Marine Pollution Bulletin

Mercury concentrations in fish species consumed in a Spanish Mediterranean region between 2011-2021: Bayesian multi-group Gaussian processes and hierarchical missing imputation

--Manuscript Draft--

Manuscript Number:	MPB-D-25-00928
Article Type:	Research Paper
Keywords:	Mercury; Fish; Trends; Gaussian processes; Multi-level modeling; Bayesian imputation
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Abstract:	Background Mercury (Hg) is a toxic metal, with fish consumption being the main source of exposure in humans. This study aimed to describe Hg concentrations in fish species consumed in the Valencian Community (Spain) and their trends during the period 2011-2021. Methods A retrospective study was conducted on Hg levels in fish meat between 2011-2021, using data from the Food Safety Monitoring Program of the Valencian Regional Government's. Descriptive analyses were performed for total Hg (THg) (n=799) and methylmercury (MeHg) (n=271) levels by fish group, year of sampling, and fishery origin. Bayesian Gaussian processes for multiple groups were used to evaluate trends in Hg concentrations by fish groups throughout the study period. Results Swordfish showed the highest Hg concentrations (median THg: 0.76 mg/kg; IQR: 0.47-1.17), with 30% of samples exceeding European limit values, followed by fresh tuna (0.46 mg/kg) and canned tuna (0.22 mg/kg). THg and MeHg levels in swordfish tended to decrease by around 0.5 mg/kg from 2011 to 2016 but then increased again to near their initial levels. Fresh and canned tuna showed decreasing trends in the first half of the study period, while between 2016-2021 levels increased in fresh tuna and decreased in canned tuna. Conclusions Most fish groups showed declining trends between 2011-2016. Data from the second half of the period was limited, except for swordfish, so results from this time should be interpreted with caution. Our findings on Hg levels in commercially sold fish species could be useful for guiding local fish consumption recommendations.

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To Editors of Marine Pollution Bulletin

Valencia, Spain, 05 March, 2025

Dear Editors,

Enclosed please find our manuscript entitled “**Mercury concentrations in fish species consumed in a Spanish Mediterranean region between 2011-2021: Bayesian multi-group Gaussian processes and hierarchical missing imputation”,** by Blanco-Calvo et al., submitted for your consideration for publication in ***Marine Pollution Bulletin*** as an original research article.

We consider the information contained in the manuscript is of interest to your readership. This work assessed mercury concentrations in fish consumed in the Valencian Community (Spain), a country with high fish consumption, as well as the associated factors (i.e. fish species and fish origin) and their trend throughout the study period (2011-2021). Data on Hg levels in fish comes from the fish sample collection as part of the Food Safety Monitoring Program of the Health Department of the Valencian Government and the subsequent analysis of mercury concentrations (total mercury, n=799, and methylmercury, n=271). Bayesian Gaussian processes for multiple groups (Multi-group GPs) were used to evaluate the trend of Hg concentrations by fish groups throughout the study period. Multi-group GPs are suitable models for time series data while sharing information between groups by considering correlations between them, which is especially useful with unbalanced groups as in this data set. The results of this study could be useful for guiding the Food Safety Monitoring Program, as well as for developing local fish consumption recommendations. Furthermore, the study of temporal trends of Hg in fish, at both regional and international levels, along with other factors, can help evaluate the effectiveness of measures applied to reduce exposure to mercury, such as those covered by the Minamata Convention.

This manuscript is an original work, it has not been previously published in whole or in part and it is not under consideration for publication elsewhere. All the authors have read the manuscript and agree it is ready for submission to a journal, accepting responsibility for the manuscript’s contents. The authors declare they have no actual or potential competing financial interests.

Thank you for your kind attention. I look forward to hearing from you,

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Mercury concentrations in fish species consumed in a Spanish Mediterranean region between 2011-2021: Bayesian multi-group Gaussian processes and hierarchical missing imputation

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Competing interests

The authors have no relevant financial or non-financial interests to disclose.

Funding

This work was supported by the Spanish Institute of Health Carlos III (Sara Borrell-FSE: CD21/0186) and the Spanish Ministry of Universities (Margarita Salas Grant MS21-133), and co-funded by the European Union (NextGeneration EU).

Abstract

Background

Mercury (Hg) is a toxic metal, with fish consumption being the main source of exposure in humans. This study aimed to describe Hg concentrations in fish species consumed in the Valencian Community (Spain) and their trends during the period 2011-2021.

Methods

A retrospective study was conducted on Hg levels in fish meat between 2011-2021, using data from the Food Safety Monitoring Program of the Valencian Regional Government's. Descriptive analyses were performed for total Hg (THg) (n=799) and methylmercury (MeHg) (n=271) levels by fish group, year of sampling, and fishery origin. Bayesian Gaussian processes for multiple groups were used to evaluate trends in Hg concentrations by fish groups throughout the study period.

Results

Swordfish showed the highest Hg concentrations (median THg: 0.76 mg/kg; IQR: 0.47-1.17), with 30% of samples exceeding European limit values, followed by fresh tuna (0.46 mg/kg) and canned tuna (0.22 mg/kg). THg and MeHg levels in swordfish tended to decrease by around 0.5 mg/kg from 2011 to 2016 but then increased again to near their initial levels. Fresh and canned tuna showed decreasing trends in the first half of the study period, while between 2016-2021 levels increased in fresh tuna and decreased in canned tuna.

Conclusions

Most fish groups showed declining trends between 2011-2016. Data from the second half of the period was limited, except for swordfish, so results from this time should be interpreted with caution. Our findings on Hg levels in commercially sold fish species could be useful for guiding local fish consumption recommendations.

Key words: Mercury; Fish; Trends; Gaussian processes; Multi-level modeling; Bayesian imputation

1 Introduction

Mercury (Hg) is a well-known toxic metal that comes from both natural and anthropogenic sources, the latter being the main contributor (Driscoll et al., 2013). Human activity has multiplied the atmospheric concentration of Hg by more than five-fold above natural levels (UN Environment Programme, 2018). Once released into the environment, inorganic Hg is transformed into methylmercury (MeHg)—its most toxic form—through biomethylation by anaerobic microorganisms present in water, and becomes part of the food chain (Bjerregaard et al., 2015). Dietary exposure is the main source in humans, especially fish and other marine species, and MeHg represents approximately 90% of total Hg (THg) in this food group (EFSA, 2012). Due to bioaccumulation and biomagnification processes, MeHg increasingly accumulates in the species at the highest trophic levels of aquatic food chains, hence predators, larger and/or long-lived species show the highest levels (Bjerregaard et al., 2015), such as swordfish, shark, or bluefin tuna (AESAN, 2019; Food and Drug Administration, 2012).

The main health effect of MeHg exposure among humans is neurotoxicity, with fetuses and children, especially in the early postnatal stage, being the most vulnerable groups due to the susceptibility of the developing neurological system (Afandiyev et al., 2010; ATSDR, 2024). In this population, apart from affecting neurobehavioral development, MeHg can cause intrauterine growth restriction, fetal malformations, or spontaneous miscarriage since it can pass through the placental membrane (Chen et al., 2022). In the general population, it has also been linked to hypertension, renal disorders, and immunological toxicity (ATSDR, 2024; Chen et al., 2022; Nordberg et al., 2007).

Spain is a region with a high fish consumption and, therefore, a high risk of Hg exposure. In 2012, the European Food Safety Authority (EFSA) highlighted that the highest MeHg exposure levels were found in the diets of Mediterranean countries (France, Greece, Italy, and Spain), compared with other European countries (EFSA, 2012). In 2023, the European Market Observatory for Fisheries and Aquaculture Products stated that Spain was the country of the European Union (EU) with the second highest fish consumption, after Portugal (data from 2021) (EUMOFA, 2023). According to the 2010-2011 Total Diet Study in the Valencian Community (a Mediterranean region in Spain), 8.5% of adults and 12.3% of children had a MeHg intake greater than the tolerable weekly intake (TWI) (Marín et al., 2017, 2018). Fish species that contributed the most to MeHg exposure in this population were swordfish (28%) and both fresh and canned tuna (27.6%) (Generalitat Valenciana, 2017).

Different official organizations issued recommendations regarding Hg intake through diet, targeted especially to protect vulnerable groups (AESAN, 2019; WHO, 2011). The advice is to limit the consumption of species highest in Hg levels, especially during pregnancy and childhood (EFSA, 2014). It is important to balance the health benefits and risks since the intake of n-3 long-chain polyunsaturated fatty acids (LCPUFA) and other nutrients through fish consumption appears beneficial for early neurodevelopment and protects against cardiovascular diseases in adults (Mozaffarian et al., 2006; EFSA, 2015). Besides, maximum Hg levels in foodstuffs are settled. In the EU, these levels were regulated in 2006 (EUR-Lex, 2006) and modified in 2023 to be between 1, 0.5, and 0.3 mg/kg, depending on the fish species (EUR-Lex, 2023).

Apart from the control of Hg exposure through fish consumption, worldwide efforts have been made to reduce anthropogenic emissions of this metal. An international agreement on Hg, the Minamata Convention, was signed in 2013 by 128 countries, although it did not come into force until 2017. The objective was that party nations take measures to reduce the use of Hg in certain products and industrial processes, as well as to curb its use in artisanal and small-scale gold mining. (UN Environment Programme, 2024). A review published in 2019 stated that, since 1990, Hg emissions from Europe and North America have decreased while total global anthropogenic Hg emissions have increased. They also examined global temporal trends in fish Hg concentrations between 1970 and 2017 and found an overall decrease in marine species from the North Atlantic but, on the other hand, an increase in species from the North Pacific (Grieb et al., 2019).

In Spain, very few studies have evaluated Hg temporal trends in fish species. González et al. measured Hg levels in fish species sold in the Tarragona region (Catalonia) in 2018 and compared the results to those obtained in previous years (1998 and 2013) by the same research group, finding no significant differences (González et al., 2021). Perelló et al. observed a 44% increase from 2008 to 2012 in Hg concentrations in fish consumed in Catalonia (Perelló et al., 2014). However, a decrease was observed in the levels measured in 2017, returning to the average Hg concentration in fish found in 2008 in this region (ACSA, 2017). Furthermore, few studies have been carried out worldwide to assess trends of Hg concentrations in fish over the last few years.

Traditionally, approaches estimating temporal trends for different categories, e.g., species of fish, were based on linear or semi-parametric regression, which usually imply analyses that are, to a greater extent, independent between groups/categories and, therefore, sensitive to unbalanced groups and groups with small sample sizes (Gelman et al., 2020; Gelman et al., 2006). Pooling information between groups through using random effects in linear and semi-parametric regression can be easily generalized considering structured random effects with covariances between category effects (Gelman et al., 2006). Most modern and robust techniques to estimate non-linear functions for different groups have recently appeared in the fields of advanced statistical modeling and machine learning based on Gaussian processes (GPs) (Rasmussen et al., 2006) for multiple correlated groups, either combining continuous and categorical kernel functions in a separate way (Kaufman et al., 2010) or using more recent kernels defined in the mixed domain of the continuous and categorical variables (Li et al., 2021).

Thus, the aim of this study was to evaluate the levels and temporal trends of Hg concentrations in fish consumed in the Valencian Community (Mediterranean region in Spain) differentiated by fish species, fish origin area, and type of Hg (THg and MeHg), by using the modern statistical model of multi-group Gaussian processes in comparison to traditional linear mixed-effects models.

