflecting baseline differences in site environmental levels.

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

[DDX_{sed}], diet, and habitat explained most variation in [DDX_{fish}], however, species-specific factors, reflected in random effects, further impacted [DDX_{fish}] (Table 1). Deviations from group means (i.e., species random effects) could reflect unmodeled aspects of life history such as growth, lifespan, or habitat utilization. For instance, species that grow quickly often exhibit low contaminant concentrations due to growth dilution, wherein fish with high growth efficiency add more tissue per unit contaminant consumed in prey, diluting contaminants in a larger biomass (Karimi et al. 2007, Madenjian et al. 2016). Growth dilution is perhaps evidenced by the large negative anomaly for Market squid (*Doryteuthis opalescens*), a pelagic species that lives only 9 months (Yaremko 2001), indicating lower than expected lipid-normalized DDX concentrations. Our modeling framework also implicitly assumes that fish composites reflect the distribution of sediment contaminants within their zone of capture; however, certain species preferentially use particular habitats within a given fishing zone, decoupling an individual's exposure from the average benthic conditions (Wolfe and Lowe 2015, Burns et al. 2021). For example, due to the documented positive relationship between depth, organic content of sediments, and [DDX_{sed}] within the SCB (SI Appendix, Figure S2), deeper dwelling fish that reside in organic-rich sediments, such as Starry rockfish (*Sebastes constellatus*), may accumulate more contaminants. Proximity to point sources of contamination, such as rivers, estuaries, and sewer outflows, may also increase contaminant burdens of individuals (Ahr et al. 2015). Future research could use more specific location-of-capture information in conjunction with movement patterns to optimize sediment exposure calculations (Melwani et al. 2009).

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

proxy for fish contamination in regions without a current monitoring program. Modeling results also indicate that using generalized ecological classifications produces results that are comparable to more detailed, species-specific models (Table 3). Most consumption advice to consumers generally occurs on a species-by-species basis (Oken et al. 2012), and consumption recommendations for unmeasured species can be absent or confusing; a more generalized framework would allow consumption advisories to be extended out to unmeasured species. Lastly, differing patterns across habitat groups have implications for how advisories are created. For example, a region-wide advisory for mobile pelagic species may be sufficient, whereas advisories for benthic fishes need to consider highly localized conditions. This integrated approach enhances our ability to address the complex dynamics of contaminant exposure and bioaccumulation, ultimately contributing to more informed environmental and public health policies.

Environmental contaminants are both increasing and diversifying through time, posing substantial threats to human and ecosystem health (Bernhardt et al. 2017). Despite more than half a century since the cessation of industrial dumping in the SCB, the impacts remain prominent. DDX concentrations in marine mammals frequenting the region are among the highest worldwide (Mackintosh et al. 2016) and critically endangered species, such as coastal populations of the California condor, are exposed to levels capable of impacting reproduction (Stack et al. 2022). Our findings, however, suggest spatial and ecological nuance to the DDX ecosystem pathways of toxin transport in the region. Leveraging this nuance should be an important part of efforts to safeguarding both people and the environment against the legacy of ocean dumping.

