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Widespread presence of metallic compounds and organic contaminants across Pacific coral reef fish

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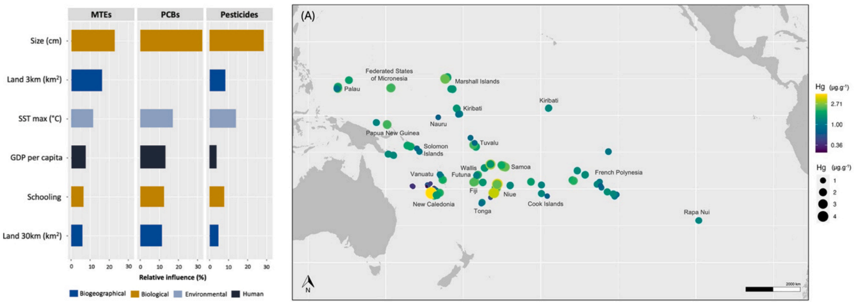
HIGHLIGHTS

- We studied the distribution of MTEs and POPs in Pacific coral reef fish.
- All the components analyzed were measured in low to modest concentrations overall, but with a wide spatial distribution.
- The biogeographic, biological, human and environmental factors driving these spatial patterns have been identified.
- The specific cases of mercury and glyphosate are discussed.
- The distribution maps for the various contaminants provide essential information for fisheries managers and consumers.

GRAPHICAL ABSTRACT



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ABSTRACT

Coral reef fishes represent an invaluable source of macro- and micro-nutrients for tropical coastal populations. However, several potentially toxic compounds may jeopardize their contribution to food security. Concentrations of metallic compounds and trace elements (MTEs), and persistent organic pollutants (POPs, including pesticides and polychlorobiphenyls PCBs), totalizing 36 contaminants, were measured in coral reef fish from several Pacific islands. The objective of this study was to describe the spatial distribution of these compounds and contaminants in order to identify potential variables explaining their distribution at a Pacific-wide scale. To achieve this, we applied Boosted Regression Trees to model species-specific and community-level contaminant and inorganic compound concentrations at the scale of the tropical Pacific Ocean. Overall, using 15 easily accessible explanatory variables, we successfully explained between 60 and 87 % of the global variation, with fish body size being the most important correlate of MTEs and POPs concentrations in reef fish. Our modeling approach allowed us to estimate and map the distribution of the community-level concentration of 19 contaminants and inorganic

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compounds at the scale of the equatorial and south Pacific Ocean. Spatial patterns varied significantly depending on the compound, with modeled quantities per 100 g of fish flesh generally being higher in the central and southwest Pacific than in the eastern part of the basin. These patterns were influenced by a combination of biological, environmental, anthropogenic and biogeographical variables. Overall, this approach represents an important step toward the estimation of concentrations of the main compounds on the basis of species identity and fishing location. Our results enhance our understanding of the extent of contamination in the Pacific while underscoring the urgent need for long-term and large-scale spatial monitoring of diverse compounds in this region.

1. Introduction

Most of the islands and atolls of the Pacific Ocean are geographically isolated from continental margins, with limited industrial and agricultural production and relatively small human populations (Fey et al., 2019; Nalley et al., 2023). In these territories, human communities have traditionally relied on harvesting marine resources, with a particular emphasis on the consumption of coastal and pelagic fish (Pratchett et al., 2011; Johnson et al., 2017; Bell et al., 2018). Per capita fish consumption in the Pacific is among the highest in the world, ranging from 2 to 4 times higher than the global average (Bell et al., 2009; Gillett and Fong, 2016), and ranges from 55 kg to 110 kg per person per year (Bell et al., 2009). Coral reef and other coastal fish are often favored in subsistence fishing practices over pelagic species due to the accessibility of their stocks. Moreover, coral reef fish represent a major source of proteins, essential vitamins e.g., retinol (A), nicotinamide (B3), pyridoxine (B6), cobalamin (B12), minerals e.g., calcium, iron, selenium, and zinc, as well as essential ω3 and ω6 polyunsaturated fatty acids (Wang et al., 2006; Hicks et al., 2019; Byrd et al., 2021). However, despite their high nutritional value, coral reef fishes may also represent a major vector of several chemical contaminants, which raises concerns due to their high consumption in the Pacific (Storelli, 2008; Sabino et al., 2022).

Coral reefs are currently facing a range of disturbances, both of natural and anthropogenic origin (Graham et al., 2008, 2013; Hoey et al., 2016; Mumby et al., 2016), with chemical pollution playing a significant role (Richmond, 1993). The continuous expansion of anthropogenic activities on Earth, such as urbanization, agriculture, industrialization, and mining has led to habitat degradation and environmental pollution through effluents and emissions (Ashraf et al., 2012; Kumar-Roiné et al., 2022). Three major classes of contaminants are of particular concern: polychlorobiphenyls (PCBs), pesticides (both of which are classified as persistent organic pollutants or POPs), and metallic trace elements or MTEs (Briand et al., 2014; Fey et al., 2019). These ubiquitous contaminants can be toxic even at low concentrations, they are resistant to degradation and may be dispersed over long distances (Phillips, 1995). Furthermore, they have the potential to bioaccumulate during individual life and/or to bioamplify along trophic networks, posing potential health risks to consumers (e.g., De Gieter and Baeyens, 2005; Peter and Viraraghavan, 2005; Baeyens et al., 2005). Most previous research on contaminants in coral reefs has focused on a specific type of contaminant or a limited number of species (Chouvelon et al., 2009; Metian et al., 2009, 2013; Hédouin et al., 2010, 2011; Bonnet et al., 2014; Dromard et al., 2016; Ritger et al., 2018 but see Sabino et al., 2022), thereby limiting our capacity to understand the complex multidimensional nature of fish contamination in the Pacific Ocean.

Here, we assess the potential contamination of coral reef fish assemblages across 9 islands in the tropical Pacific Ocean, with the main aim of identifying the main variables driving the spatial distribution of contaminant, including biological, environmental, biogeographic, and anthropogenic aspects. To achieve this goal, our approach is based on two main steps: (i) we used Boosted Regression Trees (BRT) to model the contamination of reef fishes according to their traits on the basis of a comprehensive dataset of contaminants concentration for 980 individual fish. Then, (ii) we integrated this model with fish community surveys across the Pacific (Ruppert et al., 2018), in order to predict and map contaminant concentrations at a larger spatial scale. Overall, we provide information on fourteen MTEs, twenty-one pesticides, and one PCB contamination index (the sum of all measured congeners), at the scale of the equatorial and south Pacific.

2. Material and methods

2.1. Study sites and fish sampling

Fish were sampled at 27 sites across 9 Pacific Island countries and territories (hereafter PICTs) from 2011 to 2021 (Fig. 1). A total of 983 individuals belonging to 146 species were sampled during this period (Suppl. Table S1). Analyses were conducted to obtain a wide range of fish species with contrasting feeding strategies. Fish species were categorized into four broad trophic groups: herbivores (mainly consuming macrophytes), omnivores (consuming both animal and plant material), micro-carnivores (consuming small prey), and macro-carnivores (consuming larger prey), based on known dietary patterns (Parravicini et al., 2020). Fish were collected either directly in the field through spearfishing, handlining or using a small quantity of non-selective anesthetic, namely 10 % diluted eugenol in ethanol, or by purchasing specimens from local fishermen. In all cases, fish were stored in coolers, and each individual was identified and total length (in cm) measured. For each fish, dorsal muscle samples were taken and frozen at −20 °C until analysis. For each sample, subsamples of approximately 1 to 5 g (wet weight) were prepared for nitrogen stable isotope analysis (Suppl. Text S1), and of approximately 5 to 10 g (wet weight) for MTEs measurements and POPs analysis, i.e., pesticides and PCBs.