2 Methodology

2.1 Study design

This work is a retrospective, descriptive, and analytical study of Hg levels in fish and fish products and their temporal trend in the period 2011-2021 in the Valencian Community. Fish samples analysed in this study were fish consumed in the Valencian Community, which is a region on the Mediterranean coast of Spain, although the origin of the fish might also be from other countries or continents.

2.2 Sample collection and Hg determination

Data on Hg levels in fish came from the fish sample collection as part of the Food Safety Monitoring Program from the Public Health Department of the Valencian Regional Government. The Valencian Community's Health Law 8/2018, which modifies Law 10/2014 (BOE, 2018), defines the Food Safety Plan as an instrument that includes interdepartmental actions leading to guaranteeing the health and safety of consumers in relation to food. One of the main objectives of this Plan is to reduce health risks for the population by organizing official controls to monitor biological and chemical hazards associated with food consumption. Regarding the control of contaminants, specifically Hg, as part of the Food Sanitary Monitoring Program, activities focus on the control of the maximum limits (established in the legislation) of this metal in fish, and in cases of non-compliance, taking the necessary measures (Generalitat Valenciana, 2021).

As part of the monitoring program, total Hg (THg) and MeHg concentrations were measured in fish and fish product samples (wet weight) that were collected in packaging, storage, and sale points by Public Health officers and analysed in the Public Health Laboratories of Alicante and Valencia. Different analytical methods were used, depending on the year and the laboratory, including Atomic Absorption Spectroscopy (AAS), Cold Vapour Atomic Absorption Spectrometry (CV-AAS), Atomic Absorption Spectroscopy-Advanced Mercury Analyzer (AAS-AMA-254), and Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

A total of 1070 Hg measurements were made in 839 samples. THg was measured in 799 samples, MeHg in 271 samples, and both compounds in 231 samples. In the case of MeHg, its concentration was only measured as of 2013. Hg levels were expressed in mg/kg and all measurements included an uncertainty value with a 95% confidence interval. Twenty-seven percent of measurements were below the quantification limit (BQL). BQL measurements per year are shown in **Table S1** in the Supplementary material. Since data were derived over a long sampling period, different analytical techniques were used and analyses were carried out in two different laboratories (Alicante or Valencia), the limits of quantification (LOQs) were not homogeneous, being 0.01 or 0.02 for MeHg and 0.02, 0.05, or 0.10 for THg.

The Sub-directorate for Food Safety of the Valencian Directorate-General for Public Health created a database that included different metal concentrations in food and complementary information. Data were collected following the "Chemical monitoring reporting guidance" by the EFSA (Brocca et al., 2022).

2.3 Study variables

To achieve the study objectives, in addition to THg and MeHg concentrations, the following covariates were added from the database described above: EFSA Product code (species or types of fish/fish product sampled), product treatment (canning, freezing, pasteurization, smoking, cooking, processed, unprocessed), fishery area of origin (sea/ocean where the product was fished), year of sampling (year of sampling matches year of Hg content analysis).

Due to the high number of different categories of the "Species of fish" variable (collected by EFSA Product code) in the original database, the original categories were distributed into six easier-to-analyse-and-interpret groups: canned tuna (n=147), fresh tuna (n=114), lean fish (n=69), swordfish (n=348), other oily fish (n=71), and others (n=321). Three shark samples were included in the "Swordfish" group due to similarities between both species (size, predators, high in the trophic level…). The fish group "Others" included cephalopods (n=136), crustaceans (n=57), mollusks (n=56), and other canned and/or processed fish (n=72).

In the same way, the original categories of the "Fishery area of origin" variable were grouped as follows: Atlantic (n=297), Indian (n=80), Mediterranean (n=93), Pacific (n=183), and Unknown (n=417). Six samples from the North Sea were included in the "Atlantic" group.

2.4 Data descriptive analysis

A descriptive analysis of THg and MeHg concentrations was conducted in all the samples, as well as according to the year of sampling, the fish group, and the area of origin. For this purpose, central tendency measures (mean, median) and dispersion measures (standard deviation, 25-75 percentiles, and range) were calculated. Hg concentrations, both THg and MeHg, cannot have negative values but rather be positive continuous variables with a distribution skewed at low values close to zero. They are expected to follow a lognormal distribution (i.e., the logarithm of the concentrations is normally distributed). Hg levels were compared to the maximum limit values in fish meat specified by current European regulations.

2.5 Statistical modeling

The aim was to formulate a probabilistic model to estimate temporal trends in Hg levels differentiated by categories and regions of origin of the fish and Hg measurement type. The dataset available was unbalanced between species and fish origin areas, where the sample size for some groups may be small. Some correlation was expected between the Hg levels of certain categories of fish; such information could be useful, especially for those groups with fewer observations. Moreover, for some samples, THg concentrations may have been missing but MeHg was available. Additionally, some samples may have undetected observations since they had BQL Hg concentrations. Also, for some samples, the LOQ may be unknown for THg or MeHg measurements, or both.

To address these challenging issues robustly, we proposed a probabilistic (Bayesian) modeling framework that allowed joint modeling of these issues: A multi-group GP model predictor function to account for correlations between groups and hierarchical prior probability distributions to impute missing and BQL data. In addition to the multi-group GP model predictor function, we also implemented, for comparison purposes, the well-known mixed-effects-based multi-level linear model.

2.5.1 Multi-level linear model

The underlying predictor function consist in a multi-level linear model based on mixed-effects (Gelman et al., 2006), where different levels/groups are defined for fish categories, origin regions, and type of Hg while pooling information between groups as random effects from a common probability distribution:

$$y_i | t_i, x_i \quad \sim \mathrm{Lognormal}(f(t_i, x_i), \sigma^2),$$

$$f_i(t_i, x_i) \quad = a_{x_i} + b_{x_i} t_i,$$

$$\begin{aligned} a_{x_i} &\quad \sim \mathrm{Normal}(\mu_a, \sigma_a^2), \\ b_{x_i} &\quad \sim \mathrm{Normal}(\mu_b, \sigma_b^2), \end{aligned}$$

where $y_i > 0$ is the Hg concentration of sample i ($i = 1, \ldots, n$, where n is the total number samples) belonging to group x and time point t. Group x results from crossing factors of fish species $s \in S = \{\text{Canned tuna, Fresh tuna, Lean fish, Swordfish, Other oily fish, Others}\}$, origin region $r \in R = \{\text{Atlantic, Indian, Mediterranean, Pacific, Freshwater, Unknown}\}$, and type of Hg $c \in C = \{\text{THg, MeHg}\}$, so that $x \in \mathcal{X} = S \times R \times C$. Observation y_i is log-normally distributed with mean predictor function $f(t_i, x_i)$ and noise σ. Function f is a multi-level linear model where a_x and b_x are random effects of groups x, for intercepts and linear coefficients, respectively. Intercept parameters a_x represents Hg base levels per group x and coefficients b_x the time trends per group x. Parameters σ, σ_a, and σ_b are the hyperparameters of the model and appropriate prior distributions for these can be found in Appendix A.

2.5.2 Multi-group Gaussian process model

The underlying predictor function is centered on a modern and robust method based on multi-group GPs, recently proposed by Li et al. (Li et al., 2021). GPs (Neal, 1997; Rasmussen et al., 2006) are non-parametric probabilistic models for functions that have become a popular approach for time series functions, as well as for spatial or spatiotemporal functions and multidimensional functions in general (Banerjee et al., 2014; Diggle, 2013; Riutort-Mayol et al., 2020). GPs are reliable and powerful models as they may infer and exploit the covariance structure of data to estimate the underlying functions and make predictions. The recent approach of multi-group GPs makes this a robust method to estimate function deviations for different groups by estimating covariances between observations within and between groups, which allows information to be shared between groups. This may be useful to estimate robust functions for unbalanced datasets with few observations in some of the groups, or groups that may have similar behaviour, as is the case of our dataset. The basics of this multi-group GP model are specified below:

$$y_i|t_i, x_i \sim \text{Lognormal}(f(t_i, x_i), \sigma^2),$$

where function f is a GP function,

$$f(t, x) \sim \text{GP}\left(0, k\left(t_i, t_j, x_i, x_j\right)\right), \quad (1)$$

with the multi-group squared exponential covariance function k proposed by Li et al. (30),

$$k\left(t_i, t_j, x_i, x_j\right) = \frac{\sigma_{GP}^2}{\left(a^2 d_{x_i,x_j}^2 + 1\right)^{(p/2)}} \exp\left(\frac{-\left(t_i - t_j\right)^2}{\ell^2\left(a^2 d_{x_i,x_j}^2 + 1\right)}\right),$$

where ℓ is the shared length-scale among pairs (i, j) of observations, observation i of group $x_i \in \mathcal{X}$ and observation j of group $x_j \in \mathcal{X}$. Length-scale represents the grade of decay in the correlation between pairs of observations as a function of the distance between them in the space of the input variables, e.g., the time variable. The parameter $a > 0$ is the overall similarity scale between groups, and d_{x_i,x_j} is the specific similarity distance between pairs of groups $\left(x_i, x_j\right)$, where the complete set of distances $D = \left[d_{x_i,x_j}\right] \in \mathbb{R}^{q\times q}$, where q is the number of groups in $\mathcal{X}$, forms a valid distance metric between the groups (Li et al., 2021). $\sigma_{GP} > 0$ is the marginal variance of the GP functions and p is the dimension of the input space. In short, the multi-group kernel scales the length-scale for observations within groups and between groups, allowing estimating different covariance structures within groups and between observations belonging to different groups. This kernel differs from the common separate GP approach in that in the latter the groups are independent. The length-scale between groups is changed by multiplying ℓ by the corresponding similarity distance d_{x_i,x_j} between the groups x_i and x_j. Parameters σ, ℓ, σ_{GP}, a, and d_{x_i,x_j} are the model hyperparameters and appropriate prior distributios for them can be found in Appendix A.

The defining property of a GP is that the collection of function values $\{f(t_i, x_i)\}_1^n$ follows a multivariate normal with an arbitrary mean function $\mu(t_i, x_i)$ and a variance-covariance matrix K,

$$\{f(t_1, x_1), \dots, f(t_n, x_n)\} \sim \mathrm{Normal}(\mu, K),$$

where each element (i, j) of matrix K is given by the covariance function,

$$K_{ij} = k\big(t_i, t_j, x_i, x_j\big).$$

For a deeper explanation of the multi-group GP, see the work by Li et al. (2021) (Li et al., 2021) and for regular GPs see Rasmussen and Williams (2006) (Rasmussen et al., 2006).

2.5.3 Imputation of missing and below quantification limit observations

Bayesian modeling (Ntzoufras, 2008) allows missing observations to be inferred by specifying prior probability distributions for them and defining them as parameters to be estimated in the model. Information on the LOQ was established as the upper limit of prediction for those BQL observations by defining a bounded prior distribution for each of them. And, since hierarchical modeling (Ntzoufras, 2008) is natural in Bayesian modeling, the missing LOQ values can also be inferred by defining a hierarchical prior distribution over them. Finally, since the observations of THg and MeHg, as well as their LOQs, were highly intercorrelated, multivariate prior distributions can be defined such that they share information.

