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Critical Review

# A Global, Cross-System Meta-Analysis of Polychlorinated Biphenyl Biomagnification

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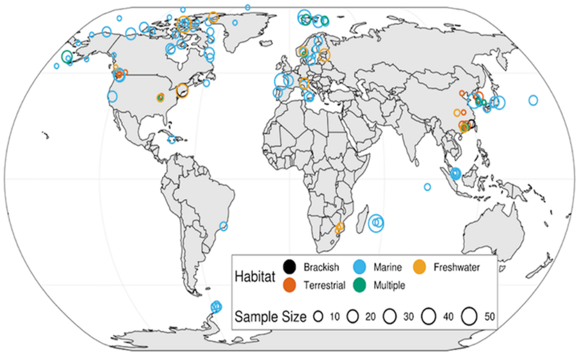
**ABSTRACT:** Studies evaluating the mechanisms underpinning the biomagnification of polychlorinated biphenyls (PCBs), a globally prevalent group of regulated persistent organic pollutants, commonly couple chemical and stable isotope analyses to identify bioaccumulation pathways. Due to analytical costs constraining the scope, sample size, and range of congeners analyzed, and variation in methodologies preventing cross-study syntheses, how PCBs biomagnify at food web, regional, and global scales remains uncertain. To overcome these constraints, we compiled diet (stable isotopes) data and lipid-normalized concentrations of sum total PCB ($PCB_{ST}$), seven indicator PCB congeners, and their sum ($PCB_{\Sigma 7}$). Our analyses revealed that the number of congeners analyzed, region, and class most strongly predicted $PCB_{ST}$, while similarly, region, class, and feeding location best predicted $PCB_{\Sigma 7}$ and all seven congeners. We also discovered that $PCB_{ST}$, $PCB_{\Sigma 7}$, and the seven indicator congeners all occur in higher concentrations in freshwater than marine ecosystems but are more likely to biomagnify in the latter. Moreover, although the seven congeners vary in their propensity to biomagnify, their trophic magnification factors are all generally greater in the Atlantic than the Pacific. Thus, novel insights regarding PCB biomagnification across taxonomic, food webs, regional, and global scales can be gleaned by leveraging existing data to overcome analytical constraints.



## INTRODUCTION

Once widely utilized as pesticides or in industrial processes, persistent organic pollutants (POPs) are among the most pervasive threats to environmental and human health. Due to their organic structures, many POPs are hydrophobic and lipophilic, resulting in their predisposition to bind with particulate matter in aquatic environments and adipose tissues in organisms. These characteristics enable POPs to resist biological and chemical breakdown,[1] bioaccumulate and biomagnify up food webs through trophic transfer,[2] and travel long distances through atmospheric processes and ocean currents.[3−6] As a result of these transport processes, POPs have become a global challenge that require an equally comprehensive solution.

In 2001 the Stockholm Convention called for the cessation of production of the 12 most hazardous POPs ("the Dirty Dozen").[7] Despite nearly two decades of international regulation, some of these POPs are still documented at high concentrations.[8−12] Understanding how these contaminants continue to biomagnify and where they are most likely to accumulate to toxic levels therefore remain pressing scientific, public health, and conservation challenges. To mitigate the effects of these harmful chemicals, a comprehensive understanding of POP bioaccumulation mechanisms across taxa, trophic levels, ecosystem types, and regions is needed. However, the high cost of analyses (e.g., an average of US $787.50 per sample of water, sediment, or soil analyzed for polychlorinated biphenyls,[13] hereafter PCBs; min−max: ∼$75 to $1,000 U.S. dollars[14]) constrains sampling size in individual studies, and the variation in both methodology and reported units of POP concentrations prevents the cross-study syntheses necessary to garner holistic insights about biomagnification dynamics.

PCBs are a group of particularly harmful POPs that are among the Dirty Dozen.[2] PCBs have been linked to cancer[15−18] and a multitude of other detrimental human and animal health impacts to the immune system,[19−21] reproductive system,[22,23] nervous system,[24] and endocrine system.[25−27] Their acute toxicity and environmental persistence have since motivated significant research on PCB bioaccumulation pathways. These studies typically couple measurements of PCB congeners (of which there are 209) with stable isotope analyses to expose the dietary mechanisms for biomagnification by linking consumers to prey ($\delta N^{15}$) and carbon sources

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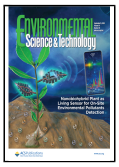