2.2. Contaminant analyses

2.2.1. Analysis of metallic compounds and trace elements (MTEs)

A total of 14 MTEs were analyzed: silver (Ag), arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), mercury (Hg), nickel (Ni), lead (Pb), selenium (Se), vanadium (V), and zinc (Zn). Except for Hg, analyses of these elements were performed on dry (lyophilized and powdered) samples (150–300 mg) using either inductively coupled plasma atomic emission spectroscopy (ICP-AES 5800 VDV, Agilent Technologies ®) or inductively coupled plasma-mass spectrometry (ICP-MS II Series Thermo Fisher Scientific®), as described in Kojadinovic et al. (2011). Total Hg concentrations were quantified in samples ranging from 3 to 100 mg of dry (lyophilized and powdered) samples by atomic absorption spectrometry with an advanced mercury analyzer (AMA 254 ALTEC®), following the procedure described by Bustamante et al. (2006).

A quality control program was implemented, including the treatment and analysis of certified reference materials (CRMs, which included DOLT-5 dogfish liver and TORT-3 lobster hepatopancreas from the National Research Council, Canada) and blanks alongside the samples. These CRMs were processed and analyzed simultaneously with the samples. The recovery rates for the CRMs ranged from 83 % to 115 %. All MTEs concentrations are reported in $\mu g.g^{-1}$ of fish dry mass (dm), and were conducted on 980 fish samples. The detection limits (in $\mu g.g^{-1}$ dm) were 0.005 $\mu g.g^{-1}$ (Hg), 0.015 $\mu g.g^{-1}$ (Ag, Cd), 0.02 $\mu g.g^{-1}$ (Cr, Co, Pb), 0.03 μg.g^{-1} (Ni), 0.08 μg.g^{-1} (Mn), 0.1 μg.g^{-1} (Cu, Se), 0.2 μg.g^{-1} (As), 0.3 μg.g^{-1} (V), and 3.3 μg.g^{-1} (Fe, Zn). When concentrations were below the detection limit, half of the detection limit was used.

2.2.2. Analysis of persistent organic pollutants (POPs)

PCBs comprise a large family of synthetic compounds (209 congeners). A total of 41 PCB congeners were analyzed on 710 fish samples, and identified by their IUPAC (International Union of Pure and Applied Chemistry) numbers: 17, 18, 20, 28, 31, 33, 44, 49, 52, 60, 70, 74, 82, 87, 92, 95, 99, 101, 105, 110, 118, 128, 132, 136, 138, 141, 149, 151, 153, 156, 170, 174, 177, 180, 183, 187, 191, 194, 195, 196, and 201. The overall PCB contamination i.e., the sum of all analyzed congeners (Σ PCB) was calculated, and expressed in ng.g^{-1} dm. Quantification was performed using gas chromatography (Agilent Technologies HP6890®) equipped with an electron capture detector at 300 °C and an automatic injector in the column (DB5 J&W column, 60 × 0.32 i.d × 0.25 μm), with helium as the carrier gas. During injection, the temperature is 60 °C, then it increases by 10 °C per minute to 160 °C, and then by 25 °C per minute to 280 °C. The detection limit was 0.01 ng.g^{-1} dm. When concentrations were below the detection limit, half of the detection limit was used.

Pesticides, a group of chemical compounds widely used to hinder the development of various pests such as insects, weeds, and others (Abhilash and Singh, 2009; Kim et al., 2017), were analyzed in this study on 716 fish samples, and expressed in ng.g^{-1} dm. Twenty-one pesticides were examined, including aldrin, atrazine, α- and cis-chlordane, diazinon, dieldrin, α- and β-endosulfan, endrin, glyphosate, heptachlor, heptachlor epoxide A and B, isodrin, lindane, linuron, malathion, simazine, pp'-DDT, pp'-DDE, and pp'-DDD, chosen due to their persistence and public health interest. Glyphosate was quantified using liquid chromatography, and the other compounds were quantified using gas chromatography–mass spectrometry following the methods fully described in Dierking et al. (2009). The detection limits (DL, in ng.g^{-1} dm) were 0.01 ng.g^{-1} for heptachlor epoxide A and B, 0.02 ng.g^{-1} for linuron and malathion. For aldrin, diazinon, endosulfan II, endrin, heptachlor, lindane, pp'-DDT, pp'-DDE, and pp'-DDD, the DL is 0.1 ng.g^{-1}, 0.2 ng.g^{-1} for atrazine, dieldrin, and endosulfan I. Finally, the DL for glyphosate is 1 ng.g^{-1}. When concentrations were below the detection limit, half of the detection limit was used.

2.3. Selection of potential predictive variables

To examine the relationships between site-specific predictive variables potentially influencing MTEs and POPs concentrations in coral reef fish, we have selected four categories of variables related to environment, biogeography, human activities, and fish biological characteristics. Within each of these four categories, several variables were considered, totalizing 25 potential variables influencing contaminants' concentrations (see Suppl. Text 2 for details and justification of variables' choice).

Species-specific biological variables are known to influence contaminant bioaccumulation in marine organisms (Žižek et al., 2007). Here, we accounted for 7 biological variables: δ^{15}N, individual size (total length, in cm), dietary habits, mobility, gregariousness, position in the water column, and the activity period.

Water temperature is a key-environmental variable because it significantly influences the distribution of environmental contaminants, such as POPs, in sediment, water, and the atmosphere (Noyes et al., 2009), and impacts the distribution, movement and activity of marine species (Lea et al., 2015). Sea Surface Temperature (SST) was obtained from data provided by the Group for High Resolution Sea Surface Temperature (GHRSST) project (http://podaac.jpl.nasa.gov/). It represents the monthly temperatures over a ten-year period from 2004 to 2014, with a spatial resolution of 0.011° latitude and 0.011° longitude. Three variables were considered for modeling: minimal, averaged and maximal monthly SST.

Seven biogeographical variables were included in the models to account for the spatial extent of the study. Sandin et al. (2008) demonstrated the positive effect of island surface area, which can provide insights into the various potential interactions between terrestrial and marine ecosystems, including terrestrial runoffs. Additionally, POPs are often semi-volatile, allowing them to be transported over long distances and leading to contamination of sites far from their initial use or production location (Polder et al., 2014). These biogeographical variables