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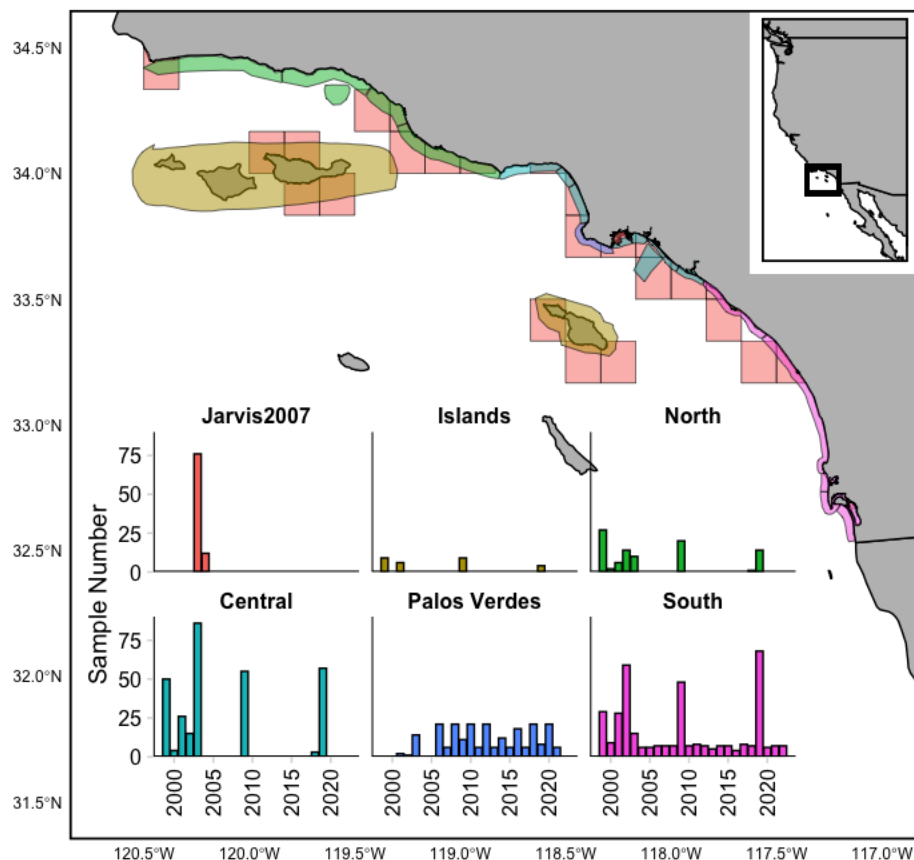
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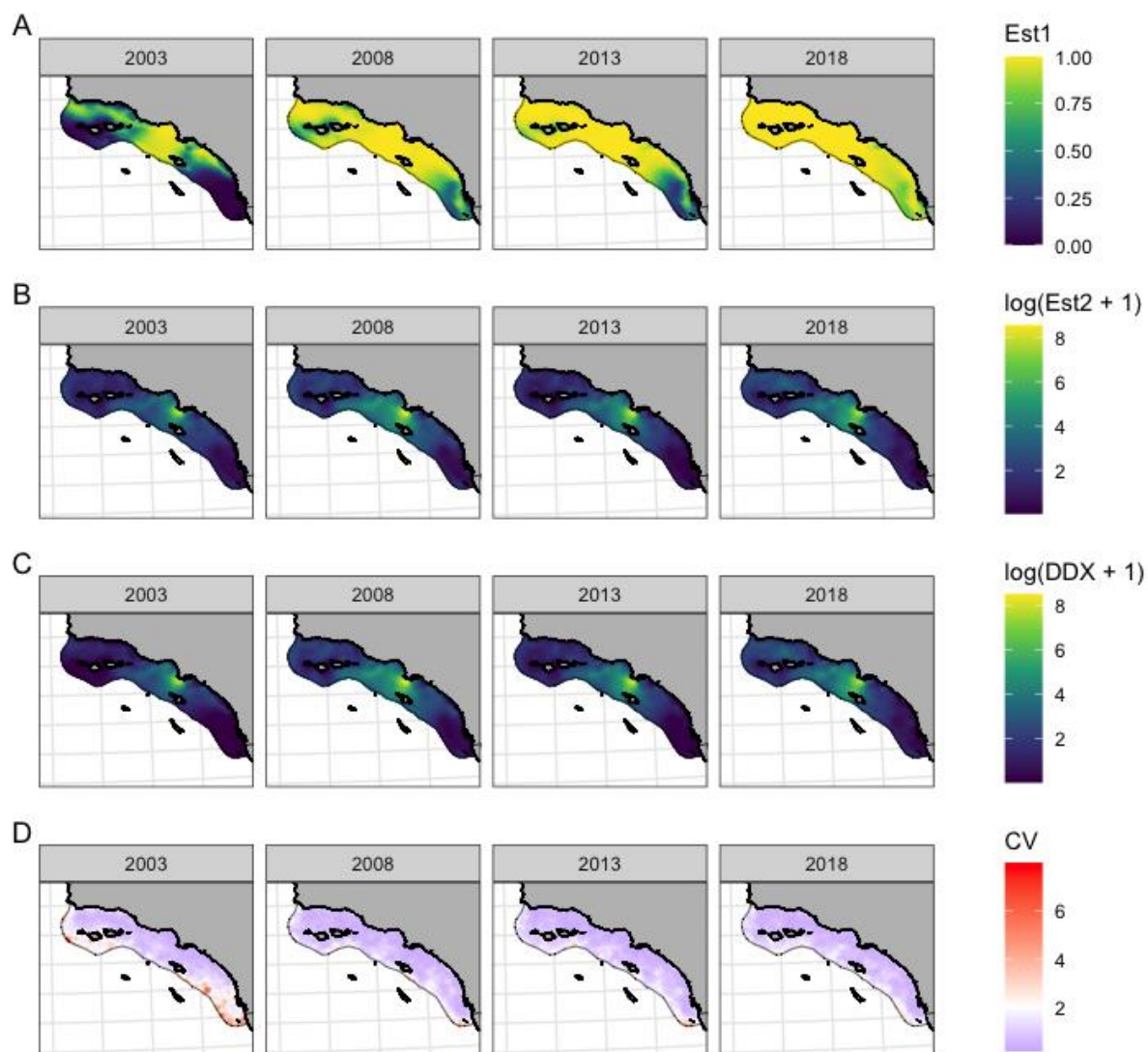
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Figure 1. Map of fishing zones and the number of fish samples through time, by region (inset). Nearshore polygons are derived from McLaughlin et al. (2021) and pink blocks are California Department of Fish and Game 256 km² fishing blocks.