Thus, multivariate prior distributions can be defined for pairs of THg, $y_{i|c_i=\text{THg}}$ (observations y_i belonging to group $c_i = \text{THg}$), and MeHg concentrations, $y_{i|c_i=\text{THg}}$ (observations y_i belonging to group $c_i = \text{MeHg}$), of sample i so that they share information,

$$\big(y_{i|c_i=\text{THg}}, y_{i|c_i=\text{MeHg}}\big) \sim \mathrm{Lognormal}\big(\mathbf{0}, \Sigma_y\big), \tag{2}$$

with a mean vector of zero (one could also define a function for the mean of this distribution) and a covariance matrix, Σ_y, between pairs of THg and MeHg values of sample i, which is shared across samples. For those observations of THg and MeHg that are missing, denoted by $y^*_{i|c_i=\text{THg}}$ and $y^*_{i|c_i=\text{MeHg}}$, respectively, additional prior distributions, such as

$$\begin{aligned} y^*_{i|c_i=\text{THg}} &\sim \mathrm{Lognormal}(0, \delta) \\ y^*_{i|c_i=\text{MeHg}} &\sim \mathrm{Lognormal}(0, \tau), \end{aligned} \tag{3}$$

can be defined for them as parameters and inferred from the model, where δ and τ are the variance hyperparameters of these priors shared across samples i (Note that the y^* notation is intended to denote a missing Hg concentration among the complete set of Hg concentrations y).

Furthermore, for those missing observations of THg and MeHg that are BQL observations, bounded prior distributions can be defined for them as follows:

$$\begin{aligned} y^*_{i|c_i=\text{THg}} &\sim p\left(0, LOQ_{i|c_i=\text{THg}}\right), \\ y^*_{i|c_i=\text{MeHg}} &\sim p\left(0, LOQ_{i|c_i=\text{MeHg}}\right), \end{aligned}$$

where p represents a probability distribution with the lower bound of zero and the upper bound of $LOQ_{i|c_i=\text{THg}}$ and $LOQ_{i|c_i=\text{MeHg}}$, respectively. As it is slightly more likely that the predictions of BQL observations be closer to the upper bound $LOQ_{i|c_i=\text{THg}}$ or $LOQ_{i|c_i=\text{MeHg}}$ than to the lower bound of zero, the probability distribution p is defined as a Beta distribution scaled by the LOQs as follows:

$$\begin{aligned} p\left(0, LOQ_{i|c_i=\text{THg}}\right) &= LOQ_{i|c_i=\text{THg}} \cdot \text{Beta}(2,1), \\ p\left(0, LOQ_{i|c_i=\text{MeHg}}\right) &= LOQ_{i|c_i=\text{MeHg}} \cdot \text{Beta}(2,1). \end{aligned}$$

Similarly to THg and MeHg concentrations, a multivariate prior distribution can be defined for the LOQs, $LOQ_{i|c_i=\text{THg}}$ and $LOQ_{i|c_i=\text{MeHg}}$, of THg and MeHg concentrations, respectively, of sample i,

$$\left(LOQ_{i|c_i=\text{THg}}, LOQ_{i|c_i=\text{MeHg}}\right) \sim \text{Normal}\left(\mu, \Sigma_{LOQ}\right). \tag{4}$$

For those LOQs that are missing, denoted by $LOQ^*_{i|c_i=\text{THg}}$ and $LOQ^*_{i|c_i=\text{MeHg}}$, respectively, additional prior distributions, such as

$$\begin{aligned} LOQ^*_{i|c_i=\text{THg}} &\sim \text{Uniform}(0,0.1) \\ LOQ^*_{i|c_i=\text{MeHg}} &\sim \text{Uniform}(0,0.02), \end{aligned}$$

can be defined for them as parameters and inferred from the model. Note that the largest LOQ values for THg and MeHg are, in our case study, 0.10 and 0.02, respectively. Covariance matrices Σ_y and Σ_{LOQ} and the variance parameters δ and τ are the model hyperparameters and details about appropriate prior distributions for them can be found in Appendix A. Additional prior information about the values to be predicted can be encoded through these prior distributions.

In a Bayesian framework, complete posterior probability distributions are estimated for the missing and BQL observations in a fully probabilistic fashion, where uncertainties are fully and adequately propagated across model parameters and assumptions, yielding a reliable estimate of the parameters of interest (Ntzoufras, 2008). These posterior distribution estimates will be more concentrated or more dispersed depending on the correlation between the parameters and the information provided by the data. This contrasts with traditional approaches where missing observations are often discarded or estimated in advance, often using point estimates or fixed imputations, which may have an excessive influence on the model and, thus, biasing parameter estimates.

2.5.4 Model inference, checking, and selection

The models were formulated from a Bayesian perspective and inference was performed using the Hamiltonian Monte Carlo sampling method in the Stan probabilistic programming software (Carpenter et al., 2017; Stan Development Team, 2017) to estimate the marginal posterior probability distributions of every parameter of interest. Brief details regarding the model inference, checking, and selection performed in the case study are included in Appendix B.

3 Results

3.1 Description of THg and MeHg by fish groups

Table 1 and **Figures S1** and **S2** (Supplementary material) show descriptive statistics and boxplots of THg and MeHg concentrations by fish species, respectively. The swordfish group exhibited the highest levels (median THg: 0.76 mg/kg), followed by fresh tuna (0.46 mg/kg) and, in third place, canned tuna (0.22 mg/kg). Lean fish was the following group, with a median THg of 0.13 mg/kg. Levels in the other groups were significantly lower than in these aforementioned groups.

Swordfish presented a wide range of concentrations (THg: 0.11-2.70 mg/kg), and 28.7% of swordfish samples (n=100, which included the three shark samples) exceeded the permitted EU levels (1 mg per kg of weight). MeHg levels in this group presented a median value of 0.61 mg/kg. Fresh tuna also had a wide range of concentrations (THg: 0.10-1.30 mg/kg). In this case, 2.6% of samples (n=3) exceeded the maximum levels of 1 mg/kg. A single sample of canned tuna exceeded this limit (1.02 mg/kg). In the lean fish group, one sample of Pink cusk-eel (Genypterus spp.) exceeded the 1 mg/kg limit for this species (THg: 1.31, MeHg: 1.12 mg/kg).

3.2 Distribution of swordfish Hg levels by area of origin

Due to the limited dataset, the descriptive analysis of the sample's area of origin was conducted on the swordfish group as it contained a significant number of samples per origin area and, also, it was the group with higher Hg concentrations. This descriptive analysis is shown in **Table 2**. For marine swordfish samples of known origin, those from the Indian Ocean (n=24) showed the highest levels (median THg: 1.13 mg/kg), followed by the Atlantic Ocean (n=93) (median THg: 0.84 mg/kg). Samples from the Mediterranean Sea had a median THg of 0.59 mg/kg, however, it should be noted that there were only three samples with this origin. Swordfish from the Pacific Ocean (n=73) showed the lowest THg levels (median THg: 0.53).

3.3 Imputation of missing and below quantification limit observations

Posterior distributions were estimated for THg concentrations in 34 samples where only MeHg had been measured, BQL observations in 252 samples of THg and 33 of MeHg concentrations, and LOQ values in 40 THg samples (5 in 2013 and 35 in 2014) and in 2 MeHg samples in 2014, from the proposed model as explained in Section 2.5.3. **Figure C.1** in Appendix C shows the estimated posterior distributions of the 34 missing THg concentrations along with the corresponding observed MeHg concentrations, where a high correlation between the THg and MeHg values can be seen, which is in line with the high correlation of almost 0.9 found between THg and MeHg concentrations in the estimation of their multivariate distribution in equation (2). **Figure C.2** shows the estimated posterior distributions for THg and MeHg BQL concentrations along with their corresponding LOQs. These estimated posteriors were mostly concentrated close to the upper-bound LOQ as expected, except for the samples belonging to the categories Other oily fish and Others and with a LOQ of 0.1, where the estimated posteriors were more centered. **Figure C.3** shows the estimated posterior distributions for THg and MeHg BQL concentrations along with the estimates of their corresponding LOQs. The estimated posterior of the LOQs were always larger than the estimated Hg concentrations, since they were BQL observations.

3.4 Time trends by fish group and Hg type

Figure 1 shows the estimated trends of THg and MeHg concentrations per fish species. The multi-group GP model detected a non-linear pattern in THg and MeHg levels in swordfish throughout the study period, where levels tended to decrease around 0.5 mg/kg from 2011 to 2016. However, from 2016 to 2021, the tendency changed and levels increased to approximately those at the start of the period.

In the latter part of the study period, there were very few data points for fresh tuna; therefore, the resulting trend estimated by the multi-level linear model would not be sufficiently robust. However, the multi-group GP model locally adapted trend estimates over the period and reliably estimated a decreasing trend from 2011 to 2016 for both THg and MeHg. It also estimated an ascending trend between 2016-2021, although with less evidence due to the lower number of samples analysed.

For canned tuna, the multi-level linear model estimated an overall decreasing trend of Hg levels throughout the study period, although the number of observations in the second half of the period was low, especially for MeHg. In this case, the multi-group GP model was more reliable as it estimated a clear decreasing trend between 2013 and 2016 for both THg and MeHg. From 2016 to 2021, it also estimated a descendent trend for THg but showed less evidence due to the low number of samples.

Finally, a slight decrease throughout the period was observed for the other species.

3.5 Swordfish time trends by area of origin

In general terms, THg and MeHg concentrations in swordfish showed a significant descending trend in samples from the Indian Ocean, although this trend somewhat softened when using the multi-group GP model (**Figure 2**). Those from the Pacific Ocean showed an increasing trend in the period 2011-2021. However, THg levels in Atlantic Ocean swordfish maintained a horizontal trend, and MeHg levels showed a slightly ascending trend using the linear model, while the multi-group model indicated that levels tended to be stable throughout the period. In the case of swordfish from the Mediterranean Sea, models showed mild variations in THg levels, considering that there were only three samples from this origin.

4 Discussion

The current study assesses THg and MeHg concentrations and their trends in fish intended for consumption over a decade in a Spanish Mediterranean region, a country with high fish consumption. As far as we know, this is one of the very few studies that have been carried out worldwide since the entry into force of the Minamata Convention. Temporal analyses of Hg in fish species consumed by humans are an important aspect of the effectiveness evaluation of Minamata Convention, and monitoring efforts must include the measurement of potentially confounding factors, such as fish body size, fish trophic level, ecosystem shifts, atmospheric mercury data or the influence of climate change) (Grieb et al., 2019). Although our study does not aim to evaluate the impact of the Minamata Convention, it measures Hg trends in a large number of fish samples from different global origins.

Swordfish was the group that showed the highest Hg levels (median THg: 0.76 mg/kg), followed by fresh tuna, canned tuna, lean fish, and other oily fish. Almost 30% of the swordfish samples studied presented levels over the legal limit established by the EU for this species. This is an issue of concern since swordfish is one of the most commercially valuable species in the EU market (EUMOFA, 2023) and has a particularly high consumption rate in Spain (Torres-Escribano et al., 2010). López-González et al. studied Hg exposure among a cohort of Valencian adolescents and demonstrated a clear association between fish consumption and higher concentrations of THg in hair samples, with swordfish being the species with the strongest association, while canned tuna was the main absolute contributor due to the frequency of consumption in this population (López-González et al., 2023).

In general, a decreasing trend of Hg concentrations was found in the different fish species in the first half of the study period, between 2011-2016. In the second half of the study period, between 2016-2021, there was a significant lack of samples in all the species studied except swordfish, which may hinder the interpretation of the results for those species. In the longitudinal study of Hg exposure in a Valencian cohort mentioned above, hair THg concentrations at 11 years of age (2015-2016) represented a reduction of around 69% with respect to that estimated at childbirth and a reduction of around 27% with respect to the concentration at 4 years of age (2008-2009) (López-González et al., 2023).