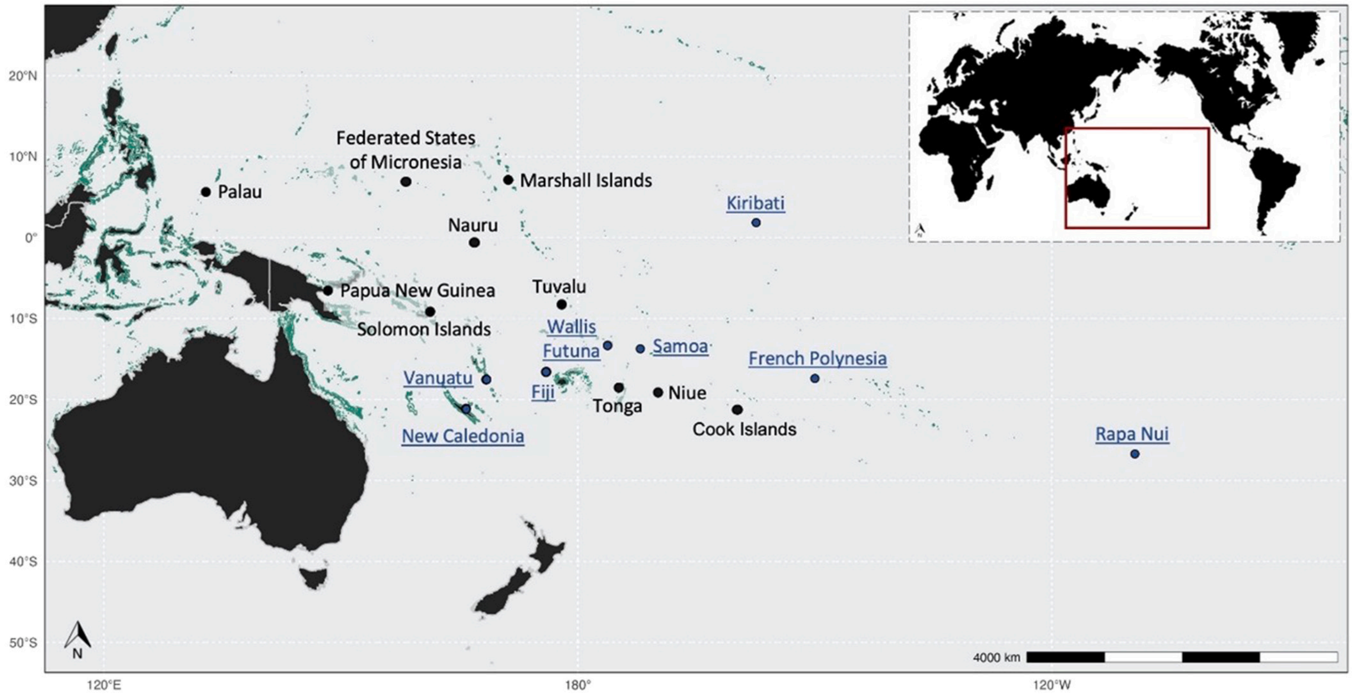

Fig. 1. Map showing the 17 PICTs where coral reef fish assemblages were assessed by Ruppert et al. (2018) (in black and underlined in blue, except Rapa Nui), and the 9 PICTs where fish individuals were sampled for contamination analysis (in blue and underlined).

used in the models were total land area of the country/island, land area within 3, 30, and 600 km buffer zones, and coral reef area within a radius of 3, 30, and 600 km.

Finally, eight human variables were considered, because human activities in coastal areas adjacent to coral reefs may also affect coastal ecosystems through pollution, urbanization, agriculture or fishing (Mora et al., 2011): the surface of the nearby city or town at the sampling site, the coastal population size, the total human population size for each country/island, the Gross Domestic Product (GDP) per capita, the Human Development Index (HDI), the human gravity, the land area used for agriculture, and the pesticide trade, i.e. the quantity of pesticides imported annually.

Among the 25 variables initially considered and presented above (see also Suppl Text S2), we conducted correlation tests (Pearson's r) between the predictive variables to identify and remove those that were highly correlated. In accordance with a recommended practice (Dormann et al., 2013), we eliminated collinearity among the explanatory variables by removing those with a Pearson correlation coefficient exceeding 0.7. As a result of these tests, data related to δ^{15}N, position in the water column, minimum and averaged SST, the size of the nearby city or town at the sampling site, coastal population sizes, 600 km coral reef radius, 600 km land buffer zones, the HDI, and population density were removed from our potential driving variables. Therefore, for the first part of our analysis (see Section 2.4 below), we utilized 15 predictive variables, including individual size, diet, mobility, activity, gregariousness, coral reef area within a 3 km and 30 km buffer zone, land area within a 3 km and 30 km buffer zone, maximum SST, total population, GDP per capita, human gravity, agricultural land area, and pesticide trade (Suppl. Table S2).

2.4. Drivers of fish contamination

To identify potential predictive variables influencing the contamination of sampled coral reef fish, we employed Boosted Regression Trees (BRT). BRT can handle non-linear relationships and interactions between variables, and generate a series of regression trees, with each new tree explaining the residuals from the previous one (Elith et al., 2008). Four main parameters need to be specified: the bag fraction (bf), the learning rate (lr), tree complexity (tc), and the number of trees (nt). According to the recommendations of Elith et al. (2008), to determine the best model parameters, we tested different combinations of lr and tc to generate multiple BRT models. Consequently, a combination of lr (0.01, 0.005, 0.001) and tc (1 to 5) was used to run BRT models. BRTs with the best cross-validation (10-fold) correlation were kept, and then fitted again keeping only variables with >5 % importance in the model. The BRTs were fitted using the *gbm* package in R (Ridgeway, 2024) in addition to functions provided by Elith et al. (2008). Before starting the analyses, it was suggested that eliminating collinearity among explanatory variables with a Pearson correlation coefficient exceeding 0.7 is a recommended practice (Dormann et al., 2013). Once each model has been run and selected relative to its predictive deviance, a modified simplification procedure according to Elith et al. (2008) was applied so irrelevant factors were removed objectively from the model.

2.5. Modeling contamination in coral reef fish communities

After completing the first phase of our analysis, during which we modeled the contamination of coral reef fish based on their measured concentrations of contaminants while evaluating the potential role of various variables, we then proceeded to the second phase of our analysis, which consists of two fundamental steps. In the first step, we used an existing dataset on Pacific fish densities and biomasses (Ruppert et al., 2018), combined with similar data from the Marquesas Islands (SO CORAIL, 2023) and Rapa Nui (i.e., Easter Island) (Hinojosa et al., 2021). In summary, this dataset included a total of 86 sites in 17 PICTs, ranging from Palau in the west to Rapa Nui in the east, and from the Marshall Islands in the north to Tonga in the south (Fig. 1). To do that, we used the *predict.gbm* function from the *gbm* package in R. This function is specifically designed to make predictions based on the regression model established in the first phase (i.e., in 9 PICTs), while ensuring that exactly the same explanatory variables are used as in the initial model. By applying the *predict.gbm* function, we were able to estimate fish contamination levels for the densities and biomass data, enabling us to extend our 1st phase' analysis to a wider spatial scale. These data were then associated with the contaminant concentrations measured during the first phase; only contaminants with a cross-validation (CV) correlation $\geq$0.6 were retained to ensure the reliability and robustness of our model, resulting in a total of 22 contaminants being selected. Thus, 14 contaminants were excluded: Co, Pb, aldrin, α-chlordane, cis-chlordane, dieldrin, endrin, heptachlor, heptachlor epoxide A, isodrin, linuron, malathion, pp'-DDD and pp'-DDE.

It should be noted that the BRT models developed in this second phase initially considered the 18 environmental, biogeographic and human variables used in the first part, but not biological ones as the collected data consisted of fish community censuses. In line with the recommendations of Dormann et al. (2013), we removed variables showing collinearity i.e., a Pearson correlation coefficient > 0.7.

Following these tests, 9 variables were used for the analyses, including mean SST, human gravity, reef area within 3 km, 30 km, and 600 km buffer zones, land area within 3 km and 30 km buffer zones, total population, and pesticide trade (Suppl. Table S3). The 9 variables that were removed (due to collinearity, which could have introduced bias in the model) were minimum and maximum SST, land area in 600 km buffer zones, size of town or village near sampling site, size of coastal population, total population density, GDP per capita, human development index and land area used for agriculture. The second step of our process focused on normalizing contamination for each element, based on a quantity of 100 g of fish. This facilitated mapping at the Pacific scale, providing an overview of estimated contamination levels in the region. By combining these two phases, we established a framework for assessing coral reef fish contamination at the regional scale, while also providing information on the predictive variables used in the BRT models.

3. Results

3.1. Contaminants' concentrations in sampled fish

Overall, averaged MTE concentrations, all trophic categories pooled, ranged from very low (e.g. Ag, Cd; elements often being under the DL) to higher (e.g. As, Fe, Zn) values; the other elements being at intermediate levels (Suppl. Table S4a).

Regarding POPs concentrations, glyphosate was globally and by far the most abundant pesticide reaching ~85 % of all pesticides, with an average concentration slightly lower than 90 ng.g^{-1}, all trophic categories pooled (Suppl Table S4b). Other pesticides showed very low (e.g., pp-DDT, isodrine; elements often being under the DL) to intermediate values (pp-DDD, β-endosulfan). The sum of PCB, all values pooled, reached ~30 ng.g^{-1} (Suppl. Table S4b).