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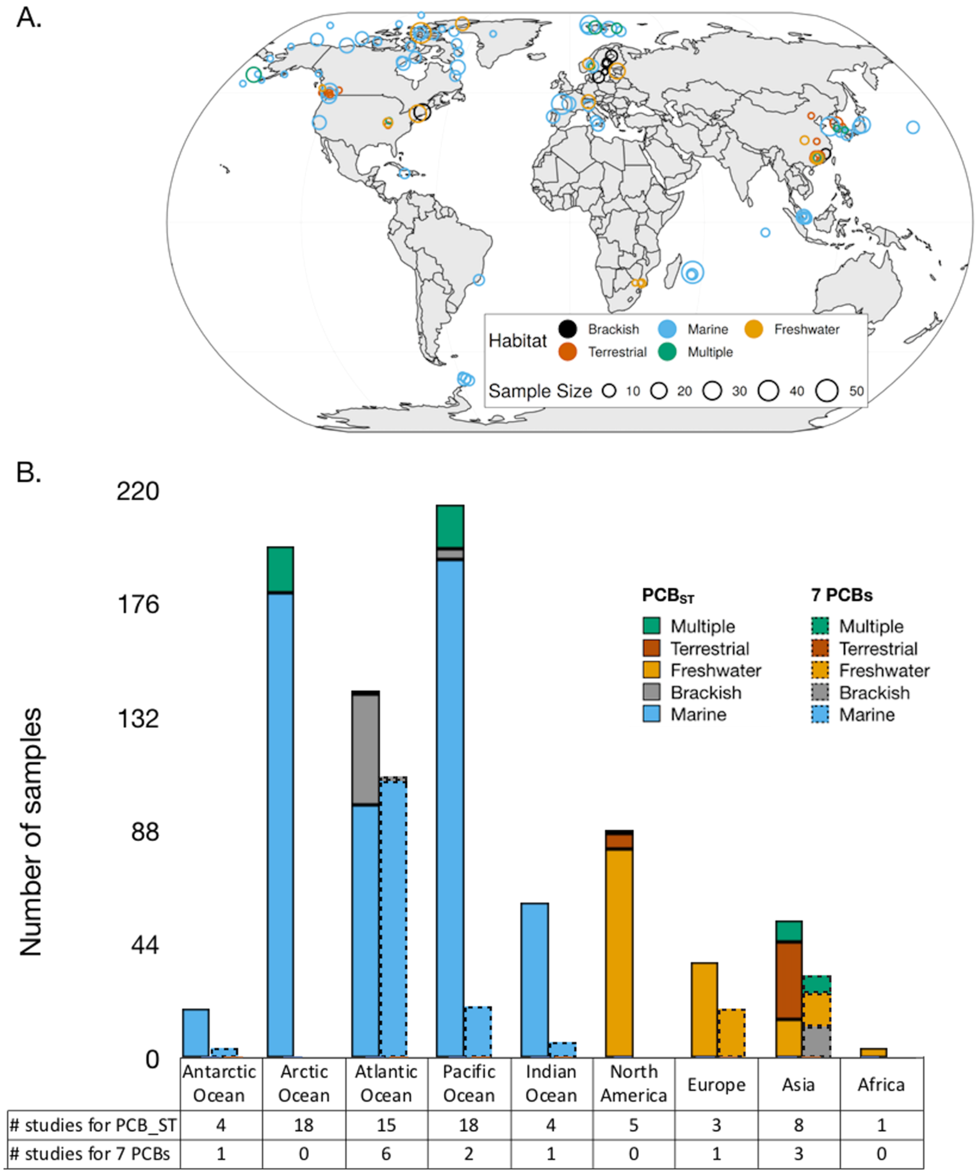
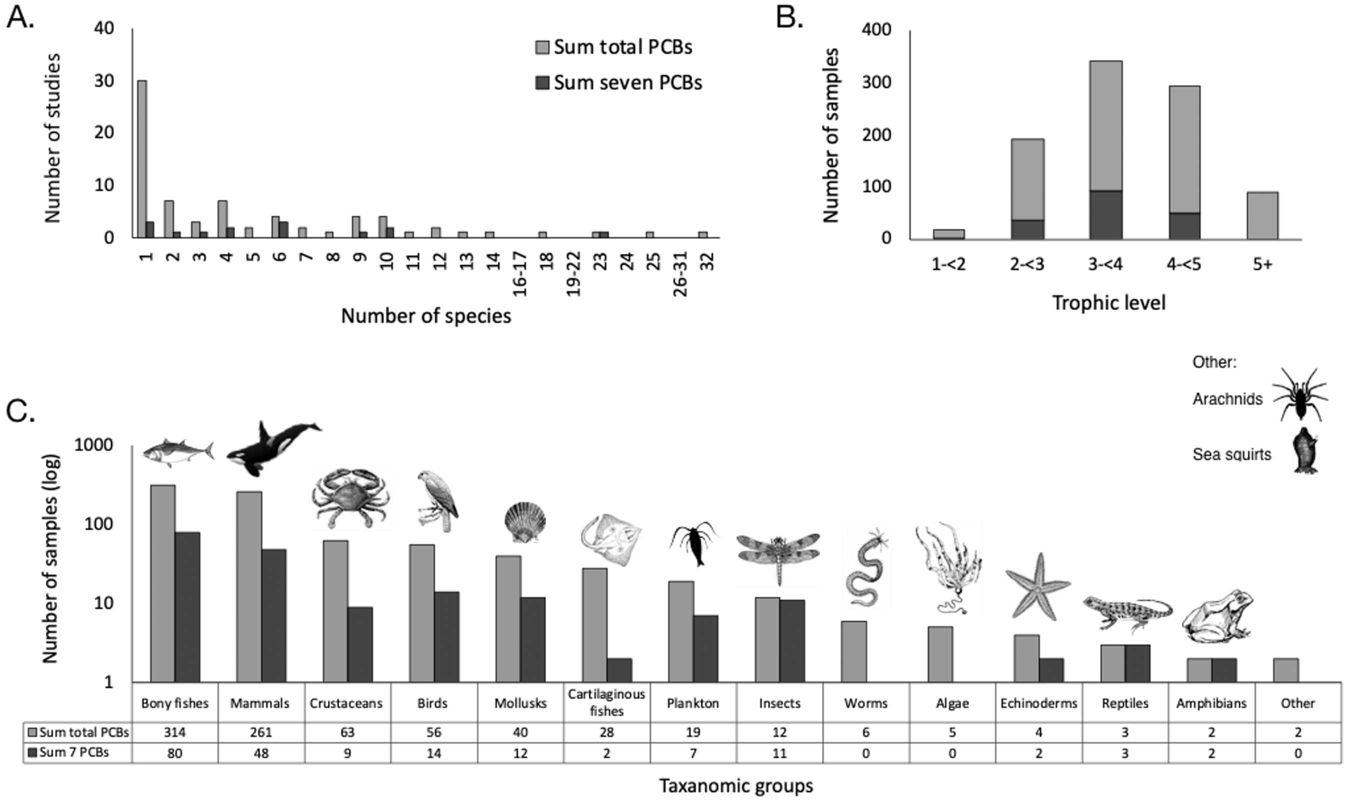


| | Antarctic Ocean | Arctic Ocean | Atlantic Ocean | Pacific Ocean | Indian Ocean | North America | Europe | Asia | Africa |
|---|---|---|---|---|---|---|---|---|---|
| # studies for PCB_ST | 4 | 18 | 15 | 18 | 4 | 5 | 3 | 8 | 1 |
| # studies for 7 PCBs | 1 | 0 | 6 | 2 | 1 | 0 | 1 | 3 | 0 |

**Figure 1.** Geographic distribution of PCB and stable isotope sampling reported in peer-reviewed studies. Locations where organisms measured for lipid normalized PCB concentrations and stable isotopes in our database were collected (A) and a summary of the distribution of both PCB$_{ST}$ (solid lines) and that of the seven most commonly analyzed congeners (dashed lines) by ocean and continent (B). In A, each circle denotes the location where organisms were collected, the size of the circle denotes the number of samples reported in that location, and the circle color represents ecosystem type: brackish (black), terrestrial (orange), marine (blue), multiple systems (green), and freshwater (yellow). In (B), samples from various ecosystems are identically color-coded.

($\delta C^{13}$).[28,29] Most studies conducted to date have sampled PCBs and stable isotopes of only one or a few species (but see Zhang et al., 2017[30] and Byun et al., 2013[31] for analyses of 25- and 32-species, respectively) and typically focus on a specific water body or region (Figures 1 and 2). Moreover, the majority of studies report total sum PCB concentrations, rather than the concentrations of individual congeners, a norm that obstructs robust analyses of how congeners that vary in their chemical properties and environmental behavior biomagnify in different contexts.[32] Consequently, PCB levels measured for a given species are rarely placed in larger food web or spatial contexts (but, see Walters et al., 2016[32] and Corsolini and Sara, 2017[33]), despite widespread awareness that a species' vulnerability to PCB enrichment depends on the resources it consumes and on the contamination levels of its environment.[2,25]

To advance knowledge of PCB biomagnification processes and begin to overcome cost and methodological constraints, we synthesize peer-reviewed studies that incorporate both stable isotope and PCB analyses. Using a subset of these studies that report lipid-normalized PCB concentrations we identify gaps in data coverage and opportunities for compiling data across studies to assess global PCB contamination at geographic, taxonomic, and food web scales. We also apply a machine learning approach to quantify the relative importance of different drivers in mediating PCB concentrations and explore whether PCB biomagnification rates differ across ecosystems (e.g., marine, terrestrial, freshwater, brackish, and multiple systems) and oceanic regions. We then compare these analyses to identify whether the global trends observed for our 73-study data set of 815 sum total PCB concentrations align with those detected for a standardized set of PCB congeners,



| | Bony fishes | Mammals | Crustaceans | Birds | Mollusks | Cartilaginous fishes | Plankton | Insects | Worms | Algae | Echinoderms | Reptiles | Amphibians | Other |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ▨ Sum total PCBs | 314 | 261 | 63 | 56 | 40 | 28 | 19 | 12 | 6 | 5 | 4 | 3 | 2 | 2 |
| ■ Sum 7 PCBs | 80 | 48 | 9 | 14 | 12 | 2 | 7 | 11 | 0 | 0 | 2 | 3 | 2 | 0 |

**Figure 2.** Taxonomic scope of PCB biomagnification studies conducted to date. The number of species evaluated for both PCBs (e.g., summed total PCBs and seven ICES congeners) and stable isotopes per study (A), and the distribution of samples ($n = 815$ for summed total PCBs; $n = 190$ for the subset of seven PCBs) across trophic levels (B) and across taxonomic families (C). In C, "Other" includes arachnids and sea squirts ($n = 1$ sample each). Images were labeled for reuse and found using Google Images. All panels present data from summed total PCBs in light gray and the standardized subset of seven ICES congeners in dark gray.

the seven most reported congeners in our database, a data set that included 190 samples derived from 31 studies.