Throughout the study period, Hg concentrations in swordfish showed a significant decrease between 2011 and 2016; however, during the second half of the study period, from 2016 to 2021, levels increased, returning to their initial point in 2011. Nevertheless, unequal Hg concentrations according to their origin were observed over time. Swordfish samples from the Indian Ocean showed the highest Hg levels and a significant decreasing trend throughout the period. On the other hand, Pacific Ocean swordfish presented the lowest levels among the different fish origin areas and a significant increasing trend, especially in the last part of the study period. This was probably related to the general increasing trend observed in swordfish from 2016 to 2021, considering the lack of Indian Ocean samples from these years, in contrast with the increasing number of Pacific Ocean samples. These results align with the overall increase in Hg levels in fish from the Pacific Ocean found by Grieb et al. (Grieb et al., 2019). Swordfish Hg levels from the Atlantic Ocean maintained certain stability throughout the period.

Both fresh and canned tuna showed a descending trend in the first half of the period. Total Hg levels in fresh tuna increased between 2016-2021, while a descending trend was observed in canned tuna.

In previous studies, Hg levels in fresh tuna seemed to be ascending or remaining stable (Drevnick et al., 2017; Médieu et al., 2021). Drevnick et al. (Drevnick el al., 2017) observed an increasing trend in two tuna species from the North Pacific Ocean during the period 1971-2008. Meanwhile, Médieu et al. studied Hg trends in three tuna species from the South Western Pacific Ocean and observed a decadal stability (2001-2018), with significant inter-annual variability related to the variability in tuna lengths, biogeochemical and physical processes, and ecological parameters (Médieu et al., 2021).

Therefore, canned tuna is the only fish group seen to have an overall significant descending trend in THg levels in the present study, although the trend observed in the second part of the study period should interpreted with caution, since the number of observations between 2016-2021 was low.

As highlighted in other studies, predator and large species show the highest Hg concentrations (Ahmad et al., 2015; Chacón et al., 2016; Mozaffarian et al., 2006; Torres-Escribano et al., 2010). In the 2017 Total Diet Study in Catalonia (Spain) (ACSA, 2017), the highest mean Hg levels were observed in swordfish (0.86 and 0.78 µg/g of THg and MeHg, respectively) and fresh tuna (0.43 and 0.35 µg/g of THg and MeHg, respectively), which were very similar to the THg and MeHg concentrations found in our study in both species. Mozzafarian et al. (Mozaffarian et al., 2006) reported an average Hg concentration of 0.98 µg/g in swordfish and 0.99 µg/g in shark based on data from the

US Department of Agriculture (USDA) and the Food and Drug Administration (FDA); in our study, the mean THg content in swordfish was 0.86 mg/kg. Rodrigues et al. evaluated Hg concentrations in South Atlantic swordfish from the coast of Brazil. Of a total of 697 swordfish analysed, 1.6% had Hg concentrations above 1 mg/kg, 60.4% were between 0.5 and 1.0 mg/kg, and 38% were below 0.5 mg/kg (Rodrigues et al., 2013). We obtained a range of 0.11-2.70 mg/kg and a median of 0.84 mg/kg in samples from the Atlantic. Jinadasa et al. (Jinadasa et al., 2018) studied Hg content in commercial swordfish collected from the Indian Ocean (n=75), finding a wide range (0.07-4.30 mg/kg) and 13.3% of measurements exceeded the 1 mg/kg limit for Hg. Mean levels were 0.62 mg/kg, whereas, in our study, they were 1.11 for THg in swordfish from the Indian Ocean. Esposito et al. studied THg content in 220 commercial swordfish samples from different FAO fishing areas imported into Italy between 2014 and 2017. Among the samples investigated, 17.7% exceeded the maximum Hg limit established by EU legislation, in comparison with 28.7% of samples in the present study. They obtained a mean value of 0.71 mg/kg, lower than that observed in our study (0.86 mg/kg). Samples from the Western Indian Ocean (n=13) had the highest median (0.90 mg/kg), while samples from the North-East Atlantic Ocean (n=23) had the lowest (0.32 mg/kg) (Esposito et al., 2018).

Some regional studies have examined mercury trends in fish but most of them are not recent; **Table 3** summarizes the results from these studies. During the period 1969-2012, Bonito et al. studied Hg trends in multiple species from a wide range of origins and found a significant overall decreasing trend (Bonito et al., 2016). Zhang et al. found a 70% decrease in Hg levels from 1980 to 2014 in multiple species consumed in China (Zhang et al., 2022). In more recent studies, Kammann et al. observed a significant increase in Hg levels in dab (*Limanda limanda),* a lean fish species, samples from the North Sea in the period 1995-2020, with an annual percentage change of 1.4% (41% increase within 25 years) (Kammann et al., 2023). Morris et al. assessed temporal trends of THg concentrations in Arctic biota as part of the 2021 Arctic Monitoring and Assessment Programme Mercury Assessment and observed a recent 20-year (1999-2021) increasing trend in sculpin from Greenland, which is a predatory fish species, and Atlantic cod from the northwest Faroe Islands and northern Norway (Morris et al., 2022). Rudershausen et al. studied THg trends in blue marlin, considered one of the largest fish species, from the Atlantic Ocean (North Carolina) and obtained an interdecadal decline of 45% between the 1970s and late 1990s. They observed that concentrations remained stable during the first two decades of this century (Rudershausen et al., 2023). Bank et al. assessed Hg concentrations in Northeast Arctic cod from the Barents Sea sampled between 1994-2021 and observed low and consistently stable concentrations over the period, despite a significant increase in Barents Sea temperature and a sharp decline in regional sea ice extent, suggesting climate change dynamics did not translate into major increases or decreases in Hg bioaccumulation in the fish species studied (Bank et al., 2023). Médieu et al. assembled THg concentrations in tuna species in six regions of the global ocean from 1971 to 2022, with variable periods depending on the region. Significant temporal trends of Hg levels were only evidenced in skipjack in the late 1990s in the northwestern Pacific, likely resulting from concomitant increasing Asian mercury emissions. However, in all the other regions, stable long-term trends of tuna Hg concentrations were found, which reflect the inertia of surface ocean Hg with respect to declining emissions, as it is supplied by legacy Hg that accumulated in the subsurface ocean over centuries (Médieu et al., 2024) Therefore, recent evidence suggests an increasing or stable trend in worldwide Hg concentrations in fish.

This research is not devoid of limitations. In over 27% of THg measurements in swordfish, the sample's origin was unknown (75 out of 269); thus, the analysis of this variable should be interpreted with caution. Furthermore, Hg measurements in the second half of the study period were gathered in swordfish. Therefore, there is a significant lack of samples from other species, resulting in unbalanced Hg measurements between species and with small sample sizes in some of the species. However, as study strengths, this issue has been addressed using Bayesian Gaussian processes for multiple groups (Multi-group GP) which estimates a complete covariance matrix between observations so that correlation between groups are used to estimate the treatment/group effects, which can be especially useful for those groups with fewer observations since they gather information from correlated groups. Other methodologies, such as semi-parametric or mixed-effects linear regression, do not estimate and use a full covariance matrix between observations, but may instead correlate model coefficients, like intercepts, time trends, or spline coefficients, to attempt to share information between groups.

Another frequent limitation of the assessment of metal concentrations in observational studies is that some measurements might be below the LOQ and others just be missing. Our dataset contained about 3.5% missing THg concentrations and 26.6% BQL observations, as well as some missing LOQs. With traditional frequentist analysis it is often necessary to discard such observations, thus reducing the study sample size, or to estimate them in advance, often using point estimates or fixed imputations, which may have an excessive influence on the model. Within the Bayesian model framework developed in this study, those missing and BQL observations were naturally considered parameters and estimated in a fully probabilistic fashion, where uncertainties were fully and adequately propagated between parameters and model assumptions, obtaining full predictive posterior distributions. By defining upper-bounded prior distributions for the BQL observations, their estimated posterior distributions were limited by the LOQ. Furthermore, the accuracy of the estimates was improved by modelling the covariance structure between the data.

As discussed throughout the paper, a multi-group GP model has some advantages compared to traditional semi-parametric or mixed-effects linear regression for modeling the covariance structure in the data and using it for prediction or imputation. Furthermore, in this work, we evaluated the predictive performance and model calibration of both models, and the multi-group GP model showed significantly better results than the linear mixed-effects model.

Additionally, this work approximates recent Hg trends, both THg and MeHg, while very few studies have been carried out worldwide to assess trends of Hg levels in fish in the last years, and, to our knowledge, there are no studies evaluating this in our region. Besides, even though the results come from a single Spanish region, there is a high number of measurements over a long period of time in fish samples that are representative of fish consumption in our region. Furthermore, the globalization of the fish consumption market, as reflected in the origin of the samples, provides data on a general situation of mercury concentration in fish sold, and consumed, internationally.

6 Conclusions

In the present study, among fish consumed in the Valencian Community, swordfish stands out as the species with the highest Hg concentration. In the period studied, near a third of swordfish samples exceeded the European limit values, and no clear trend was observed. Hg levels in swordfish tended to decrease from 2011 to 2016, but then showed an increasing trend from 2016 to 2021. However, this ascending trend in the second half of the study period appears to be related to the increasing trend observed in swordfish samples from the Pacific Ocean given the non-homogenous distribution of swordfish samples by origin during the studied period. Furthermore, decreasing trends for most of the studied species were observed from 2011 to 2016.

Our findings on Hg levels in different species of commercially sold fish in a high-consumption country, along with their trends, could be useful for guiding the Food Safety Monitoring Program, as well as for developing local fish consumption recommendations.

Appendices

A. Prior distributions for model hyperparameters

Multi-level linear model.

Appropriate prior distributions for the hyperparameters σ, σ_a, and σ_b of the multi-level linear model in Section 2.5.1 can be a positive truncated zero-mean normal distribution with an appropriate variance, for example, in our case study, a variance of 10:

$$\sigma, \sigma_a, \sigma_b \quad \sim \mathrm{Normal}^+(0, \sigma^2), \text{ with } \sigma^2 = 10.$$

Alternatively, Gamma distributions with appropriate parameters could also be used.

Multi-group GP model.

Appropriate prior distributions for hyperparameters σ, ℓ, σ_{GP}, a, and group similarity distances d_{x_i, x_j} of the multi-group GP model in Section 2.5.2 can be the following:

$$\begin{aligned} \sigma, \sigma_{GP} &\sim \mathrm{Normal}^+(0{,}10), \\ \ell &\sim \mathrm{InverseGamma}(5{,}15), \\ a &\sim \mathrm{Exponentila}(0.2), \\ d_{x_i, x_j} &\sim \mathrm{Beta}(1{,}1). \end{aligned}$$

It is important to mention that similarity distances between groups, $D = \left[d_{x_i, x_j}\right]$, have to fulfill the requirements to form a valid distance matrix such that the multi-group covariance function k of the GP model in equation (1) is positive definite, so the constraints for the distances need to be defined accordingly. Alternatively, Min-plus matrix multiplication can be used to update an initial unconstrained distance matrix to become a valid distance matrix (Zwick, 2002).

Imputation of missing and below quantification limit observations.

For the covariance matrices Σ_y and Σ_{LOQ} in equations (2) and (4), Wishart distributions (Wishart, 1928) can be specified. Another option for specifying a prior distribution for a covariance matrix is to define a LKJ distribution (Lewandowski et al., 2009) for the correlation matrix and scale it by the coefficient variances. Details on how to implement the Wishart and LKJ distributions for covariance matrices in probabilistic modeling can be found in https://mc-stan.org/docs/functions-reference/covariance_matrix_distributions.html and https://mc-stan.org/docs/stan-users-guide/regression.html#multivariate-hierarchical-priors.section, respectively.