3.2. Drivers of fish contamination

Using 15 predictive variables, BRT modeling explained 60 % to 87 % of the global variation (cross-validation, CV) in concentrations of 36 compounds and contaminants (14 MTEs, 21 pesticides, and the sum of PCBs) in coral reef fish (Suppl. Table S5). Averaging the contributions of the predictive variables for each contaminant category (i.e., MTEs, pesticides and PCBs) revealed that individual size was consistently and by far the most important variable determining the concentration of all contaminant categories, with an importance for MTEs, PCBs and pesticides of 22.4 %, 31.5 %, and 27.5 % respectively. Land area in a 3 km buffer zone was a secondary variable for MTEs, with a contribution of

14.8 %. Maximum SST was the second most influential variable for PCBs and pesticides, with relative importance of 11.6 % and 12.6 % respectively, and the third for MTEs with an importance of 10.5 %. Several other predictive variables played a lower or even a marginal role (i.e., < 10 %) as drivers of fish contamination, depending on contaminant categories (Fig. 2 and Suppl. Fig. S1).

Although individual size was the main driver for most MTE, significant exceptions highlighted the diversity of variables influencing fish contamination (Table 1). For example, Pb followed the general trend (i.e., the trend found for all MTE pooled), showing a significant influence of individual size (62.1 %). In contrast, Fe exhibited a strong relationship with human and biogeographical variables such as agricultural land area (31.2 %) and land within a 3 km radius (30.4 %), relegating individual size to the fourth position (13.8 %). Similarly, Ni relied more on environmental variables such as SST (31.0 %), thereby reducing the influence of individual size to the second place (21.5 %). This diversity of interactions was also present in the in-depth study of pesticides, where the individual size of fish emerged as a major determinant of concentrations. Notably, for example, aldrin (29.7 %), cis-chlordane (26.6 %), diazinon (26.3 %), dieldrin (28.1 %), and many others followed the general trend. Conversely, certain pesticides, such as Heptachlor epoxide B, heavily depended on human and biogeographical variables (total population (25.5 %), land area 3 km (18.8 %), agricultural land area (16.5 %), and GDP per capita (14.0 %)), diminishing the importance of individual size as a determining variable. Similarly, for atrazine, which relied more on environmental variables as SST (35.2 %) and biogeographical variables (land 3 km (22.2 %)), reduced the role of individual size to a lower contribution (19.3 %). These additional examples highlighted the variety of responses observed among various MTEs and pesticides, underscoring the intrinsic complexity of interactions between these contaminants and fish. While individual size remained a key variable in most cases, these results emphasized the importance of a nuanced approach to understanding fish contamination by these pollutants.

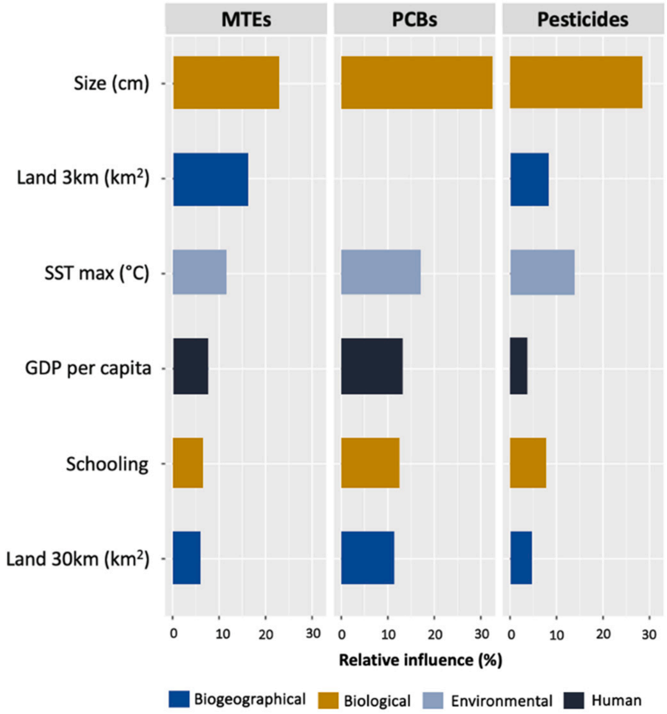

Fig. 2. Summary of the average relative contributions (%) of the six most significant variables (i.e., > 10 %) used in BRTs explaining their relative influence on coral reef fish contamination (See Suppl. Fig. S1 for more details of all variables).

For illustrative purposes, we specifically highlighted the cases of Hg and glyphosate because they are considered priority contaminants in terms of public health due to their high established (Hg) or suspected (glyphosate) toxicity levels. The Hg concentration in coral reef fish was predominantly influenced by biological variables, such as individual size (34.9 %) and diet (28.7 %) (Fig. 3A, B). The maximum SST also drove Hg concentration, although to a lower extend (11.8 %) and ranking third. The concentration of Hg in fish clearly increased with increasing fish size but also with increasing SST, whereas carnivore diets implied higher concentrations (Fig. 3B). Glyphosate concentration in reef fish was primarily determined by individual size (42.4 %) (Fig. 3C, D). Other variables driving glyphosate concentration are only half as important as size, i.e., the total population (17.5 %) and coral reef area within a 30 km buffer zone (13.2 %) (Fig. 3C, D). The concentration of glyphosate in fish clearly increased with the increase in fish size and the total population, whereas the driving role of reef area mostly appeared for 30–50 km^2 (Fig. 3D).

3.3. *Modeling Pacific coral reef fish communities' contamination*

By integrating fish community data with contamination data, we then determined the quantities of contaminants per 100 g of fish at 86 sites across 17 PICTs. The BRT modeling revealed that contamination at the fish community level was mainly explained by biogeographical variables, followed by human and environmental variables (Fig. 4). The land area within a 30 km radius was the primary predictive variable determining the concentration of MTEs in coral reef fish communities (mean contributions of 22.7 %), followed by the human gravity (18.6 %), and the coral reef area within a 30 km radius (12.4 %). For pesticides, the most important variables were also of biogeographical nature, i.e., the coral reef area within a 30 km radius (16.3 %), the land area within a 30 km radius (15.4 %), and the human gravity (13.4 %). In contrast, for PCB concentration, the most influential variable was an environmental driver, i.e., the average SST (18.3 %). The coral reef area within a 30 km radius was the secondary variable (15.0 %), and the third variable was the land area within a 30 km radius (12.7 %) (Fig. 4).

3.4. *Mapping contamination distribution in the Pacific*

Modeled concentrations of contaminants, whether for MTEs or POPs, were generally higher in the central and southwest Pacific, and lower in the eastern part of the basin, encompassing regions such as French Polynesia, the Cook Islands, and Rapa Nui (Fig. 5, Suppl. Fig. S3, S4). For example, the highest Hg concentrations were found in New Caledonia, in the southwest Pacific (4.4 μg.g^{-1}), the lowest in the Tuamotu archipelago, French Polynesia (0.7 μg.g^{-1}), the Solomon Islands (0.8 μg.g^{-1}), and Tuvalu (0.9 μg.g^{-1}), while moderately low concentrations were found in the Cook Islands (1.2 μg.g^{-1}), in Niue (1.3 μg.g^{-1}) and in the Marshall Islands (1.7 μg.g^{-1}) (Fig. 5A). Despite these large differences across the regions, the concentration of Hg in coral reef fish in PICTs was generally low.

The areas with the highest concentrations of glyphosate in coral reef fish were mainly located in the central and southern Pacific, such as the Fiji Islands (198.0 ng.g^{-1}) and Tonga (178.5 ng.g^{-1}). Elevated concentrations were also found in the western part of the Pacific basin, including the Solomon Islands (159.9 ng.g^{-1}), Papua New Guinea (142.9 ng.g^{-1}), New Caledonia (132.0 ng.g^{-1}), and even in some eastern parts, as in Tahiti, French Polynesia (120.3 ng.g^{-1}). Conversely, areas with the lowest glyphosate concentrations did not show a clear spatial pattern, such as Niue (54.9 ng.g^{-1}), Nauru (31.9 ng.g^{-1}) and the Cook Islands (28.5 ng.g^{-1}) (Fig. 5B).

4. Discussion

By combining fish community data with contamination measurements, this work constitutes, to our knowledge, the most extensive

Table 1
Summary of the most important drivers explaining MTEs and POPs concentrations in coral reef fish.