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

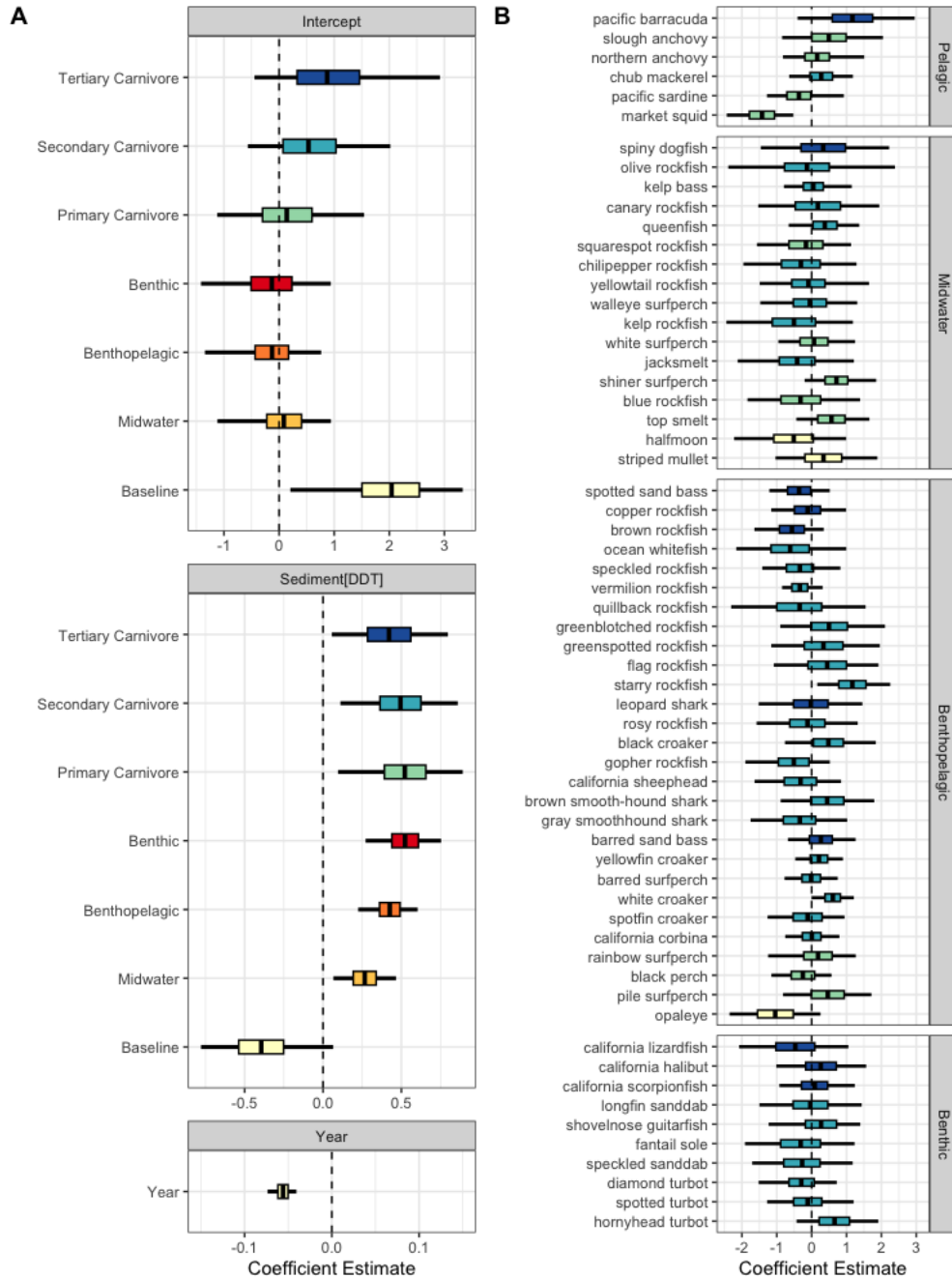


Figure 3. Model estimated posterior distributions for fixed (A) and random (B) effects from the diet-habitat-species-year model (Table 1). The reference category is herbivore and pelagic. Center lines are the mean estimated parameter, colored boxes represent the 80% credible interval, and the black lines 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.