Our goals in coupling analyses of $\mathrm{PCB_{ST}}$, $\mathrm{PCB_{\sum 7}}$, and seven ICES congeners were to (1) evaluate whether similar driving factors emerged as being most and least important in predicting the two aggregate measures of PCB exposure ($\mathrm{PCB_{ST}}$ and $\mathrm{PCB_{\sum 7}}$), and thus assess the ability of this machine-learning approach to control for the number of congeners analyzed and reliably predict large-scale trends in PCB exposure, (2) evaluate potential differences in key drivers predicting the reported concentrations of individual congeners, and (3) assess which of the seven ICES congeners exhibit similar and dissimilar trends to those observed for $\mathrm{PCB_{ST}}$ and $\mathrm{PCB_{\sum 7}}$ and may therefore be useful indicators of aggregate PCB exposure. In identifying variation in data coverage, key drivers of PCB concentration levels, hotspots of PCB bioaccumulation, and correspondence between trends in the total sum and standardized set of seven congener data sets, this synthesis sets the stage for future efforts to better leverage existing data to understand and address global contamination issues.

## ■ MATERIALS AND METHODS

To synthesize stable isotope and PCB data, we conducted a search in April 2019 on Web of Science (www.webofknowledge.com) using topic words "polychlorinated biphenyl* or PCB*" AND "stable isotope*" with no time restriction. To expand the coverage of this search, we applied the same search criteria and incorporated any additional studies from the following journals that commonly publish on this topic: *Environmental Research, Chemosphere, PLOS one, Environmental Toxicology and Chemistry, Environmental Science and Technology, Science of the Total Environment, Environmental Pollution, Ecotoxicology and Environmental Safety, Marine Pollution Bulletin, Journal of Wildlife Diseases, Marine Ecology Progress Series, Environmental Science Pollution and Restoration, Chemistry and Ecology, Environment International, and Frontiers in Ecology and the Environment.* Relevant studies cited within studies identified in our initial search were also incorporated in our database. Across all studies, we only included those that published original data and reported both PCB concentration and stable isotope data, resulting in a total of 231 studies published between 1995 and 2019.

Since studies varied in reported units of measurement, we further reduced this database to the subset of 73 studies that reported PCB concentrations in ng/g lipid-normalized weight (lw). In these studies, we extracted 815 unique sum total PCB concentration ($\mathrm{PCB_{ST}}$) values, i.e., the summed concentration of all congeners measured for a given sample. $\mathrm{PCB_{ST}}$ was overwhelmingly the most commonly reported response variable, and only 31 of the 73 studies provided concentrations for individual congeners in either the main text or supplement files in addition to $\mathrm{PCB_{ST}}$ values. In the few studies that only reported individual PCB congener concentrations, we calculated $\mathrm{PCB_{ST}}$ ourselves. We herein present analyses focused on evaluating the relative importance of different drivers of $\mathrm{PCB_{ST}}$, as a measure of aggregate PCB exposure in line with prior studies,[34−36] at global, regional, and food web scales given the large and geographically well-distributed characteristics of this data set (Figure 1). Though a complete list of congeners analyzed were often reported in studies, the concentrations of individual congeners were not, precluding us from resolving the relative contributions of individual congeners in the $\mathrm{PCB_{ST}}$ analyses. However, we do include the number of congeners analyzed an explanatory variable to help control for between-study differences in this important dimension of analytical effort (see Data Analysis below) in our $\mathrm{PCB_{ST}}$ analyses.

We complement the $\mathrm{PCB_{ST}}$ data set with one in which the number and composition of congeners analyzed are standardized. In this second data set, we focus only on the seven most commonly analyzed and reported PCB congeners (PCBs 28, 52, 101, 118, 138, 180, 153) as well as their sum ($\mathrm{PCB}_{\sum 7}$), which we derived from 13 of the 73 studies. This particular set of seven congeners happens to be the full suite of indicator PCBs recommended for monitoring by the International Council for the Exploration of the Sea (ICES) given their high concentrations in commercial mixtures and wide chlorination range (Supporting Information (SI) Table S1).

For each data set, we then categorized samples by geographic region (i.e., country and, if relevant, continent or ocean including associated seas and bays) and ecosystem type: marine, freshwater, brackish (mix of freshwater and saltwater), terrestrial, and multiple systems (if the sampled organism utilizes multiple ecosystems). We also cataloged geographical coordinates when available and, if not provided, estimated them based on the study site description using Google Maps. In addition, we recorded the taxonomic family and class of each species sampled, the type of tissue sampled (i.e., fat, blubber, blood, red blood cells, plasma, egg, embryo, heart, kidney, liver, muscle, milk, spleen, pylori, whole body, or multiple tissues), the composition and total number of congeners analyzed, and relevant characteristics of the sampled organism including its biomass, age, and trophic level. If trophic level was not provided, we assigned the sampled species a trophic level using Fishbase.org or used the organism's consumer status, which we based on published information about its diet. Primary producer, primary consumer, secondary consumer, tertiary consumer, or apex predator organisms corresponded to trophic levels 1−5. To assess the validity of the assigned trophic levels, we first fit a linear model between isotopic nitrogen and trophic level using 252 samples for which both values were reported in the study. We then found that 92% of the assigned trophic level values for the remaining 526 samples in our data set were within the 95% prediction interval of this linear model, indicating that they correspond well with the nitrogen-derived trophic levels reported in studies (SI Figure S1).

We further classified organisms by feeding location. Organisms occurring exclusively within aquatic systems were categorized as benthic, demersal, benthopelagic, pelagic, bathypelagic, or bathy-demersal, and primarily land-based species were categorized as exclusively terrestrial (e.g., sparrows, spiders, owls, moths, pigeon), terrestrial/marine (e.g., Artic foxes, Polar bears, herring gull), or terrestrial/freshwater (dragonflies, amphibians, ducks). We also classified organisms by feeding behavior (filter feeder, deposit feeder, autotroph, herbivore, carnivore, or omnivore). When necessary, we used Web Plot Digitizer (https://automeris.io/WebPlotDigitizer/) to extract isotope data, trophic level, and PCB concentrations.

## DATA ANALYSIS

**Boosted Regression Trees.** Boosted regression trees (BRT) are a machine-learning algorithm based on an ensemble of classification and regression trees and are well-suited for modeling nonlinear relationships and interactions.[37,38] For our analyses, we used BRTs to assess the relative importance of 10 explanatory variables (trophic level, ecosystem, region, taxonomic class, age, biomass, feeding location, feeding behavior, tissue type, and the number of congeners analyzed) in driving $\mathrm{PCB_{ST}}$ across the entire data set. We repeated the same analyses for $\mathrm{PCB}_{\sum 7}$ and each of the seven ICES congener concentrations, but in these iterations only nine of the original ten explanatory variables were used (i.e., the number of congeners was not included because the same seven congeners were universally evaluated in this data set). After initial model evaluation (SI Figures S2−S4), we fit BRT models using $\mathrm{PCB_{ST}}$, $\mathrm{PCB}_{\sum 7}$, and the concentrations of the seven individual congeners as response variables. The BRT models were each fit using 50 bootstrapped iterations of the data wherein each iteration used a random sample with replacement.[38] In a BRT model, the importance of an explanatory variable is a relative value that is calculated from the average decrease in error when that variable is chosen in a tree node;[37−39] thus, explanatory variables with high relative importance are those that decrease the error of the model most.