	Rank 1	Rank 2	Rank 3	Rank 4	Rank 5
MTEs					
Ag	GDP per capita (44.4 %)	SST max (16.9 %)	Land 3 km (15.8 %)	Reef 30 km (11.8 %)	Land30km (11.1 %)
As	Diet (30.0 %)	Size (27.0 %)	Reef 30 km (km2) (19.1 %)		
Cd	Land 3 km (68.1 %)	Size (14.4 %)			
Co	Schooling (21.0 %)	Land 30 km (17.7 %)	Size (13.6 %)	Reef 30 km (10.4 %)	
Cr	Land 3 km (20.4 %)	Reef 30 km (19.5 %)	Agricultural land (16.0 %)	Human gravity (15.1 %)	SST max (13.8 %)
Cu	Agricultural land (34.2 %)	Size (23.8 %)	Pesticides trade (22.0 %)	Reef 30 km (13.4 %)	
Fe	Agricultural land (31.2 %)	Land 3 km (30.4 %)	Reef 30 km (20.1 %)	Size (13.8 %)	
Hg	Size (34.9 %)	Diet (28.7 %)	SST max (11.8 %)		
Mn	SST max (23.8 %)	Size (20.1 %)	GDP per capita (16.5 %)	Land 3 km (13.9 %)	Reef30km (10.8 %)
Ni	SST max (31.0 %)	Size (21.5 %)	Schooling (13.7 %)		
Pb	Size (62.1 %)	Land 30 km (29.3 %)			
Se	Size (25.2 %)	Diet (22.3 %)	Land 3 km (15.3 %)	SST max (11.0 %)	Pesticides trade (10.6 %)
Vn	Total population (32.2 %)	GDP per capita (22.9 %)	Land 3 km (13.4 %)	Agricultural land (11.5 %)	Size (10.9 %)
Zn	Total population (35.3 %)	Size (25.1 %)	Land 3 km (22.5 %)	GDP per capita (17.1 %)	
POPs					
Aldrin	Size (29.7 %)	Schooling (11.5 %)			
Atrazine	SST max (35.2 %)	Land 3 km (22.2 %)	Size (19.3 %)	Reef 3 km (10.9 %)	
α-chlordane	Reef 3 km (27.6 %)	Size (23.6 %)	Total population (21.6 %)	Schooling (15.6 %)	Diet (11.5 %)
cis-chlordane	Size (26.6 %)	Reef 3 km (21.5 %)	Land 3 km (12.9 %)	Human gravity (12.7 %)	
Diazinon	Size (26.3 %)	SST max (22.0 %C)	Reef 3 km (14.4 %)	Human gravity (14.0 %)	Pesticides trade (12.7 %)
Dieldrin	Size (28.1 %)	Land 3 km (14.2 %)	SST max (13.5 %)	Schooling (12.1 %)	Diet (11.4 %)
α- endosulfan	Land 3 km (34.4 %)	Human gravity (28.1 %)	Size (22.4 %)		
β-endosulfan	Land 30 km (36.0 %)	Size (29.1 %)	Pesticides trade (13.2 %)		
Endrin	Size (38.4 %)	Reef 3 km (18.0 %)	SST max (16.1 %)	Total population (10.5 %)	
Glyphosate	Size (42.4 %)	Total population (17.5 %)	Reef 30 km (13.2 %)		
Heptachlor	Size (37.3 %)	Human gravity (15.1 %)	Agricultural land (15.0 %)	Land 3 km (14.0 %)	
Heptachlor epoxide A	Size (34.9 %)	Pesticides trade (14.1 %)	Human gravity (11.8 %)	Reef 3 km (10.8 %)	Land 3 km (10.2 %)
Heptachlor epoxide B	Total population (25.5 %)	Land 3 km (18.8 %)	Agricultural land (16.5 %)	GDP per capita (14.0 %)	
Isodrin	Size (35.4 %)	Total population (15.3 %)	Schooling (12.4 %)		
Lindane	Size (29.3 %)	SST max (24.8 %)	Reef 3 km (13.4 %)	Pesticides trade (10.1 %)	
Linuron	Land 30 km (30.8 %)	SST max (21.2 %)	Size (20.6 %)	Pesticides trade (16.0 %)	
Malathion	Size (24.5 %)	Pesticides trade (20.4 %)	SST max (14.0 %)	GDP per capita (13.1 %)	Diet (10.8 %)
pp'-DDD	Size (27.7 %)	Reef 3 km (25.0 %)	SST max (18.4 %)	Land 3 km (11.5 %)	Diet (10.5 %)
pp'-DDE	Size (41.4 %)	Reef 30 km (13.4 %)	SST max (12.6 %)	Schooling (12.5 %)	Reef 3 km (10.7 %)
pp'-DDT	SST max (49.6 %)	Agricultural land (21.1 %)			
Simazine	Size (30.3 %)	Total population (21.8 %)	Pesticides trade (14.0 %)	GDP per capita (11.7 %)	
Σ PCBs	Size (29.0 %)	SST max (18.5 %)	Land 30 km (16.3 %)	GDP per capita (13.7 %)	Total population (11.7 %)

Only contributions >10 % were mentioned. SST in °C, land, agricultural and reef areas in km^2, size in cm.

research on fish contamination in coral reef ecosystems, involving the most common and potentially deleterious contaminants at the scale of the tropical Pacific Ocean. Our results contribute significantly to the understanding of the mechanisms underlying the distribution of both organic and inorganic compounds. We revealed low/modest levels of contamination, but widely dispersed across the Pacific Ocean, although our predictive mapping approach is to be considered as an important step but requiring validation by new contaminant analyses, notably (but not only) in unsampled PICTs (see Section 4.1 below). For most compounds, the modeled concentrations were generally higher in the central and southwest Pacific, while lower in the eastern part of the basin, especially in French Polynesia, the Cook Islands, and Rapa Nui.

4.1. Methodological limits

Any modeling process involves simplifications, sometimes approximations and possible biases depending on model parameters and input variables. Although the variables influencing contamination are solidly established in our BRTs modeling on fish contamination data, it is important to acknowledge that the contamination maps produced for fish communities at the South-Pacific scale were model predictions, and should be treated with some caution. The first reason is that our sampling, while covering a relatively large number of 146 species, was not spatially and temporally homogeneous. Rather, it reflected only a modest proportion of local diversity of coral reef fish (for instance ~1450 species in New Caledonia, Letourneur et al., 2023). However, the species analyzed belong to those that globally make up the largest proportion of total density and biomass, and are among those most consumed by human populations in PICTs, making this species selection globally coherent. The second reason is that, in the absence of direct measurements of fish contamination in many PICTS, the assessments provided by our mapping are still based on the projection of values measured elsewhere to unassessed coral reef ecosystems. In addition, the sites sampled in the 9 PICTs are partial representations of the diversity of coral reef habitats in these countries. As with any modeling, many of the local variables that would be incorporated into the model might influence the processes studied and it is therefore likely that some variables will be over- or underestimated in view of the various local realities. Nevertheless, our exploratory predictive mapping produced the best indication to date of what might plausibly be coral reef fish contamination levels in the South Pacific on the basis of ~1000 fish analyzed for a wide range of contaminants. However further investigations on islands other than those we sampled are needed to refine our model and its predictive power. With these limitations in mind, our analyses nevertheless allow exploring below the mechanisms likely to explain the patterns observed, both for fish contamination drivers and for exploratory mapping on a Pacific scale.