Appropriate prior distributions for the variance hyperparameters δ and τ in equations (3) can be the following:

$$\delta, \tau \quad \sim \mathrm{Normal}(0, \sigma^2),$$

where $\sigma^2 = 10$ must be an appropriate large enough variance.

B. Model Inference, checking, and selection

The models were formulated from a Bayesian perspective and adjusted using sampling methods in the Stan probabilistic programming software (Carpenter et al., 2017; Stan Development Team, 2017). Stan software uses the Hamiltonian Monte Carlo sampling method (Brooks et al., 2012) to estimate the marginal posterior probability distributions of every parameter of interest. Four simulation chains with different initial values were launched. The convergence of the simulation chains was evaluated by the split-Rhat convergence diagnosis and the effective sample size of the chains (Gelman et al., 2021; Vehtari et al., 2021). In our case, and for both models, a split-Rhat value lower than 1.05 was obtained for all parameter simulation chains indicating good mixing of simulated chains. The convergence of the simulation chains indicates that the samples do come from the posterior distribution and that the model is correctly specified without confusion or identifiability problems between parameters.

For model checking, leave-one-out probability integral transformation (LOO-PIT) can be used to assess whether the model predictive distributions are calibrated, that is, they are describing the model predictive uncertainty well. In the case of good calibration of predictive distribution, LOO-PIT values are uniformly distributed (Gelfand, 1992; Gelman et al., 2021). To compute the LOO-PIT values, the R-package loo (Vehtari et al., 2017) was used, which performs efficient approximate leave-one-out cross-validation. **Figure B.1** shows the uniform Q-Q plot of the LOO-PIT values obtained in our case study for both the multi-group GP model (**Figure B.1-left**) and the multi-level linear model (**Figure B.1-right**). The distribution of the LOO-PIT values for the multi-group GP model was closer to uniform than for the multi-level linear model, indicating that the model better captures the main patterns of variability in the data. The multi-level linear model shows greater model underdispersion than the multi-group GP model, as the deviation from uniformity in the tails was greater.

For model selection, leave-one-out expected log predictive density (LOO-ELPD) and root mean square error (LOO-RMSE) can be compared between the multi-level linear model and the multi-group GP model. While the RMSE evaluates, by averaging over all checking observations, how far new data is from the model by using the distance (error) between the actual observation and the predictive mean, the ELPD evaluates how far new data is from the model while taking the posterior uncertainties into account (Magnusson et al., 2019; Vehtari et al., 2012). **Figures B.2** and **B.3** show the RMSE and ELPD for both models, the multi-level linear and the multi-group GP models. The RMSE and ELPD for the multi-group GP model are significantly lower and larger, respectively, indicating better performance than the multi-level linear model.

C. Estimated posterior probability distributions of missing, BQL, and LOQ observations

Plots of estimated posterior probability distributions of missing THg concentrations (**Figure C.1**), THg and MeHg BQL concentrations (**Figure C.2**), and THg and MeHg LOQ values (**Figure C.3**).

Acknowledgements

The authors are grateful to all the Food Safety Section technicians from the Directorate-General for Public Health (Conselleria de Sanitat, Valencia) who collaborated in sampling collection and analytical measurement.

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Tables

Table 1. Descriptive statistics of THg and MeHg concentrations (mg/kg) by fish groups.

Hg levels/ Fish group	Total		Swordfish		Fresh tuna		Canned tuna		Lean fish		Other oily fish		Others	
	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg
n (observed)	547	238	266	78	76	36	73	62	30	12	28	8	74	42
n (BQL)	252	33	3	1	1	1	12	0	22	5	35	0	179	26
Mean± SD	0.56± 0.50	0.36± 0.41	0.86± 0.52	0.71± 0.50	0.48± 0.26	0.31± 0.17	0.27± 0.20	0.21± 0.17	0.21± 0.27	0.16± 0.31	0.12± 0.12	0.02± 0.01	0.15± 0.17	0.09± 0.16
Median (p25, p75)	0.42 (0.17, 0.79)	0.23 (0.07, 0.51)	0.76 (0.47, 1.17)	0.61 (0.30, 1.00)	0.46 (0.30, 0.65)	0.29 (0.17, 0.43)	0.22 (0.12, 0.35)	0.16 (0.09, 0.28)	0.13 (0.10, 0.19)	0.07 (0.04, 0.09)	0.07 (0.06, 0.15)	0.02 (0.02, 0.03)	0.09 (0.05, 0.20)	0.04 (0.02, 0.05)
Range	0.02-2.70	0.01-2.23	0.11-2.70	0.02-2.23	0.10-1.30	0.03-0.76	0.03-1.02	0.02-0.75	0.03-1.31	0.03-1.12	0.03-0.59	0.01-0.04	0.02-0.85	0.01-0.75

n= number of measurements. (THg, n=547; MeHg, n=238). BQL= below quantification limit. Mean, standard deviation (SD), median, 25th and 75th percentiles (p25 and p75), minimum and maximum (range) levels were measured.

Table 2. THg and MeHg (mg/kg) concentrations in swordfish by area of origin.

Hg levels/ Origin	Total		Atlantic		Indian		Mediterranean		Pacific		Unknown	
	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg	THg	MeHg
n(observed)	266	78	93	25	24	5	3	0	73	26	73	22
n (BQL)	3	1	0	0	0	0	0		1	0	2	1
Mean± SD	0.86± 0.52	0.71± 0.50	0.90± 0.47	0.81± 0.54	1.11± 0.37	1.04± 0.20	0.76± 0.65	-	0.65± 0.39	0.54± 0.35	0.94± 0.67	0.73± 0.58
Median (p25, p75)	0.76 (0.47, 1.17)	0.61 (0.30, 1.00)	0.84 (0.56, 1.17)	0.72 (0.49, 1.08)	1.13 (0.79, 1.42)	1.08 (1.01, 1.14)	0.59 (0.40, 1.03)	-	0.53 (0.35, 0.83)	0.50 (0.29, 0.62)	0.70 (0.45, 1.35)	0.62 (0.27, 0.94)
Range	0.11-2.70	0.02-2.23	0.11-2.70	0.02-2.23	0.27-1.61	0.72-1.27	0.21-1.47	-	0.20-1.80	0.06-1.71	0.11-2.70	0.10-2.23

n= number of measurements. (THg, n=266; MeHg, n=78). BQL= below quantification limit. Mean, standard deviation (SD), median, 25^{th} and 75^{th} percentiles (p25 and p75), minimum and maximum (range) levels were measured.

Table 3. Summary of studies that assessed temporal Hg trends in fish.

Study	Geographical location	Fish species	n	Period	Temporal trends	Mercury levels
Present study	Fish consumed in the Valencian Community from different origins	Swordfish, fresh tuna, canned tuna, lean fish, other oily fish, and others.	1070 (THg:799, MeHg: 271)	2011-2021	THg and MeHg levels in swordfish tended to decrease from 2011 to 2016 but then increased from 2016 to 2021. Fresh and canned tuna showed a descending trend, in the 1st half of the study period	Swordfish: Median THg 0.76 mg/kg (0.47,1.17); Median MeHg 0.61 (0.30,1.00). Fresh tuna: Median THg 0.46 mg/kg (0.30,0.65); Median MeHg 0.29 (0.17,0.43). Canned tuna: Median THg 0.22 mg/kg (0.12,0.35); Median MeHg 0.16 (0.09,0.28).
Médieu et al. (2024) Médieu et al., 2024)	Western Indian, Northwestern Pacific, Southwestern Pacific, Central North Pacific, Northwestern Atlantic and Eastern equatorial Atlantic	Tuna (3 tuna species)	2910	1971-2022 (variable depending on the region)	Increasing Hg concentrations in skipjack in the late 1990s in the northwestern Pacific. In all the other regions, Hg concentrations remained stable over each study period	Western Indian (2022): Yellowfin: 0.53 ± 0.26; Bigeye: 3.10 ± 1.04; Skipjack: 0.59 ± 0.29. Northwestern Pacific: Skipjack: 0.66 ± 0.13 (2011). Southwestern Pacific: Yellowfin: 0.57 ± 0.34 (2020); Bigeye: 4.68 ± 1.04 (2021); Skipjack: 0.65 ± 0.22 (2021). Central North Pacific: Yellowfin: 0.89 ± 0.38 (2008); Bigeye: 2.14 ± 0.85 (2007). Northwestern Atlantic: Yellowfin: 0.73 ± 0.27 (2011). Eastern central Atlantic (2022): Yellowfin: 0.63 ± 0.09; Bigeye: 0.76 ± 0.09. *All Mean THg ± SD (µg/g)
Bank et al. (2023) (Bank et al., 2023)	The Barents Sea (Northeast Arctic)	Cod (*Gadus morhua*)	1999	1994-2021	Consistently stable Hg concentrations, with a slight decline in the most recent sampling periods	Yearly, least-square means range 0.022–0.037 mg/kg wet weight

Rudershausen et al. (2023) (Rudershausen et al., 2023)	North Carolina. Mid-Atlantic coast of the USA	Blue marlin	148	1975-1977 and 1998-2021	Interdecadal decline of 45% between the 1970s and late 1990s. Concentrations remained stable during the 1st two decades of this century	Mean (SD) THg 9.47 (4.11) µg/g (1975-77) to 4.17 (2.61) µg/g (2020-21)
Kammann et al. (2023) (Kammann et al., 2023)	German Bight (North Sea)	*Limanda limanda* (Common dab)	496	1995-2020	A significant increase, with an annual percental change of 1.4% (41% increase within 25 years)	Mean Hg concentration 107.06 µg/kg at study site 1, and 136.20 µg/kg at study site 2
Ho et al. (2021) (Ho et al., 2021)	North-East Atlantic Ocean	12 commercially important fish species	25631	2006-2019	Concentrations of Hg increased from North to South	Geometric mean 0.0523-0.244 mg/kg in fillet and 0.0114-0.39 mg/kg in liver
Médieu et al. (2021) (Médieu et al., 2021)	South Western Pacific Ocean	Tuna (3 tuna species)	590	2001-2018	Decadal stability. Significant inter-annual variability	Mean (SD) 2.7 (1.7) µg/g in bigeye tuna, 0.7 (0.5) µg/g in yellowfin tuna and 0.7 (0.3) in skipjack
Morris et al. (2022) (Morris et al., 2022)	Across the circumpolar Arctic	Short-horn sculpin, Atlantic cod, and sea-run Arctic char	11 time series (n=3, 7 and 1, respectively)	Recent 20-year trends (1999-2021)	All recent trends were non-significant or significantly increasing: in sculpin from central East and central West Greenland (2.1% and 6.1% per year, respectively) and Atlantic cod from the northwest Faroe Islands and northern Norway (3.9% and 7.4% per year, respectively)	Not reported
Bank et al. (2021) (Bank et al., 2021)	Norwegian sea	Greenland halibut	625	2006-2015	Significant decrease in THg levels (by 35-50%)	Median 0.25 mg/kg in 2006 to 0.14 mg/kg in 2015

Zhang et al. (2022) (Zhang et al., 2022)	Marine and freshwater fish consumed in China	Multiple species	35464	1980-2014	Hg levels decreased over the past three decades (by 70% from 1980 to 2014)	Mean Hg from 0.196 mg/kg during 1980–1985 to 0.054 mg/kg during 2011–2014
Perelló et al. (2014) (Perelló et al., 2014)	Food samples purchased in local markets (Catalonia, Spain)	Multiple species	Not reported	2012 vs 2008	A 44% increase was observed in Hg levels	0.16 and 0.22 µg/g in 2008 and 2012, respectively. 1.5 µg/g in swordfish and 0.68 in fresh tuna (2012)
Bonito et al. (2016) (Bonito et al., 2016)	Global (Atlantic, Pacific, Mediterranean, Baltic, Indian, and Gulf of Mexico)	Multiple species	841	1969-2012	Significant overall decreasing trend	Not reported
Cross et al. (2015) (Cross et al., 2015)	North Carolina. Mid-Atlantic coast of USA	Bluefish	40	1972-2011	43% decline, with an average rate of decline of about 10% per decade	Mean mercury concentration from 0.58 µg/g in 1972 to 0.33 µg/g in 2011
Polak-Juszczak et al. (2012) (Polak-Juszczak, 2012)	Baltic sea	Cod	253	1994-2010	Significant downward trend of 57% in cod muscle and 90% in cod liver	Mean (SD) Hg from 0.132±0.043 to 0.057±0.022 in muscle and from 0.228±0.028 to 0.024±0.006 in liver
Drevnick et al. (2017) (Drevnick et al., 2017)	North Pacific Ocean	Tuna (yellowfin tuna and bigeye tuna)	288 and 172 (yellowfin tuna and bigeye tuna, respectively)	1971-2008	Increasing trends	Yellowfin: Least-square mean 0.227 ± 0.008 ppm in 1971 to 0.338 ± 0.024 ppm in 2008. Bigeye: Least-square mean 0.533 ± 0.016 ppm in 1971 to 0.644 ± 0.029 ppm in 2008

Figure captions

Figure 1. Temporal trends (mean, 2.5% and 97.5% credible intervals) in THg and MeHg levels per fish species estimated using the multi-group GP and multi-level linear models.