**Partial Dependence Plots.** Partial dependence plots derived from BRT models show how the predicted response variable, here PCB concentration reported on a $\log_{10}$ scale, varies with respect to one of the explanatory variables, while holding all other variables constant.[38] For example, partial dependence plots allow us to evaluate changes in PCB concentration with respect to trophic level, while accounting for taxonomic, regional, and analytical differences—including the number of congeners analyzed—among samples in the analyses. While this method enables us to control the number of congeners analyzed (i.e., in $\mathrm{PCB_{ST}}$ models, we hold the number of congeners constant and estimate $\mathrm{PCB_{ST}}$ concentrations as if all 209 were measured), it is limited in that it used only the total number of congeners analyzed and not the proportion of individually identified congeners. Thus, we highlight that this approach was not able to account for congeners which may disproportionately influence $\mathrm{PCB_{ST}}$ concentrations. We made partial dependence plots of the topmost important variables identified by the BRT model for the $\mathrm{PCB_{ST}}$, $\mathrm{PCB}_{\sum 7}$, and seven individual congener data sets (SI Figure S5). We also created partial dependence plots for PCB concentration with respect to trophic level across different ecosystem types and feeding locations, two of the top influential variables that contained sufficient data for comparisons within both data sets to explore biomagnification dynamics. Trophic magnification factors (TMFs) were then calculated from these partial dependence plots as $10^b$ for $\mathrm{PCB_{ST}}$ and $\mathrm{PCB}_{\sum 7}$ as well as for each of the seven ICES congeners, where b is the regression slope of modeled output. Confidence intervals for the partial dependence plots were calculated from the 0.025 and 0.975 quantiles of the bootstrap iterations. All analyses was performed in R version 3.6.1 using the packages "gbm",[40] "caret",[41] "pdp",[42] "purrr",[43] "dplyr",[44] and "ggplot2".[45]

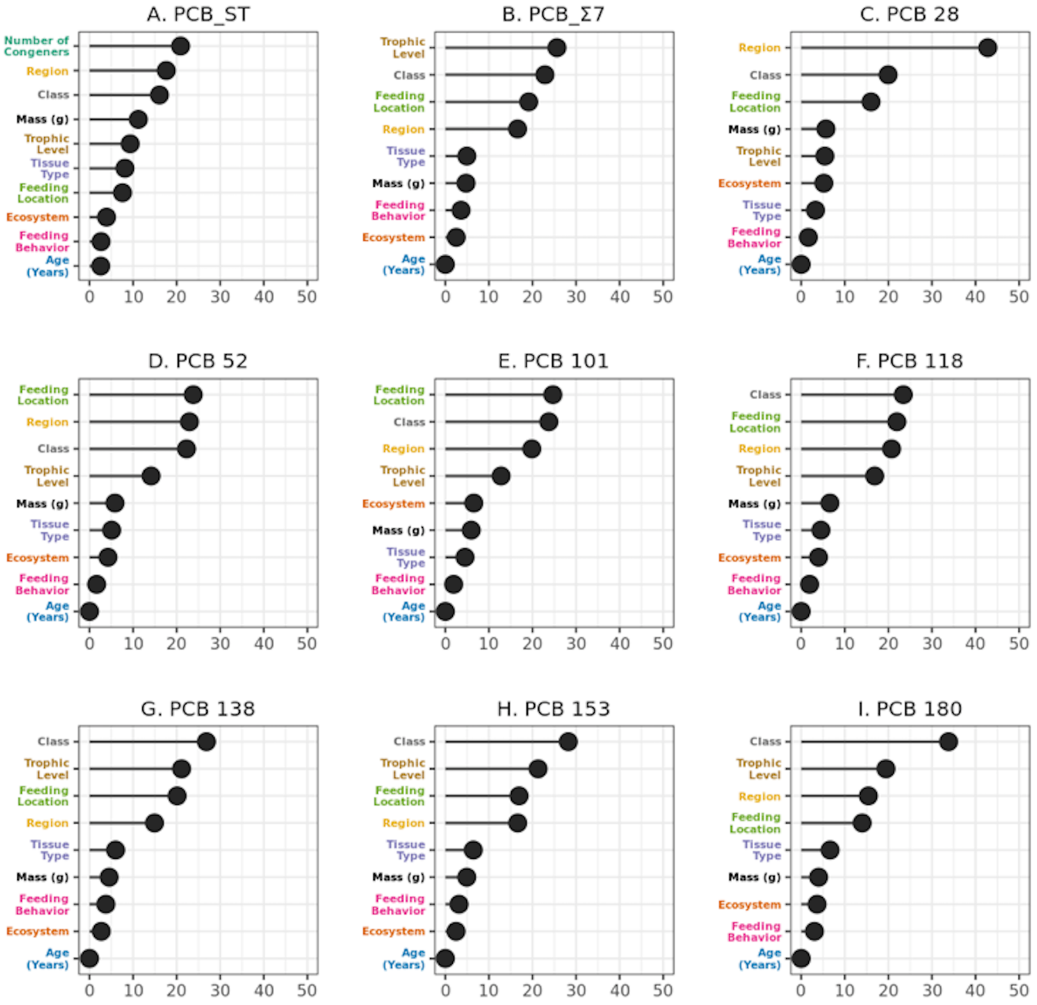


**Figure 3.** Drivers of PCB concentration levels. Boosted regression trees reveal the relative importance of 10 variables, arranged from top to bottom from highest to lowest relative importance in predicting $\mathrm{PCB}_{\mathrm{ST}}$ (A) and nine variables in predicting $\mathrm{PCB}_{\Sigma 7}$ (B), as well as concentrations for each of the seven individual congeners (C–I).

## RESULTS AND DISCUSSION

**Global Distribution of PCB Biomagnification Studies: Data Gaps and Data Synthesis Opportunities.** Of the 73 studies reporting lipid-normalized $\mathrm{PCB}_{\mathrm{ST}}$ and stable isotope data for a total of 815 samples, more were collected from the Pacific Ocean than any other location ($n = 214$ samples; 55% of which are from regional seas/bays), followed closely by the Arctic Ocean ($n = 198$ samples; 84% from regional seas/bays), the Atlantic Ocean ($n = 142$ samples; 45% from regional seas/bays) and North America ($n = 88$ samples; 34% from the United States, 66% from Canada; Figure 1A,B). In contrast, only 7% of all samples were collected in the Indian Ocean ($n = 60$ samples), 6.5% in Asia ($n = 53$ samples; of those 79% were from China, and 21% from South Korea), 4.5% in Europe ($n = 37$ samples, of those 51% were from Latvia, 33% from Italy, and 16% from Norway), 2% in the Antarctic Ocean ($n = 19$ samples), and a mere 0.5% was recorded in all of Africa, with all four samples from South Africa (Figure 1A,B). Not a single study in our database analyzed samples from Australia, the Caribbean, the Indo-Pacific, Central America, or South America, with only one study sampling off the coast of Brazil in the Atlantic Ocean. The scarcity of lipid-normalized PCB data in these regions highlights where additional sampling is needed, particularly in developing regions where only 6% of samples in our database were collected and where illegal dumping and improper disposal of toxic waste, as has occurred at the infamous ship graveyard in Mauritania, West Africa,[46] are current sources of elevated PCB concentrations.[47]

The level of research reporting $\mathrm{PCB}_{\mathrm{ST}}$ varied across ecosystem types as well, whereby the majority of $\mathrm{PCB}_{\mathrm{ST}}$ measurements were sampled from marine (67% of all samples; $n = 550$) and freshwater systems (17%, $n = 137$ samples; Figure 1A,B). Far fewer organisms were sampled from brackish systems (6%, $n = 47$ samples), multiple systems (5.5%, $n = 45$ samples), and terrestrial systems (4%, $n = 36$ samples; Figure 1A,B). The lack of data from brackish ecosystems and from organisms occupying multiple systems is concerning given that brackish systems occur at the base of watersheds and are therefore vulnerable to the cumulative input of pollution from upstream sources,[48,49] while organisms that forage within multiple ecosystems have been shown to be important vectors for POP movement into ecosystems that would otherwise be relatively unimpacted.[50]

In comparison, 57% of the 190 samples within the standardized set of seven congeners were collected from the Atlantic Ocean ($n = 107$ marine and $n = 2$ brackish ecosystems), followed by Asia (17%; $n = 13$ freshwater, $n = 12$ brackish, and $n = 7$ from multiple ecosystems), the Pacific

Ocean (11% all from marine systems), Europe (10%, $n$ = 19 from freshwater systems), the Indian Ocean (3%; $n$ = 6 marine ecosystem), and last the Antarctic Ocean (2%; $n$ = 4 marine ecosystem).