4.2. Factors influencing fish contamination

BRT allowed us to explore the complex interactions between various predictive variables (e.g. related to coral reef environments, biogeography, human influence, and fish biology) to explain the variation in contaminant concentrations across space and species. Our models explained a substantial portion of the global variation, with cross-validated r^2 ranging from 60 % to 87 % for the entire groups of

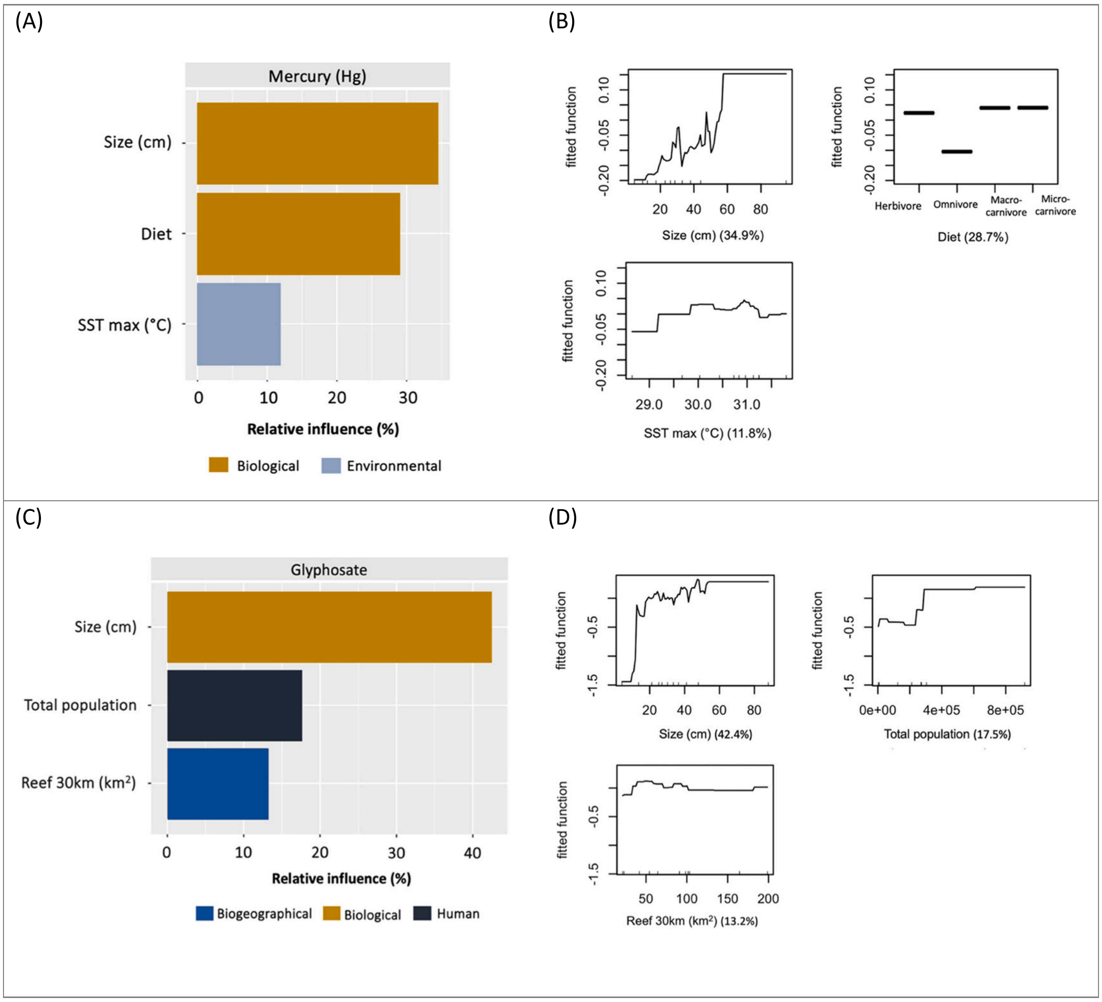

Fig. 3. (A) Summary of the relative contributions (%) of the three most significant variables (i.e., >10 %) used in BRT explaining Hg contamination in coral reef fish (CV = 0.75) and, (B) their three fitted functions (ranked by percentage of relative influence from left to right). (C) Summary of the relative contributions (%) of the three most significant variables (i.e., >10 %) used in BRT explaining glyphosate contamination in coral reef fish (CV = 0.65) and, (D) their three fitted functions (ranked by percentage of relative influence from left to right).

contaminants. One of the main outcomes of our modeling approach is the consistent and predominant role of fish size as a key-variable influencing contaminant concentrations. These results align with previous research showing that fish size, as a proxy of age, plays a crucial role in the bioaccumulation of some MTEs (Monteiro and Lopes, 1990; Zhang and Wang, 2007; Jonathan et al., 2015; Fey et al., 2019). Similarly, contamination by PCBs can vary depending on many parameters such as species, fish body size, locations, and environmental conditions (Bayarri et al., 2001; Storelli, 2008; Fey et al., 2019). Likewise, the rate of pesticide bioaccumulation can also vary based on species, fish size, pesticide structural composition, and the extent of contamination in associated aquatic systems (Banaee, 2013; Rohani, 2023). Biogeographic variables, especially land area and coral reef area, also played a significant role as drivers of contamination, suggesting strong interactions between terrestrial/coastal and marine ecosystems. Indeed, human activities in coastal areas adjacent to coral reefs generate pollution that may affect the entire food web (Mora et al., 2011). For example, overgrazing, deforestation, and agriculture in watersheds alter the nitrogen cycle, introduce pollutants, and increase sedimentation rates on coastal ecosystems. These terrestrial activities ultimately degrade benthic communities, reducing coral cover and altering the structure and functioning of the coral reef ecosystem (Mora et al., 2011). Our analysis of concentration levels of MTEs and POPs revealed a generally fluctuant effects of variables related to human activity. However, their explanatory power was on average relatively high (relative average contributions >10 %, depending on the contaminant). For example, human gravity appears to play a significant role in contamination at the community scale, although its impact was not as pronounced as at the individual level. There is evidence of increasing anthropogenic activity in PICTs altering marine environments such as land degradation on watersheds (Atkinson et al., 2016; Wairiu, 2017), plastic debris (Markic et al., 2018), sewage discharges (Ford et al., 2017), uncontrolled fishing activities (Veitayaki et al., 1995), and even occasional oil spills (Dutra et al., 2018).

We paid particular attention to Hg and glyphosate due to potential risks to public health. Mercury raises particular concern in marine environmental studies due to its sources being both natural and anthropogenic (Chouvelon et al., 2009; Eagles-Smith et al., 2018). Our results indicate that biological variables, especially individual body size and diet, were the main determinants of Hg concentration, suggesting bioaccumulation in fish. This is consistent with numerous studies showing that Hg in marine organisms can vary based on biological and environmental variables such as age, size, diet, and geographical origin (e.g., Burger et al., 2007; Žižek et al., 2007; Briand et al., 2018; Fey et al., 2019). Size is a well-known variable correlated with Hg concentrations in fish (Monteiro and Lopes, 1990; Mathieson and McLusky, 1995; Adams, 2004; Chouvelon et al., 2009; Briand et al., 2018). Furthermore, SST was correlated with higher Hg concentrations, suggesting an