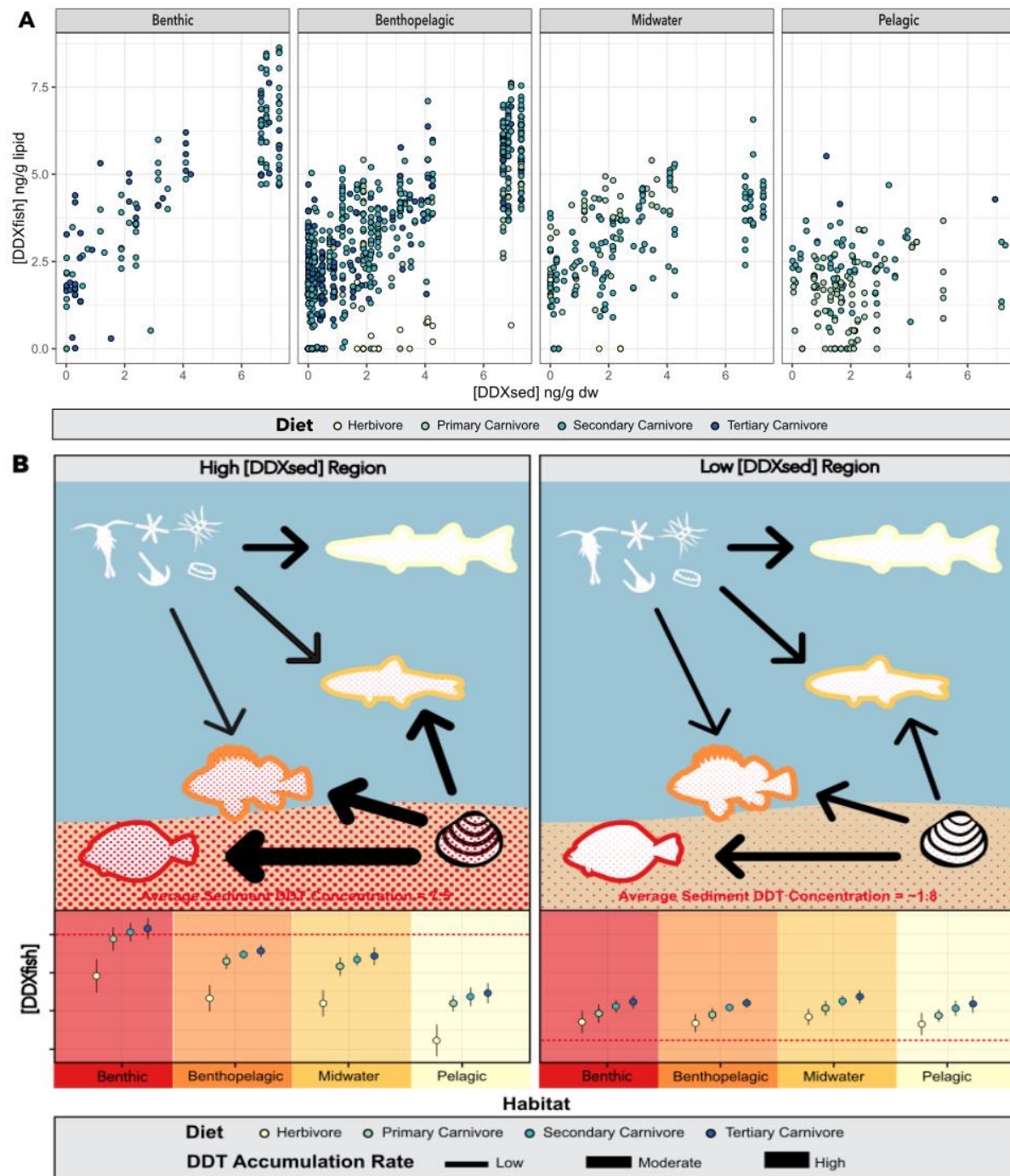


Figure 4. (A) Transformed [DDX_{sed}] data for each fishing zone versus [DDX_{fish}] values, separated by diet (color) and habitat (position), and (B) a conceptual figure of bioaccumulation in fishes within the SCB, illustrating that for high [DDX_{sed}] values habitat is the most important determinant, 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.

Table 1. Results of model selection for fish and sediment linear and mixed-effects models, showing model name, structure, LOOIC (leave-one-out information criterion), and elpd_{loo} . LOOIC describes the support of each candidate model where lower values indicate better models and the difference elpd_{loo} was used to compare model predictive capacity relative to the best model. For all models, $[\text{DDX}_{\text{fish}}]$ was treated as a left-censored variable, where values were constrained to fall between zero and the composite-specific MDL.

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 $[\text{DDX}_{\text{sed}}]$ and $[\text{DDX}_{\text{fish}}]$?	Null	$[\text{DDX}_{\text{sed}}]$		3642.4 (57.3)	-350.1 (25.5)	0.51
2. How do ecological factors mediate the relationship between $[\text{DDX}_{\text{sed}}]$ and $[\text{DDX}_{\text{fish}}]$?	Diet (intercept only)	$[\text{DDX}_{\text{sed}}] + \text{Diet}$		3509.4 (55.6)	-283.6 (22.3)	0.57
	Diet (slope only)	$[\text{DDX}_{\text{sed}}] + [\text{DDX}_{\text{sed}}] : \text{Diet}$		3482.2 (54.9)	-270.0 (21.7)	0.57
	Diet	$[\text{DDX}_{\text{sed}}] * \text{Diet}$		3475.1 (55.3)	-266.5 (21.7)	0.58
	Habitat (intercept only)	$[\text{DDX}_{\text{sed}}] + \text{Habitat}$		3488.5 (57.8)	-273.2 (23.9)	0.57
	Habitat (slope only)	$[\text{DDX}_{\text{sed}}] + [\text{DDX}_{\text{sed}}] : \text{Habitat}$		3413.5 (58.7)	-235.7 (23.5)	0.59
	Habitat	$[\text{DDX}_{\text{sed}}] * \text{Habitat}$		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

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