Studies also varied considerably in their taxonomic and food web coverage, such that the number of unique species analyzed in a single study ranged from 1 to 32. Over 50% of the 73 studies analyzed only one ($n$ = 30 studies) or two species ($n$ = 7 studies), and just over 10% of studies evaluated $PCB_{ST}$ and stable isotopes for over 10 species (Figure 2A). As for the subset of 14 studies reporting concentrations of the seven ICES PCBs, 46% analyzed only one ($n$ = 3) or six species ($n$ = 3), with the largest number of 23 species having been analyzed by a single study.[51]

Across all regions and ecosystem types, secondary (trophic level 3 to <4) and tertiary consumers (trophic level 4 to <5) have been analyzed more intensively for both the $PCB_{ST}$ and the seven ICES PCBs than apex predators (trophic levels ≥5), primary consumers (trophic level 2 to <3), and primary producers (trophic level 1 to <2) (Figure 2B). Of the taxa sampled, the two most intensively researched taxonomic groups for both data sets were bony fishes (39% $PCB_{ST}$ and 42% of the standardized set of seven PCBs) and mammals (32% and 25% of $PCB_{ST}$ and the set of seven, respectively), while amphibians, echinoderms, arachnids, and sea squirts have been relatively understudied overall (Figure 2C). This skew in sampling effort likely reflects the economic value and human health concerns associated with commercial and recreational fisheries, the propensity of environmental risk assessment studies to select indicator species representative of environmental health, and the economic or charismatic value of large-bodied mammals, including dolphins, whales, and seals.[52−54] The relatively low representation of primary producers, however, is due in part to our particular focus on studies reporting PCB concentrations in lipid-normalized units of measurement, as plant PCB values are more commonly reported in relation to a sample's wet weight. In general, our syntheses indicate that far more research on lower trophic levels and on a broader range of taxa is needed to gain holistic understanding of the key pathways through which PCBs are integrated into and accumulate within food webs. However, our databases, which are available through Dryad Digital Repository (10.5061/dryad.b2rbnzsbn), have the potential to support future research on PCB biomagnification at regional, ecosystem, and taxonomic scales given the comprehensive collation of existing data.

**Relative Importance of Global Drivers in Predicting PCB Concentrations.** The BRT model revealed that the number of congeners analyzed in a given study, which ranged from 1 to all 209 PCB congeners (mean ± SD: 49 ± 48), was more important relative to the nine other explanatory factors in predicting reported $PCB_{ST}$ concentrations (Figure 3A). While not surprising (i.e., the more congeners included in an analysis, the larger $PCB_{ST}$—the total sum of PCBs—will be), this result identifies a significant need for future studies that report $PCB_{ST}$ values to clearly contextualize their findings based on this dimension of analytical effort. This outcome also highlights the value of statistical techniques that can account for study-to-study variation in analytical resolution or methodology, such as the machine-learning approach we employ, in helping draw useful inferences from available data. Moreover, partial dependence plot analyses indicated that predicted $PCB_{ST}$ increases in a decelerating manner with the number of congeners analyzed, saturating at approximately 100 congeners (SI Figure S5-A). This result suggests that while researchers can gain significant resolution in evaluating a given organism's aggregate PCB exposure by quantifying more congeners (especially when they analyze congeners with known local sources and/or globally prevalent distributions), adding additional congeners beyond approximately 100 is unlikely to measurably improve resolution of aggregate PCB exposure and thus, depending on the study's objectives, may not warrant the added cost.

Along with the number of congeners analyzed, region and class were also particularly strong predictors of $PCB_{ST}$ concentrations in our full data set of 815 unique samples (Figure 3A). Similar to the $PCB_{ST}$ analysis, region and class, along with feeding location and trophic level, were consistently among the most important predictors of $PCB_{\sum 7}$, our standardized measure of aggregate PCB exposure (Figure 3B), and all seven ICES congeners (with the addition of mass being equally important as trophic level for PCB 28) (Figure 3C−I). Relative to their importance for predicting $PCB_{ST}$, feeding location and trophic level were stronger predictors of $PCB_{\sum 7}$ and the seven ICES congeners. It is likely that feeding location, which describes where a given sampled organism forages in the environment, emerged as a strong predictor of $PCB_{\sum 7}$ and the seven ICES congeners due to this factor's relevance in capturing the unique characteristics of these congeners—including their hydrophobicity, molecular weight, and chlorination—that control their behavior in different environmental media (e.g., freshwater, saltwater, and submerged soils) (SI Table S1). As for trophic level, this factor's high relative importance as a predictor of the seven ICES congeners and their sum aligns with an extensive literature documenting increasing PCB concentrations with increasing trophic level.[55−57] Since congeners biomagnify at different rates, it reasons that trophic level would be of higher importance in predicting concentrations for $PCB_{\sum 7}$ and some of the seven ICES congeners, while the ultimate effect of trophic level on $PCB_{ST}$ (which may include congeners that do not significantly biomagnify) is comparatively dampened. Thus, trophic level emerging as only a moderately influential variable for $PCB_{ST}$ corresponds with current knowledge of biomagnification dynamics and suggests other variables such as region or class better explain the variability of $PCB_{ST}$ at the global scale rather than a given species' relative position in its food web. Finally, the least influential variables (feeding behavior, ecosystem type, and age) remained roughly the same for the BRT models of $PCB_{ST}$, $PCB_{\sum 7}$, and most ICES congeners (tissue type was less important relative to ecosystem type for PCBs 28 and 101) (Figure 3A−I). Together, the BRT model results highlight a general alignment in the key predictors of both aggregate measures of PCB exposure ($PCB_{ST}$ and $PCB_{\sum 7}$) and the seven ICES congeners, providing collective evidence that class, region, feeding location, and trophic level are factors that exert particularly strong control over global trends in PCB contamination.

**Global Drivers of PCB Distribution.** Using partial dependence plot analyses, we determined how $PCB_{ST}$, $PCB_{\sum 7}$, and the seven ICES PCB concentrations vary among the four aforementioned explanatory variables exhibiting high relative importance: region, class, feeding location, and trophic level (Figure 3). In these analyses, we only included subcategories of each variable with five or more samples derived from two or more studies to ensure predicted PCB concentrations were not overtly biased by a few samples or single study (e.g., $PCB_{\Sigma 7}$ data from the Antarctic Ocean was not included because all data points were reported from a single study). Thus, due to significant differences in their size (e.g., 815 vs 190 unique data points in each database) as well as geographic and taxonomic scope, partial dependence plots for $PCB_{ST}$ and the seven congener data sets varied in the subcategories included for each of the four explanatory variables.

**Region.** In applying partial dependence plot analyses to assess how $PCB_{ST}$ levels vary across the eight regions with sufficient data in our database (see Figure 1 for distribution of data across regions), we found that $PCB_{ST}$ concentrations were predicted to be highest in Asia and Europe, followed by the Atlantic Ocean, the Antarctic Ocean, the Pacific Ocean and North America, and the Arctic and Indian Oceans (SI Figure S5-B). In contrast to these $PCB_{ST}$ results, $PCB_{\Sigma 7}$ and the seven ICES congener concentrations were all highest in the Pacific Ocean, followed by the Atlantic Ocean, and Asia (SI Figure S5-B), the three regions with sufficient data for analyses. This disagreement in the ranking of regions experiencing relatively higher $PCB_{ST}$ levels compared to $PCB_{\Sigma 7}$ and the seven ICES congeners may suggest that other PCBs, aside from the seven ICES congeners, are more prevalent in Asia and the Atlantic Ocean, causing these regions to rank relatively higher for $PCB_{ST}$ than $PCB_{\Sigma 7}$ in these analyses. To more confidently identify whether and, if so where, regional hotspots of this globally distributed toxin exist and thus resolve the disagreement regarding the most highly contaminated regions between our two aggregate PCB databases, additional analytically standardized and openly shared data sets that report individual congener concentrations are needed.