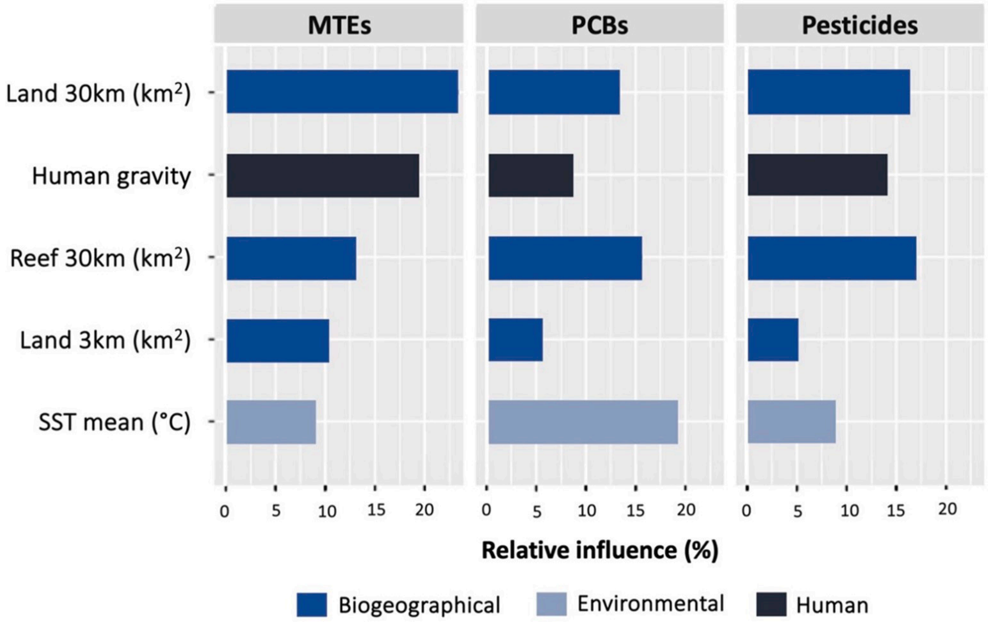

Fig. 4. Summary of the average relative contributions (%) of the five most significant variables (i.e., >10 %) used in BRT explaining their relative influence on extrapolated coral reef fish contamination (See Suppl. Fig. S2 for variables <10 %).

important role of temperature in physiological processes potentially involved in the basal metabolism of fish. This is consistent with other studies showing that biomagnification patterns in fish are complex and vary depending on food resources, feeding habitats, species metabolic activity, and physicochemical conditions in the water column (e.g., Lavoie et al., 2013; Le Croizier et al., 2016), which can directly affect the bioavailability of Hg as well as other MTEs (Tesser et al., 2021). Additionally, the organic form of Hg, i.e. methyl-Hg, usually representing >90 % of the Hg load in fish (Bloom, 1992; Sadhu et al., 2015) can naturally be found in anoxic sediments even at low depths, or form in semi-deep layers of water (~500–800 m) through complex biochemical processes (Houssard et al., 2019) mostly driven by microorganisms (Heimbürger et al., 2010), and through connections with solid and gaseous volcanic emissions during eruptions (Kumar-Roiné et al., 2022), as in Tongian and Vanuatu archipelagos. For instance, volcanic plumes released significant amounts of Hg from the Ambrym and Yasur volcanoes (Vanuatu) (Allard et al., 2016; Bagnato et al., 2011). However, a large part (~two-third) of the Hg in circulation comes from anthropogenic emissions, which have tripled concentrations in the euphotic zone since the pre-industrial era. The Hg emitted in populated areas is distributed worldwide, given its residence time in the atmosphere (6 months to a year) and the remission of Hg by the ocean, which helps disperse it everywhere (Sonke et al., 2023). It should be also pointed out that in oligotrophic environments, such as in Pacific Ocean waters, at similar concentrations (or contamination) compared to more productive environments, bioaccumulation and therefore biomagnification of Hg is more intense (Chouvelon et al., 2018).

Pesticides, a group of widely used chemical compounds to hinder the development of various pests such as insects, weeds, and others (Abhilash and Singh, 2009; Kim et al., 2017), can enter into water bodies through various pathways, including runoff from non-irrigated agricultural land and the drainage system (Rabiet et al., 2010; Bereswill et al., 2012; Stehle and Schulz, 2015). They exert harmful effects on living aquatic organisms, including fish (Gilliom, 2007; Malaj et al., 2014). In the case of glyphosate, fish size was also the most important variable explaining its concentration. This is one of the most controversial pesticides to date, suggesting a bioaccumulation process and perhaps even biomagnification. The total population and reef area in a 30 km buffer zone also played significant roles in explaining glyphosate concentrations, suggesting both a more or less general use of glyphosate and not just for agricultural purposes (for instance, also for public and private gardens), as well as its ability to disperse over such a spatial scale. The extent of pesticide introduction into aquatic systems depends on several geological and climatic factors such as the slope of the terrain and other watershed characteristics, hydrography, precipitation intensity, and soil moisture (Schulz, 2004). With the rapid development of agricultural systems to meet the increasing population demand, the use of multiple pesticides has also increased significantly (Hakeem, 2015). The use of agricultural pesticides in Oceania has seen significant growth, more than tripling between 1990 and 2021 (FAO, 2023a, 2023b). Although the quantity of pesticides used per hectare of cultivated land (1.66 kg/ha) and in relation to the value of agricultural production (0.83 kg/1000 USD) is relatively low, the quantity per capita is notably high, reaching 1.30 kg per person. This intensive and (un)controlled use, has deteriorated aquatic ecosystems in various ways, including leaching, drift, runoff, and drainage (Cerejeira et al., 2003). These results emphasize the need for particular attention to agricultural practices and the extensive use of this chemical substance, even by people to combat pests in their gardens for example. Although the values were below the health risk thresholds (the regulatory thresholds established by health agencies and institutions are 0.3 $ng.g^{-1}$ for aldrin, chlordane, heptachlor, and dieldrin, and 0.25 $ng.g^{-1}$ for glyphosate, expressed as maximum residue limits in $ng.g^{-1}$of fresh mass), the fact remains that we have observed and modeled distribution of this contaminant, as well as other MTEs and POPs, over a large part of the Pacific, which is particularly concerning for at least two major reasons. The first reason is that various PICTs are not heavily industrialized and have little or no highly polluting industry or other activities. This suggests that, although local impacts should not be neglected, most of the contamination could come from potentially (very) distant areas, through ocean currents and/or atmospheric deposits. The second reason is that various industrial activities, including the use of fossil fuels and other non-renewable resources (e.g., mining activities etc.), do not seem to be slowing down. It is therefore reasonable to assume that contamination, in general, and undoubtedly more markedly for certain elements, could increase and potentially will reach or exceed health risk thresholds in a near future.