Figure 2. Temporal trends (mean, 2.5% and 97.5% credible intervals) in THg and MeHg levels of swordfish species per area of origin estimated using the multi-group GP and multi-level linear models.

Figure B.1. Uniform Q-Q plot of leave-one-out probability integral transformation (LOO-PIT) values for the multi-group GP model (left) and multi-level linear model (right).

Figure B.2. Root mean square error (RMSE) for the multi-group GP and multi-level linear models per fish species and Hg type.

Figure B.3. Expected log predictive density (ELPD) for the multi-group GP and multi-level linear models per fish species and Hg type.

Figure C.1. **Estimated posterior distributions (mean, 2.5%, 25%, 75% and 97.5% credible intervals) of the missing THg samples along with their corresponding actual MeHg observations represented by purple dots.**

Figure C.2. **Estimated posterior distributions (mean, 2.5%, 25%, 75% and 97.5% credible intervals) of the BQL observations.** The horizontal dashed grey lines represent the different LOQs (0.1, 0.05, 0.02, 0.01) of the samples. In the “Others” category a selection of 36 out of 179 THg and 4 out of 26 MeHg BQL measurements is shown.

Figure C.3. **Estimated posterior distributions (mean, 2.5%, 25%, 75% and 97.5% credible intervals) of the BQL observations (in blue) and the LOQs (in purple).** The horizontal dashed grey lines represent the different LOQs (0.1, 0.05, 0.02, 0.01) in the dataset.

Supplementary material

Table S1. Below quantification limit (BQL) Hg measurements per year.

	TOTAL	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021
n	1070	77	90	121	169	113	163	70	65	57	68	77
BQL	285(26.6%)	21(27%)	15(17%)	20(17%)	37(22%)	51(45%)	53(33%)	21(30%)	15(23%)	12(21%)	18(26%)	22(29%)

n= number of Hg measurements. BQL= below quantification limit.

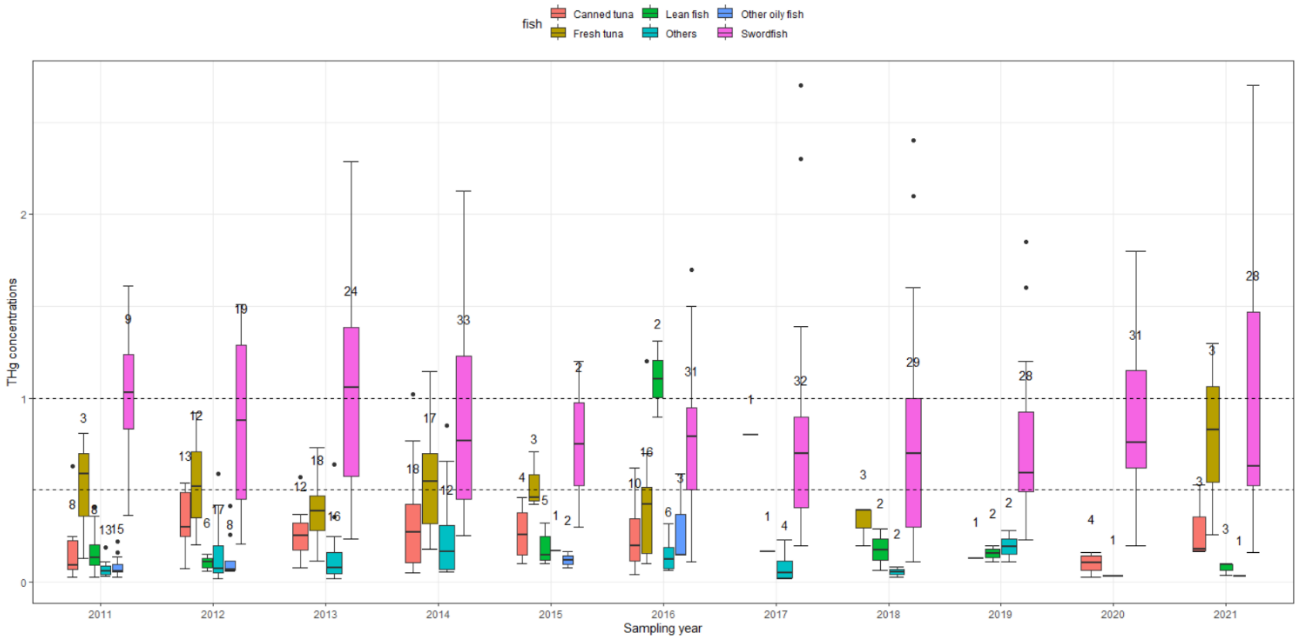

Figure S1. Distribution of THg concentrations by fish groups throughout the study period (2011-2021) (The Valencian Community, Spain). The dotted lines show the maximum limit settled for most species (1 or 0.5 mg/kg). THg= Total mercury. n=547.

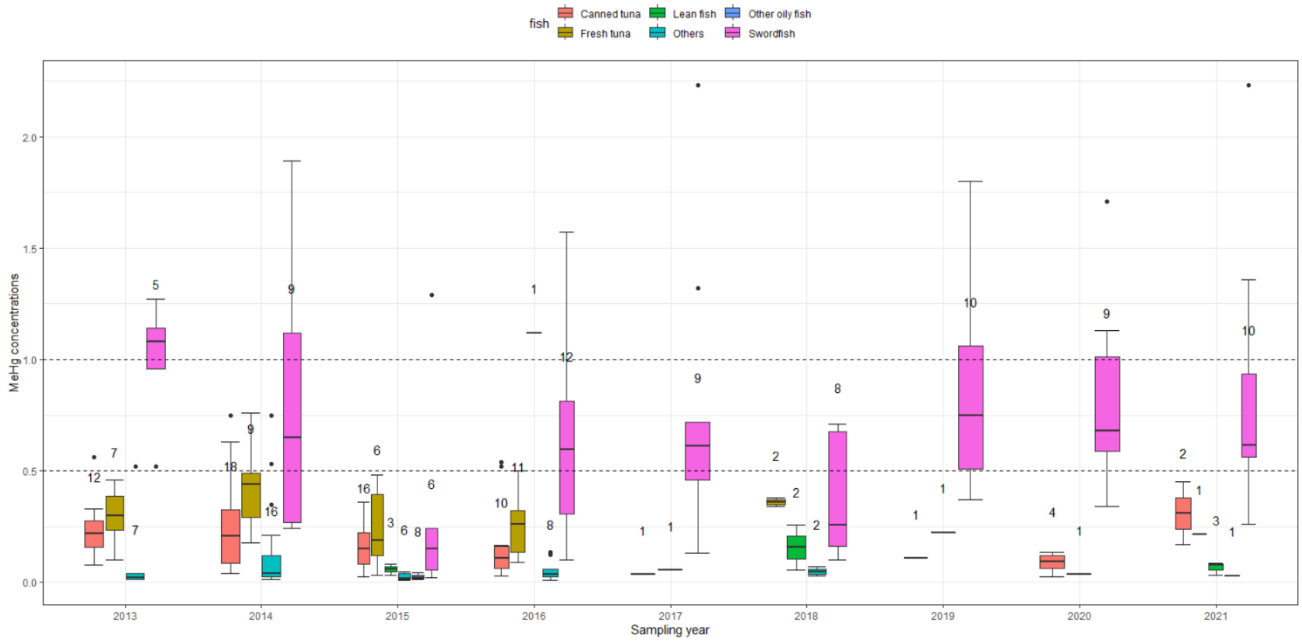

Figure S2. Distribution of MeHg concentrations by fish groups throughout the period (2013-2021) (The Valencian Community, Spain). The dotted lines show the maximum limit settled for most species (1 or 0.5 mg/kg). MeHg= methylmercury. n=238.

Figure 1

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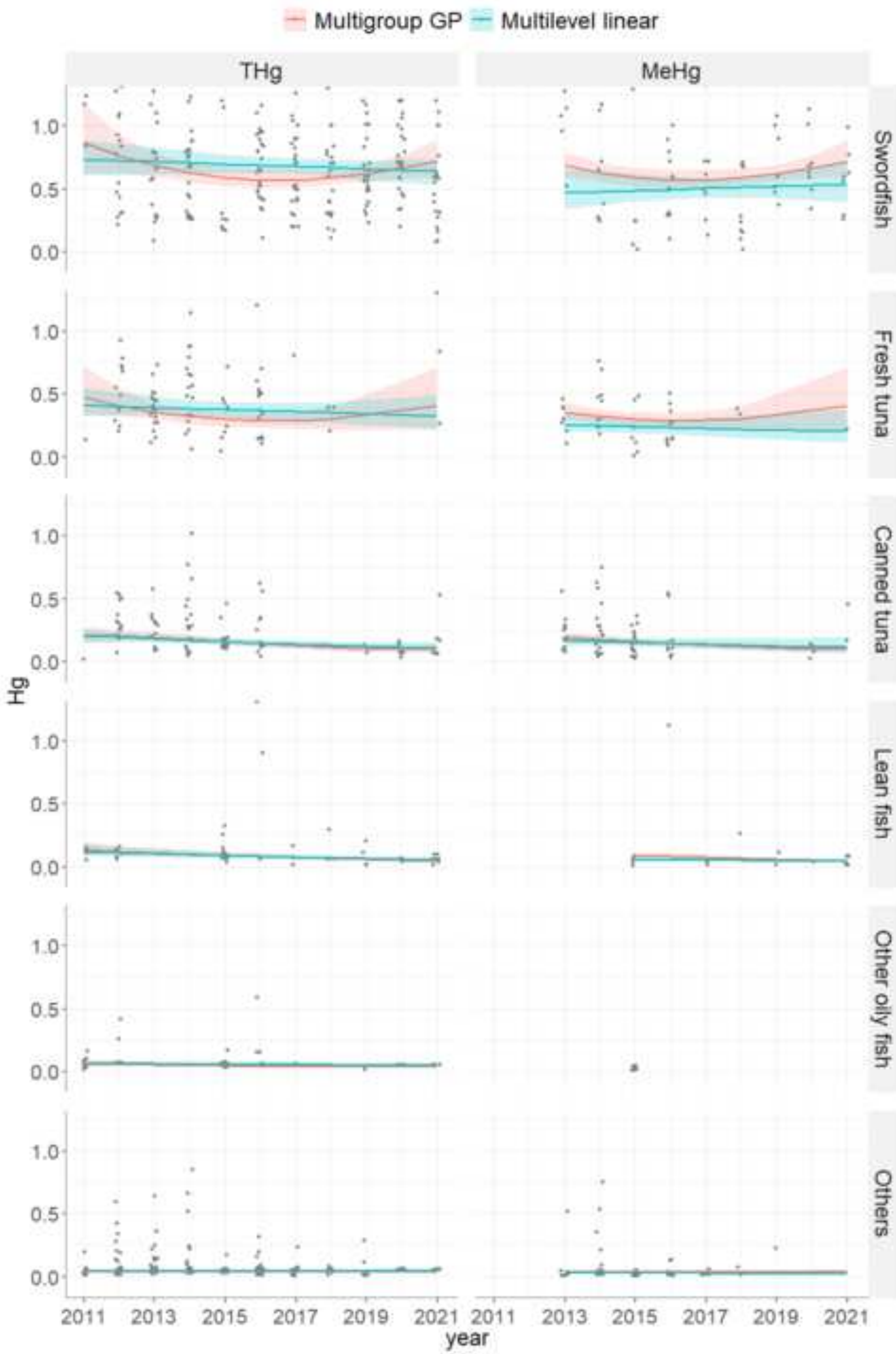