**Class.** While holding the nine other explanatory variables constant, partial dependence plots revealed that median predicted $PCB_{ST}$ concentrations were highest for Aves (birds) followed by Gastropoda (e.g., snails and whelks), Insecta (insects), Chondrichthyes (cartilaginous fishes), Plankton (both phytoplankton and zooplankton), Malacostraca (crustaceans), Mammalia (mammals), Cephalopoda (e.g., squid and octopi), Bivalvia (mollusks), Actinopterygii (bony fishes), and last, Maxillopoda (copepods) (SI Figure S5-B). Many other studies have reported relatively high aggregate PCB concentrations in birds, particularly in predatory bird species that are common in our $PCB_{ST}$ database and attribute their significant PCB exposure to their higher trophic positions among food webs.[26,58,59]

Partial dependence plot analyses of the seven ICES PCBs and their sum revealed that, of the five classes with sufficient data, $PCB_{\Sigma 7}$ concentrations were highest for bivalves, followed closely by birds, crustaceans, mammals, and bony fishes (SI Figure S5-B). Similarly, concentrations of PCBs 153 and 180, two of the heavier, more chlorinated PCBs with the lowest metabolic rates ($k_M$) and the highest measures of hydrophobicity (octanol−water distribution coefficients $k_{OW}$), were also highest in bivalves, suggesting these two congeners are particularly important in driving the high $PCB_{\Sigma 7}$ levels observed in bivalves. In general, bivalves tend metabolize organic pollutants at lower rates than other taxa, such as fish or crustaceans.[60,61] In addition, less-chlorinated PCB congeners are more readily metabolized and excreted than highly chlorinated congeners,[62] further supporting our finding that higher chlorinated PCBs are more heavily concentrated in this filter-feeding taxon relative to lesser-chlorinated PCBs. However, given that other studies have reported that the distribution of the seven ICES congeners in bivalves can vary widely by region and season,[61] we highlight that our general finding that bivalves tend to bioaccumulate particularly high concentrations of PCB 153 and 180 relative to other taxa is not universally applicable. In contrast to PCBs 153 and 180, the five other ICES congeners (PCBs 28, 52, 101, 118, 138), which range from tri- to pentachlorobiphenyls (3−5 chlorine atoms), exhibited patterns similar to $PCB_{ST}$ analyses such that their predicted concentrations were highest in birds relative to the other four taxa. These similar findings emphasize this taxa's particularly high propensity for PCB bioaccumulation.

While individual congener concentrations were generally highest among birds, bivalves, and bony fishes relative to crustaceans and mammals, PCBs 138 and 153 exhibited distinct patterns such that their concentrations in crustaceans and mammals were higher than the five other congeners (SI Figure S5-C). However, the predicted concentrations of all congeners for crustaceans and mammals exhibit particularly wide confidence intervals. This is likely due to the fact that one of the three studies reporting crustacean data was from a highly contaminated electronic waste site and that 90% of the mammal data for the seven congeners was extracted from a single study. The inferences we can draw regarding crustacean and mammal PCB exposure are therefore limited due to these significant constraints in data coverage.

**Feeding Location.** Partial dependence plots exposing the relative differences in PCB concentrations across feeding locations demonstrate that $PCB_{ST}$ concentrations are predicted to be much higher for organisms foraging in bathypelagic habitats (e.g., deep-sea aphotic, pelagic zone ∼1000 to 4000 m) relative to any other feeding location. Organisms feeding in both terrestrial and marine systems, such as polar bears or seabirds, exhibited the second highest $PCB_{ST}$ concentrations followed by organisms foraging in benthopelagic habitats (i.e., both benthic and pelagic zones), terrestrial-freshwater systems (i.e., ducks, frogs, and dragonflies), bathydemersal habitats (close to the sea floor of the deep-sea, aphotic zone), demersal (close to the sea floor in photic zones) and pelagic habitats (open ocean in the photic zone), which maintained similar $PCB_{ST}$ levels, benthic (sea floor/sediment surface) habitats, and last, those foraging exclusively in terrestrial systems (SI Figure S5-C) exhibited the lowest $PCB_{ST}$ concentrations. In short, organisms foraging in the deep-sea appear to have, on average, $PCB_{ST}$ concentrations 2.5 times higher than those foraging in the photic zone and on land, a result that aligns with prior studies that have identified deep oceanic waters as major sinks of persistent organic pollutants.[63,64] In contrast, $PCB_{\Sigma 7}$ concentrations as well as the concentrations of the three most highly chlorinated congeners, PCBs 138, 153, and 180, were greatest in organisms foraging exclusively in terrestrial systems followed by organisms foraging in benthic and benthopelagic habitats (SI Figure S5-C). Less chlorinated congener concentrations (i.e., PCBs 28, 52, 101, 118) were consistently highest in benthic habitats (SI Figure S5-C).

The large discrepancy in the relative concentrations of $PCB_{ST}$ and the seven ICES congeners measured in organisms differing in their feeding location is likely due to the congener-specific characteristics that influence the fate and transport of PCBs. Less chlorinated congeners are more water-soluble and more susceptible to environmental degradation compared to more persistent higher-chlorinated PCBs;[65] however, the aggregate measure of $PCB_{ST}$ does not reflect this congener-specific variability in chemical properties and environmental behavior, potentially driving the difference among the levels of $PCB_{ST}$ and the seven ICES congeners among organisms foraging in particular habitats. Additionally, the seven ICES PCBs have historically been included in high proportions of many PCB technical commercial mixtures. In the terrestrial habitats where these mixtures were manufactured, used, and disposed of, these particular PCBs would likely occur in relatively higher concentrations.

**Trophic Level.** For trophic level, we found concentrations of $PCB_{ST}$, $PCB_{\Sigma 7}$, and each of the seven ICES congeners all increased with increasing trophic level (Figures 4 and 5).

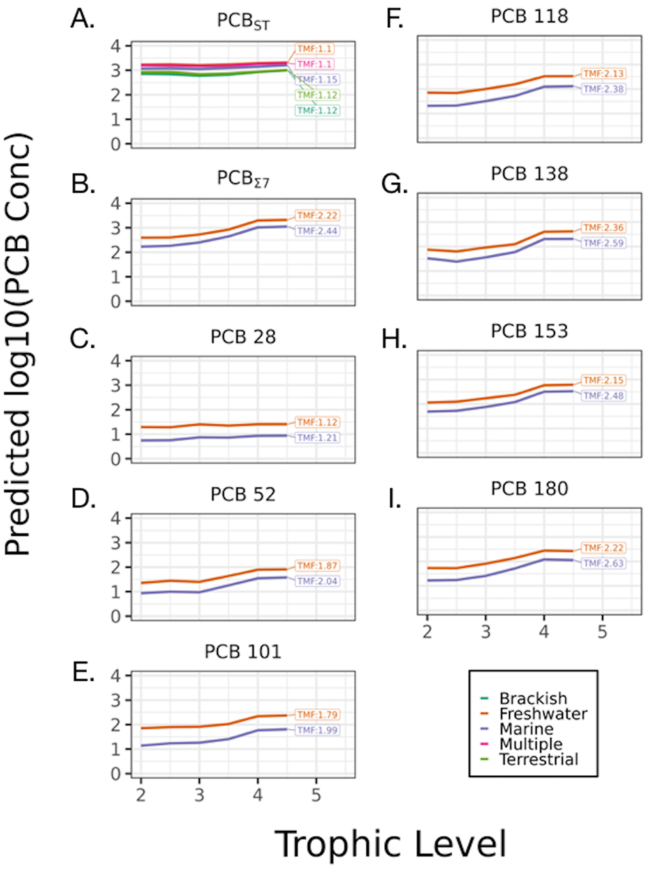


**Figure 4.** PCB biomagnification across ecosystems. Partial dependence plots reveal predicted concentrations reported on a log scale for $PCB_{ST}$ (A), $PCB_{\Sigma 7}$ (B), and the seven ICES congeners (C–I) relative to trophic level across brackish (teal), freshwater (orange), marine (purple), multiple (pink) and terrestrial (green) ecosystems. Based on these predicted PCB values, we calculated trophic magnification factors (TMFs) for $PCB_{ST}$, $PCB_{\Sigma 7}$, and each of the seven PCB congeners. TMFs are colored according to their corresponding ecosystem type. Trophic level on the *x*-axis begins at level 2, secondary consumers, to prevent skewed data due to the lack of primary producers in our data set.

These findings align with the extensive biomagnification literature that has repeatedly demonstrated positive relationships between trophic level and PCB concentration.[66,67] However, the slope of this relationship is known to vary depending on a congener's bioaccumulation characteristics and an organism's metabolic excretion rate.[32] Therefore, to garner more useful insights about PCB biomagnification dynamics, we explored whether biomagnification rates (i.e., the relationship between trophic level and PCB concentration) of $PCB_{ST}$, $PCB_{\Sigma 7}$, and each of the seven ICES congeners differ across ecosystems (e.g., marine, terrestrial, freshwater, brackish, and multiple systems) and oceanic regions. Consistent with prior partial dependence plots, we only include region and ocean subcategories with more than five samples derived from more than one study.

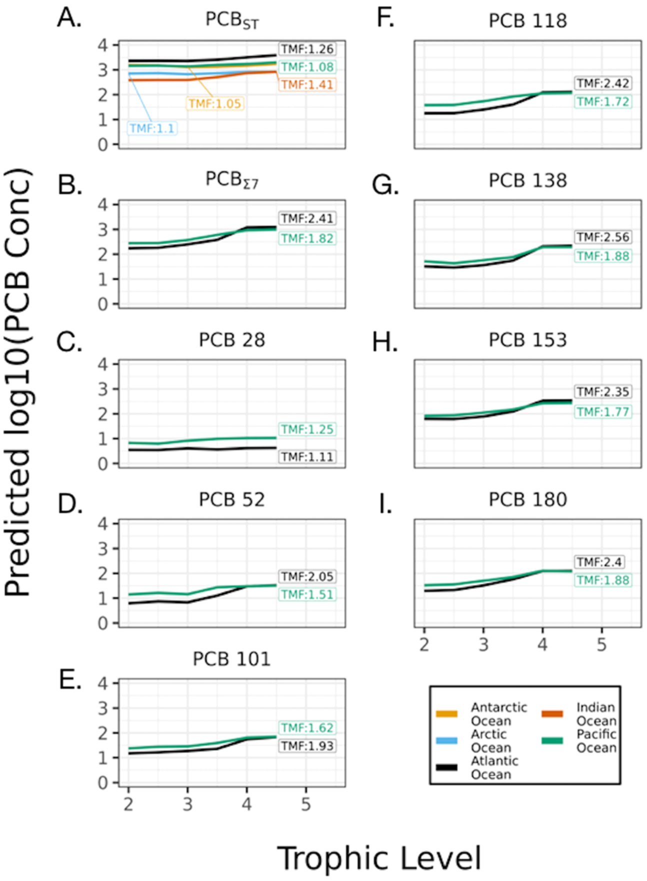


**Figure 5.** PCB biomagnification across oceans. Partial dependence plots reveal predicted concentrations reported on a log scale for $PCB_{ST}$ (A), $PCB_{\Sigma 7}$ (B), and the seven ICES congeners (C–I) relative to trophic level across the Antarctic (yellow), Arctic (light blue), Atlantic (black), Indian (orange) and Pacific (green) Oceans. Based on these predicted PCB values, we calculated trophic magnification factors (TMFs) for $PCB_{ST}$, $PCB_{\Sigma 7}$, and each of the seven PCB congeners. TMFs are colored according to their corresponding oceanic region. Trophic level on the *x*-axis begins at level 2, secondary consumers, to prevent skewed data due to the lack of primary producers in our data set.

**Biomagnification Across Ecosystems.** We found that, across all five ecosystems, $PCB_{ST}$ concentrations increased with trophic level and concentrations observed in basal species strongly predicted PCB concentrations at the top of their respective food chains (Figure 4A). Freshwater, multiple, and marine systems exhibit higher $PCB_{ST}$ concentrations compared to terrestrial and brackish systems, such that top predators exhibit $PCB_{ST}$ concentrations of 2083 ng/g lw, 1967 ng/g lw, and 1617 ng/g lw in these respective ecosystem types, but only 1016 ng/g lw in terrestrial systems and 1000 ng/g lw in brackish systems (SI Table S2).

In analyzing the effects of trophic level on $PCB_{\Sigma 7}$ and the seven ICES congeners in the two ecosystems with sufficient data, we found that similar to $PCB_{ST}$ predicted concentrations for both $PCB_{\Sigma 7}$ each of the seven PCB congeners were higher in freshwater systems than marine systems. On average, $PCB_{\sum 7}$ concentrations are nearly an order of magnitude greater per trophic level compared to marine systems (Figure 4B; SI Table S2), while the concentrations of each of the seven PCBs is two- to four-times greater in freshwater compared to marine organisms across trophic levels (Figure 4C–I; SI Table S2). While overall concentration levels among these two systems differ, we found that the pattern of relative contributions among individual congeners remained the same for both freshwater and marine ecosystems such that, on average, PCB 153 occurs at the highest concentrations across trophic levels, followed by PCBs 138, 118, 180, 101, 52, and 28.

Using the slope ($b$) of the linear relationship between trophic level and the $\log_{10}$ of predicted lipid normalized concentrations for $PCB_{ST}$, $PCB_{\sum 7}$, and the seven ICES PCBs, we estimated trophic magnification factors (TMFs) for each ecosystem type, ocean, and PCB metric as $10^b$. TMFs describe the rate of change in contaminant concentration per trophic level and are a common metric for assessing biomagnification potential across food webs, where a TMF> 1 is considered evidence of biomagnification.[56,68] In contrast to the variability seen in $PCB_{ST}$ concentrations across ecosystem types (Figure 4A), we found the rate of $PCB_{ST}$ biomagnification was similar in all five ecosystems, with TMFs ranging from 1.10 in freshwater and multiple systems (i.e., species that forage in more than one ecosystem type) to 1.15 in marine systems (Figure 4A). TMFs for $PCB_{\sum 7}$ were nearly twice as high as those of $PCB_{ST}$ and only slightly higher in marine systems (TMF 2.44) relative to freshwater systems (TMF 2.22) (Figure 4B). While aggregate PCB biomagnification for region-specific food webs vary widely throughout the literature (e.g., ∑PCB TMFs can range from 0.2–5.27 for sub-Arctic[69] vs NE-Pacific[70] marine food webs, respectively, while those reported for freshwater systems ranged from 2.8[71] in a subalpine lake to 3.68[72] in a highly contaminated reservoir in south China), and our findings suggest that ∑PCB biomagnification occurs at similar rates when analyzed from larger data sets and at broader, ecosystem scales, as done in this study.

Similarly, TMFs for each of the seven ICES congeners indicate all seven biomagnify within both freshwater and marine ecosystems, however, we found that all seven congeners have higher rates of biomagnification in marine (TMFs ranging from 1.21 for PCB 28 to 2.63 for PCB 180) than freshwater systems (TMFs ranging from 1.12 for PCB 28 to 2.33 for PCB 138) (Figure 4C–I). With the exception of PCB 138 and PCB 180 switching ranks from the highest and second highest TMFs in freshwater systems to the second highest and highest TMFs in marine systems, respectively, the remaining PCB TMFs were in the following descending order for both ecosystems: PCB 153, 118, 52, 101, and PCB 28. The differences in biomagnification rates among individual congeners is commonly attributed to congener-specific variability in hydrophobicity (as measured by octanol–water partition coefficient; $k_{OW}$), metabolic biotransformation rates ($k_M$), and chemical structure (number of chlorine atoms and molecular weight).[32] Our finding aligns with prior research in that TMFs were higher for the heavier, more chlorinated PCBs with high $k_{OW}$ and a low $k_M$[32] in our focal suite of congeners (see SI Table S1).

Collectively our findings of similarly ranked concentrations and TMFs for $PCB_{ST}$, $PCB_{\sum 7}$, and the seven ICES congeners suggest that the influence of the above-mentioned congener-specific characteristics on biomagnification remains consistent across both freshwater and marine ecosystems. In general, freshwater systems consistently exhibited higher predicted PCB levels whereas marine systems demonstrate a greater predisposition for PCB biomagnification. This may be due to the inherent differences in mechanisms of PCB exposure and food chain lengths among freshwater and marine food webs. Specifically, the higher PCB concentrations in freshwater systems may be due to the proximity and, consequently, more direct exposure of organisms in these lotic and lentic systems to land-based contamination sources via surface and storm-water runoff, whereas PCB transport to marine systems is generally through atmospheric deposition.[73−75] In comparison, studies have long-since found that the biomagnification of organic pollutants increases with each predator–prey link in the food chain. As a consequence, food webs with longer food chain lengths are generally more susceptible to biomagnification[66,76] and marine food webs tend to have significantly longer food chains relative to among freshwater food webs.[77]

**Biomagnification Across Oceans.** Since the majority of data included in this study was reported for marine organisms (67% of $PCB_{ST}$ and 72% of the standardized seven-congener data set), we further evaluate how PCB concentrations and the rate of biomagnification vary by oceanic region. Once again, only data with sufficient sample sizes were incorporated in these analyses. Similar to ecosystem type (discussed above), $PCB_{ST}$ increased with increasing trophic level in all five oceans (Figure 5A). Top predators in the Atlantic Ocean exhibited the highest average $PCB_{ST}$ concentrations (3914 ng/g lw), a level nearly double that estimated for top predators in the Pacific (2004 ng/g lw), the ocean with the second highest $PCB_{ST}$ levels (SI Table S3). As for our standardized seven-congener data set, we were only able to assess biomagnification within the Atlantic and Pacific Oceans. Like our $PCB_{ST}$ analyses, $PCB_{\sum 7}$ concentrations were higher in the Atlantic (ranged from 174 for primary consumers to 1244 ng/g lw for top predators) than in the Pacific Ocean (277 to 993 ng/g lw, for primary consumers and top predators, respectively) (Figure 5B; SI Table S3). In contrast, predicted concentrations of individual PCB congeners tended to be greater within the Pacific than Atlantic food webs, however, differences in predicted concentration varied by trophic position and congener (Figure 5C–I). More specifically, primary and secondary consumers exhibited higher concentrations in the Pacific Ocean for PCBs 28, 52, and 118 where concentrations were, on average, double that predicted for higher trophic level taxa in Atlantic food webs (SI Table S3). In contrast, with the exception of PCBs 28 and 101, which have higher concentrations in the Pacific across all trophic levels, tertiary predators demonstrated higher concentrations of the following PCBs in the Atlantic: 118, 138, 153, and, to a lesser degree, PCB 180 (SI Table S3).

In regard to biomagnification rate, $PCB_{ST}$ TMF values were greater than one in all five oceans, indicating that biomagnification is likely occurring (Figure 5A). The Indian Ocean exhibited the highest $PCB_{ST}$ TMF of 1.41, followed by the Atlantic (TMF: 1.26), Arctic (TMF: 1.10), Pacific (TMF: 1.08), and Antarctic Oceans (1.05). $PCB_{\sum 7}$ and all seven ICES congeners also had TMFs that were greater than one and were higher in the Atlantic relative to the Pacific Ocean with the exception of PCB 28, which demonstrated slightly higher TMF values in the Pacific (TMF 1.25) than the Atlantic Ocean (TMF 1.11). While PCBs 28 and 138 maintain the lowest and highest TMFs, respectively, for both oceans, the order of the remaining five PCB's propensity to biomagnify varied relative to one another between the Pacific and Atlantic Ocean.

Altogether, the metrics of aggregate PCB biomagnification for both standardized ($PCB_{\Sigma7}$) and nonstandardized data sets ($PCB_{ST}$) strongly differ from predicted concentrations of individual congeners in their distribution across the Atlantic and Pacific Ocean. This suggests that none of the individual seven ICES PCBs are suitable for serving as potential indicators for the distribution of aggregate PCB concentrations among these two oceans. In contrast, PCB biomagnification potential remained relatively similar across all PCB measures, whereby TMFs for $PCB_{ST}$, $PCB_{\Sigma7}$, and each ICES congener were all higher in Atlantic Ocean (with the exception of PCB 28) relative to the Pacific Ocean, implying that top predators in the Atlantic Ocean may be more vulnerable to PCB biomagnification than those in the Pacific.

## ■ PERSPECTIVES AND RECOMMENDATIONS FOR FUTURE RESEARCH

In collating data across peer-reviewed studies, this work garners several new insights regarding PCB bioaccumulation that we hope inspires further research to scrutinize and refine. First, we found evidence that the number of congeners analyzed is a critical factor influencing the concentrations of PCBs that are detected in biota. The outstanding importance of this factor highlights the need for future studies to present their data transparently by reporting individual congener concentrations to better support much-needed cross-study analyses. In addition, our findings of similarly ranked variables driving global concentrations for $PCB_{ST}$ and $PCB_{\Sigma7}$, and the seven individual ICES congeners, demonstrate our machine learning model, which had no a-priori information, may be a useful tool for cross-study analyses that are needed to analyze global PCB trends. Overall, our study highlights the value in collating data from multiple studies to bridge data gaps and advance understanding of the propensity of PCBs to biomagnify across multiple scales.

Due to the global nature of PCB contamination and biomagnification dynamics, illuminating the key factors controlling their transport and fate requires that investigators around the world report their PCB and stable isotope data in standardized units to enable cross-study syntheses and comparisons. Many of the studies in our literature review (68%) could not be included in our meta-analyses because of our inability to compare their reported values to those in the rest of the data set and, as discussed above, data limitations continue to obstruct our ability to resolve where global hotspots in PCB bioaccumulation exist. To push POP biomagnification analyses toward the forefront of the ecotoxicology field, future studies can continue to add to this database and those focused on other biomagnifying POPs through the reporting of pollutants and stable isotope data using standardized methods and units of measurement (e.g., lipid-normalized concentrations), thereby collectively advancing knowledge on the global transport and fate of POPs. Further, we encourage others to leverage the data compiled in our existing database to contextualize their findings relative to other organisms sampled in a similar region or from a taxonomic class. Such communal data sets provide one of the few comprehensive tools to understand and manage how PCBs move into and across ecosystems at the global scale.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.9b07693.

> Linear model demonstrating the validity of estimated trophic levels; pairwise correlation between all explanatory variables; hyperparameter selection for BRT model; recursive feature selection for BRT model; partial dependence plots for the second and third most influential predictive variables; congener-specific characteristics for the seven ICES congeners; partial dependence plot predicted PCB values by trophic level for ecosystem type and oceanic region; and studies included in our meta-analysis (PDF)

### Accession Codes

All data sets are freely available through the Dryad Digital Repository. 10.5061/dryad.b2rbnzsbn

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### Notes

The authors declare no competing financial interest.

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