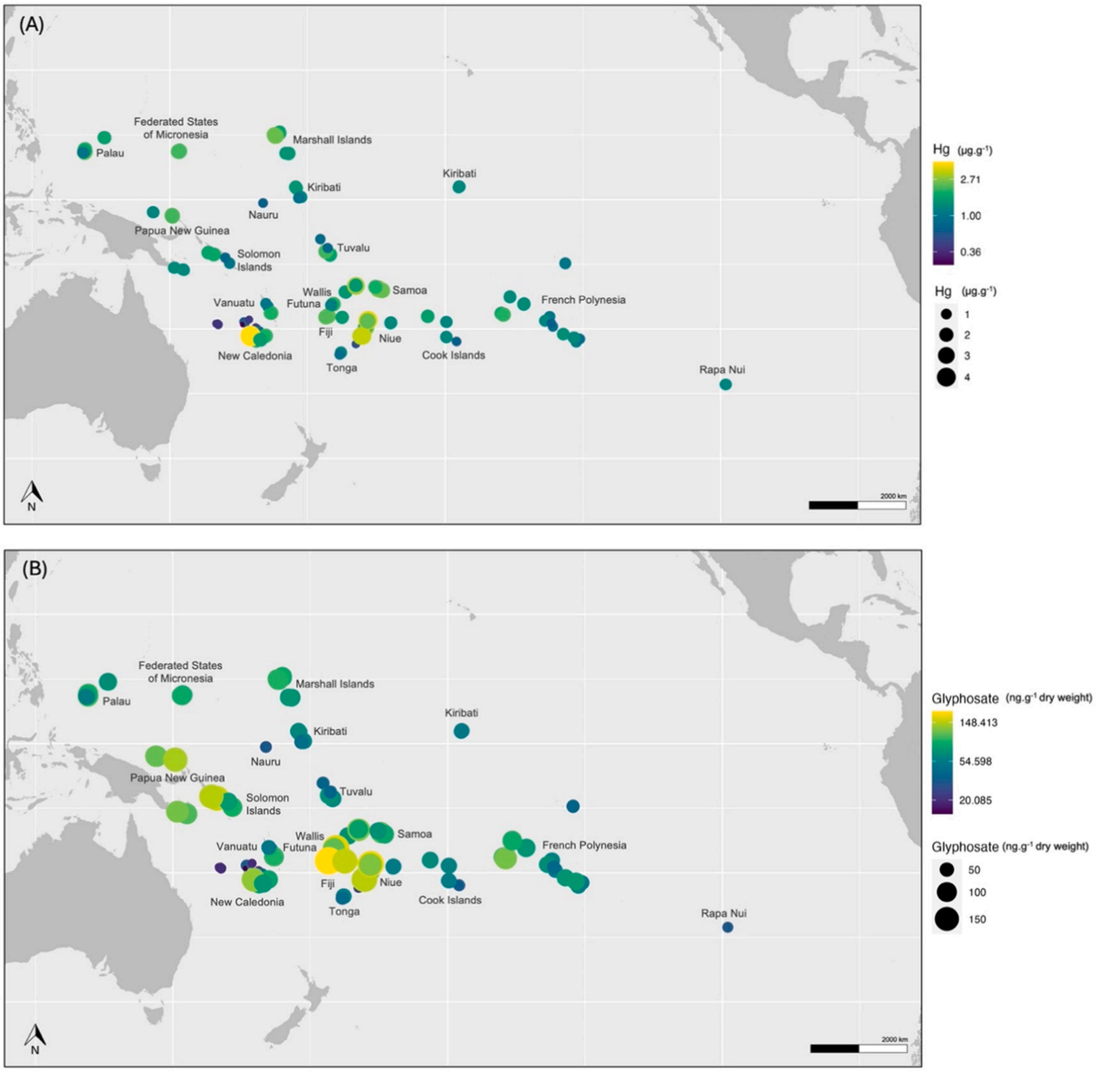

Fig. 5. Maps of predicted Hg (A) and (B) glyphosate concentration in coral reef fish using a BRT model with environmental, biogeographic and human variables as predictors. (See Suppl. Figs. S3 and S4 for other contaminants).

4.3. *Spatial patterns of contaminants in Pacific coral reef fish*

Some of the results of this large-scale mapping (i.e., 17 PICTs) are somewhat different from those obtained at lower spatial scales (i.e., 9 PICTs). In particular, we found that factors implying a larger spatial scale were more important as drivers of contamination at the Pacific scale (e.g., land 30 km > land 3 km in the 2nd part of our analysis, versus the opposite in the 1st part, land 3 km > land 30 km). This tendency toward smoothing is likely due to the change in scale, since the analysis of drivers (land surfaces close to coral reefs, in this case) on the contamination of the fish analyzed was carried out at the scale of the sites sampled, whereas the analysis at the Pacific scale was spatially more extensive and involved communities and not just the ~1000 individuals analyzed. It is therefore not surprising that the "medium" spatial scale (30 km) was more influential than the "fine" scale (3 km). At the sampling site scale, the accuracy of observations and measurements was logically greater, and therefore the robustness of interpretation was better. But this also implies that a change in spatial scale can attenuate or even modify the results, and therefore requires greater care in interpretation. More detailed work at local scale would be needed to validate or invalidate this hypothesis.

Fish significantly contribute to the economies of both subsistence and market-based PICTs, with fish consumption rates by Pacific islanders among the highest globally (55 to 110 kg per person per year; Bell et al., 2009). Thus, bridging the knowledge gap regarding contaminant levels in coral reef fish is crucial. One of the most important outcomes of this study is the creation of maps representing the modeled quantities of various contaminants per 100 g of coral reef fish in several sites from 17 PICTs.

Overall, modeled quantities were generally higher in the central and southwest Pacific, while lower in the eastern part of the basin, especially in French Polynesia, the Cook Islands, and Rapa Nui. For example, Hg concentrations in fish were generally low across all studied sites, except in certain places where concentrations reached higher values, such as in New Caledonia. In the absence of significant current anthropogenic inputs, it suggests that geothermal activities in the study area could be one of the potential sources of Hg. Documentation on hydrothermal vents in New Caledonia is very limited (Maurizot et al., 2020; Kumar-Roiné et al., 2022). However, no study has been undertaken to assess if any of these vents naturally release fluids containing Hg into seawater, although such kind of Hg emissions have been reported from the Tonga-Kermadec subduction zone (Lee et al., 2015). Another possible source for Hg contamination is volcanic activity in the Vanuatu arc region. Volcanic plumes can be rich in Hg and the release of significant amounts of Hg from volcanoes in Vanuatu (Allard et al., 2016; Bagnato et al., 2011) highlights the role of geophysical processes in the geographical distribution of Hg.

Agricultural activities represent another source of pollution in the region. Poor land management practices and the loss of agricultural land to other economic activities have led to an increased use of pesticides and fertilizers. For examples, the use of pesticides by sugarcane producers in Fiji (Szmedra, 1999) or anthropogenic nutrient incorporation of the intertidal fauna at the cost of Rapa Nui (Zapata-Hernandez et al., 2022) are well documented. Extensive and improper use of pesticides and fertilizers contributes to soil and groundwater contamination. It is estimated that up to 70 % of applied pesticides, most of which contain heavy metals, are washed away and leach into the soil and water, resulting in excessive contamination by pesticide residues in coastal ecosystems (UNESCAP, 2002). Areas with the highest concentrations of glyphosate in Pacific coral reef fish were primarily located in high islands with extensive overall areas devoted to agriculture, such as Fiji, the Solomon Islands, Tonga, and New Caledonia. The high concentrations found in fish could be associated with the use of this herbicide, sometimes in uncontrolled quantities and/or in an illegal context, in surrounding areas for agriculture or other agriculture-related practices.

Overall, all our maps highlighted geographical variations in contaminant concentrations in Pacific coral reef fish, which can have significant implications for the health of marine ecosystems and the local populations dependent on these resources. The information obtained from these maps can be invaluable in guiding conservation and management efforts for marine resources in the region, in particular for fishing activities including auto-consumption.

These findings contribute to a better understanding of the complex dynamics influencing contaminant concentrations in Pacific coral reef fish. Further research in this field could help refine our understanding of these intricate relationships and inform targeted conservation strategies. On the other hand, even if contaminants were at relatively low levels, thereby limiting the current health risk for humans, their interaction can potentially increase their hazardous effects (‘cocktail effect’) (Fey et al., 2019). This aspect should be further explored in the future, keeping in mind that ongoing industrial developments and climate changes could alter the patterns found. This perspective underscores the importance of closely monitoring the evolution of contaminant concentrations in marine ecosystems, considering the complex interactions between different pollutants. It emphasizes the need for a proactive approach to mitigate potential risks to human health and the ecological balance of Pacific coral reefs.

5. Conclusion

Our study on contamination by MTEs and POPs in Pacific coral reef fish provide new and crucial insights into the contamination levels of these marine ecosystems. Using statistical models, we shown that several variables, including biological, environmental, related to biogeography, or influenced by human activities, explained the concentrations of contaminants. Individual fish size was a major determining variable in contaminant concentrations, with significant variations in all cases. Additionally, the presence of land in a 3 km buffer zone and SST have also demonstrated their importance to understand contamination. Contamination mapping, although to be considered with caution, revealed moderate contamination levels, but widely dispersed across the Pacific for each contaminant, highlighting the complexity of their distribution. The variables influencing these concentrations vary depending on the contaminant, ranging from biogeographical variables to the biological characteristics of the fish. Ultimately, our research underscores the importance of considering a diverse range of variables for the management and monitoring of contamination in Pacific coral reef fish and emphasizes the urgency of continuous monitoring, both spatially and temporally.

CRediT authorship contribution statement

Noreen Wejieme: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Laurent Vigliola:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Conceptualization. **Valeriano Parravicini:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Formal analysis, Conceptualization. **Javier Sellanes:** Writing – review & editing. **Emmanuel Wafo:** Writing – review & editing. **German Zapata-Hernandez:** Writing – review & editing. **Paco Bustamante:** Writing – review & editing, Methodology, Formal analysis. **Yves Letourneur:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.177914.

Data availability

The data that has been used is confidential.

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