Figure 2

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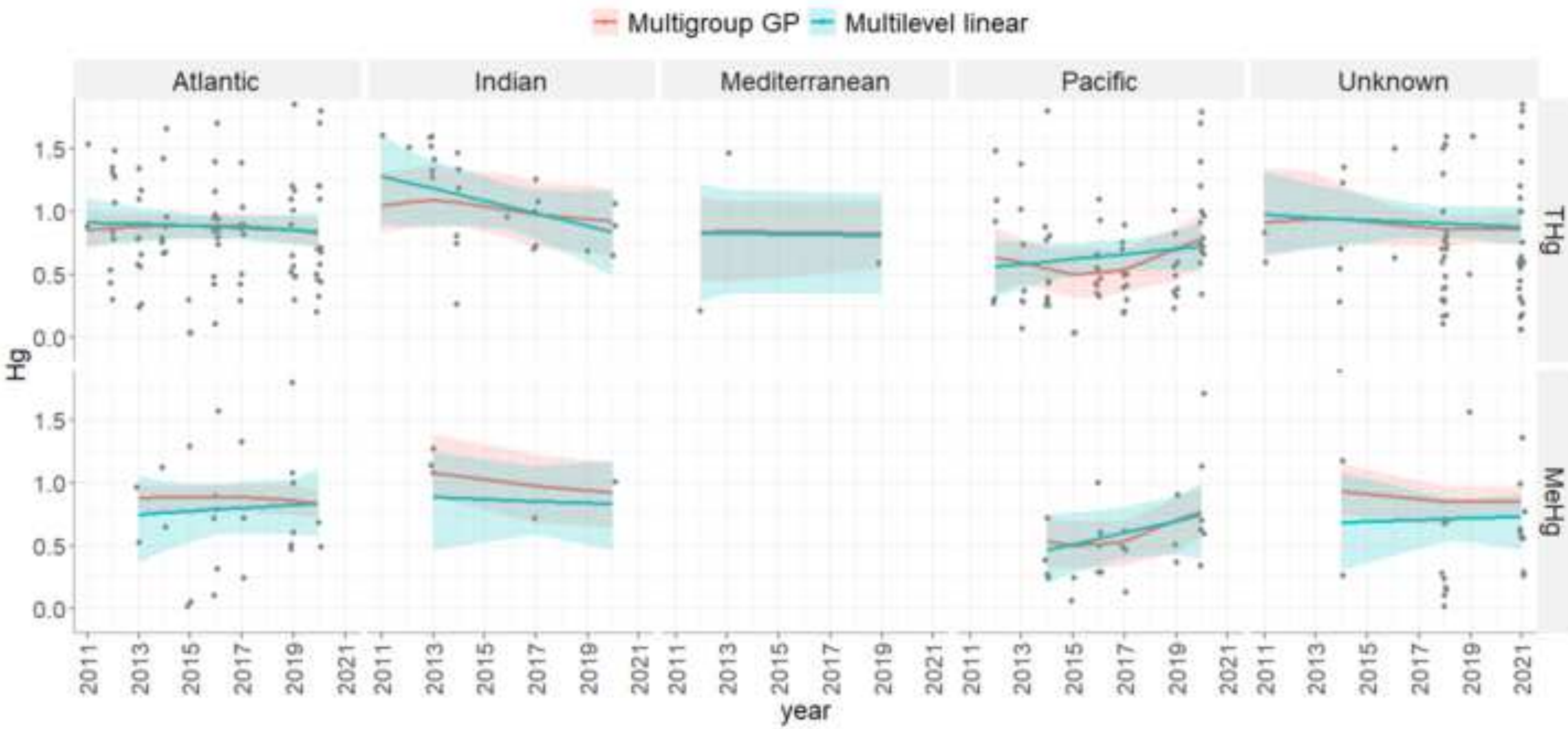

Figure B1

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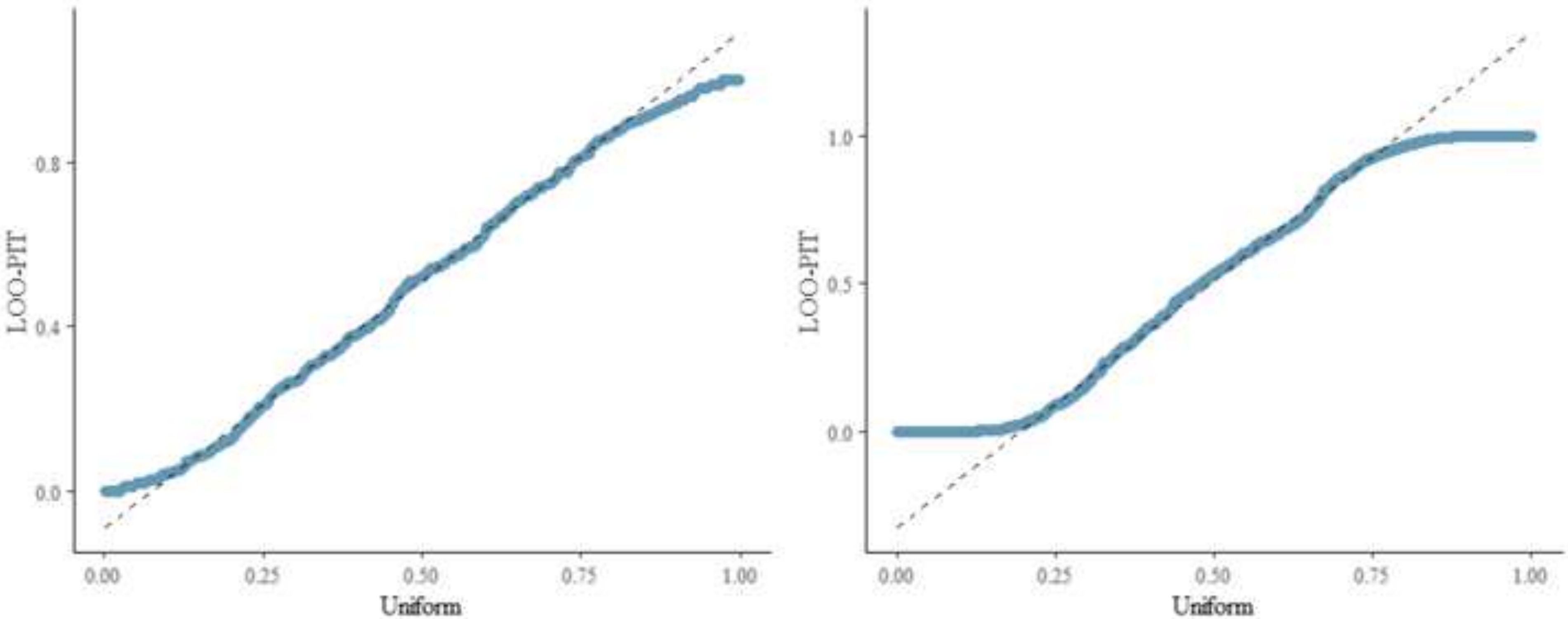

Figure B2

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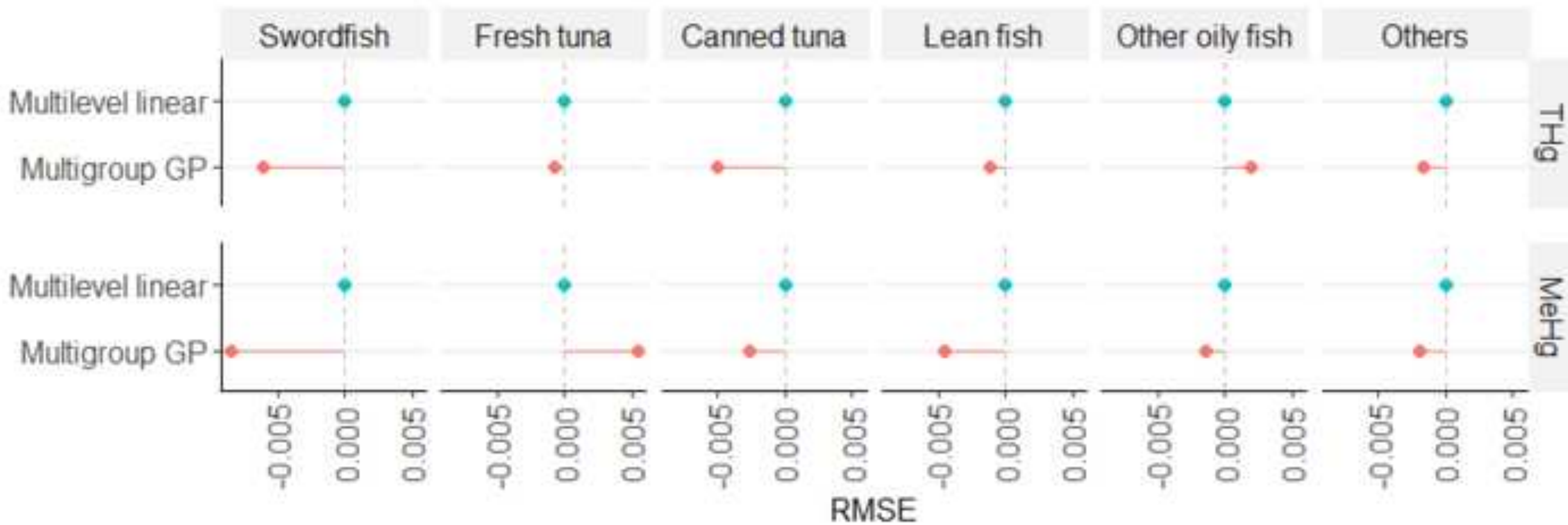

Figure B3

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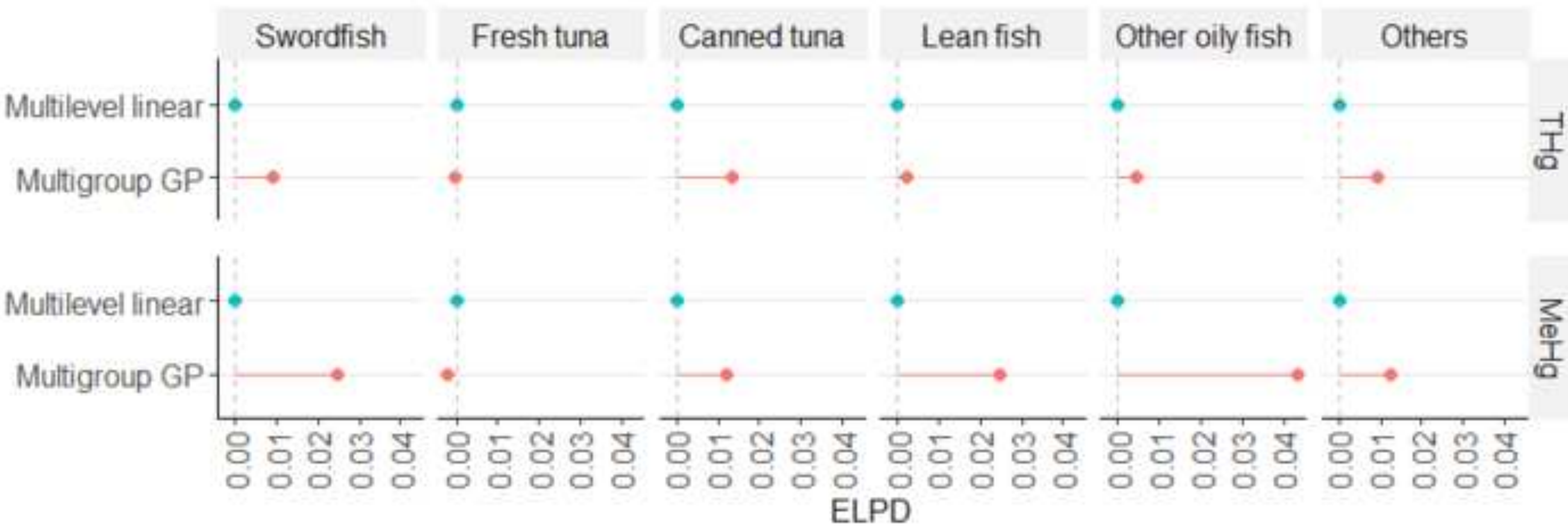

Figure C1

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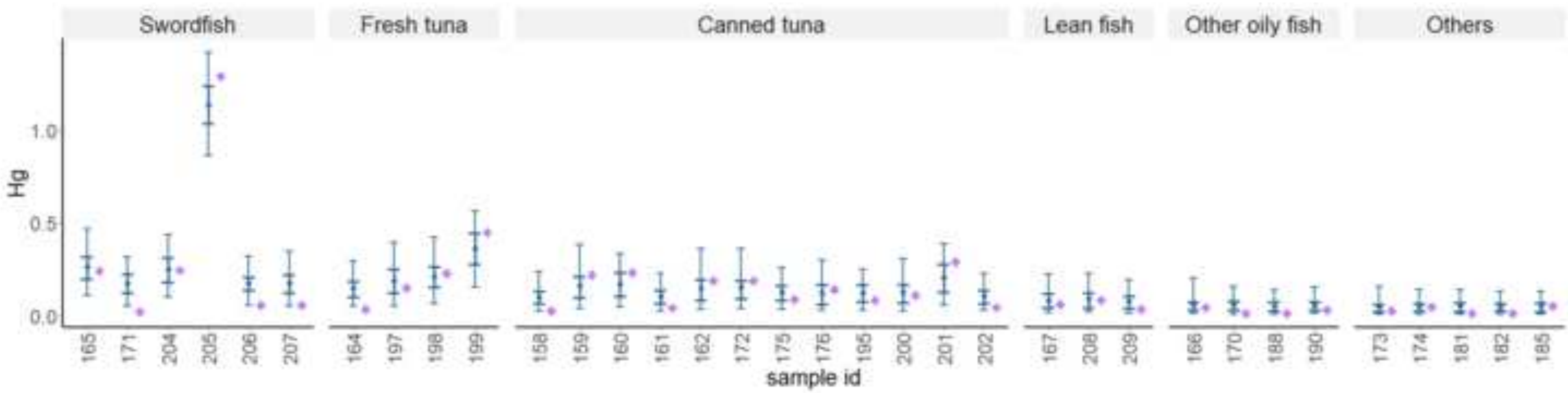


Figure C2 Click here to access/download;Figure(s);figC2.png

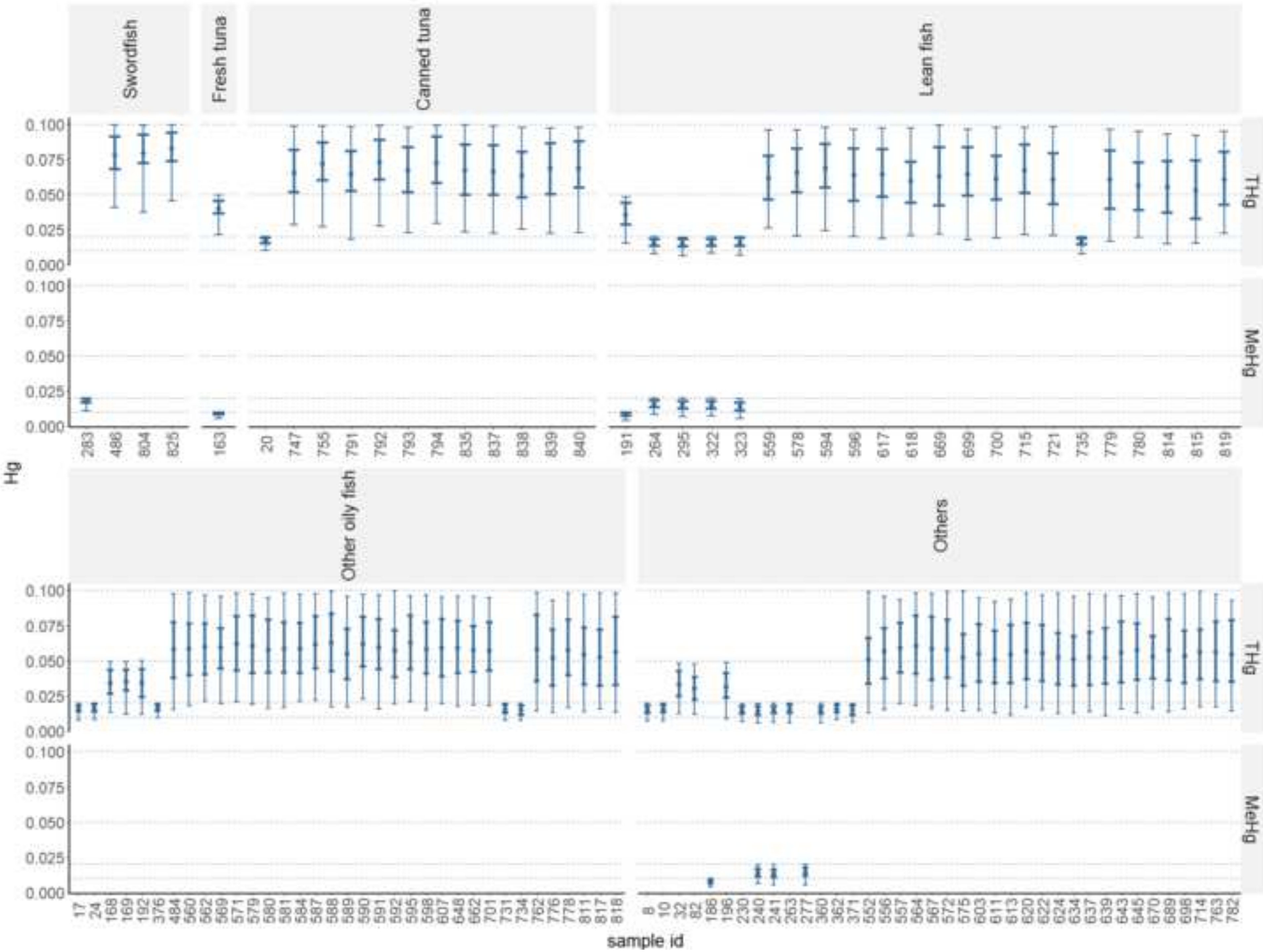
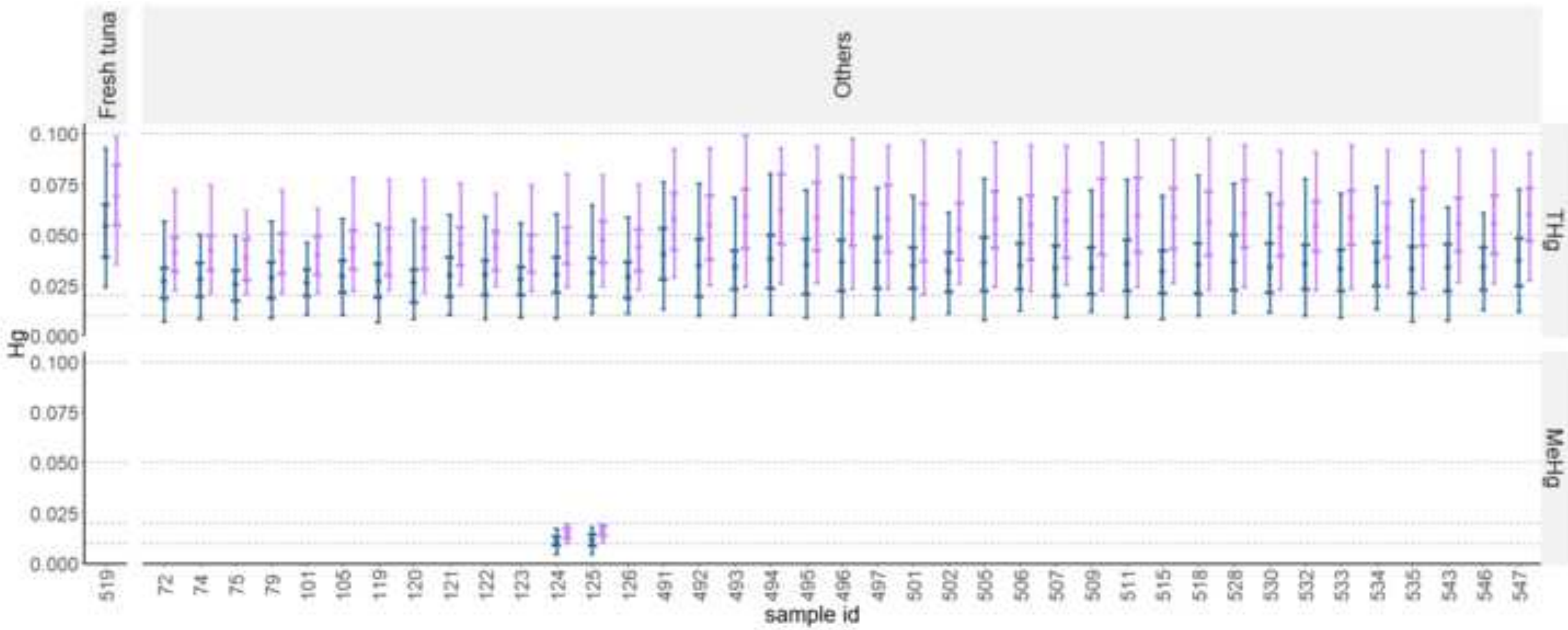


Figure C3

Click here to access/download;Figure(s);figC3.png



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Supplementary Data
MPB_Supplemental.docx

Declaration of interests

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: