

Comparative Analysis of Metals in Seafood from Rio Doce Coastal Areas and Regional Fish Markets

William Odle^{1*}, Vitor Alencar Freitas Da Silva², Lorrana Moraes Marques², Jacob Oehrig¹, Negin Kananizadeh¹, Diogo Fonseca Alves Gaspar³, Michael Wild¹

¹Independent Researcher, New Fields Atlanta, LLC., Atlanta, USA

²Independent Researcher, New Fields Brasil Consultoria Ambiental LTDA, Belo Horizonte, Brazil ³Independent Researcher, New Fields United Kingdom, LLC., Greengates, Bradford, UK

Email: *bodle@newfields.com

How to cite this paper: Odle, W., Da Silva, V.A.F., Marques, L.M., Oehrig, J., Kananizadeh, N., Gaspar, D.F.A. and Wild, M. (2023) Comparative Analysis of Metals in Seafood from Rio Doce Coastal Areas and Regional Fish Markets. *Journal of Environmental Protection*, **14**, 859-887. https://doi.org/10.4236/jep.2023.1410048

Received: September 21, 2023 Accepted: October 24, 2023 Published: October 27, 2023

Copyright © 2023 by author(s) and Scientific Research Publishing Inc. This work is licensed under the Creative Commons Attribution International License (CC BY 4.0). http://creativecommons.org/licenses/by/4.0/

Abstract

On November 5, 2015, the Fundão Dam breached (the "Event"), releasing water, tailings, dam construction material, and debris to downstream watercourses. Over 20 million cubic meters of fine particles from the tailings, as well as scoured soil and sediments, reached the Rio Doce estuary and surrounding marine area 17 days later. Fishing was banned by the Federal Court of Espírito Santo in February 2016. The fishing ban area included the coastal area near the mouth of the Rio Doce and seaward to a depth of 25 meters, south to Barra do Riacho, and north to Degredo Beach. In June 2019, the Brazilian health agency, Anvisa, published a risk assessment for fish consumption which recommended daily consumption limits for fish of 200 grams for adults and 50 grams for children for both continental and coastal areas of the Rio Doce basin. Comparative analyses were performed between metal concentrations in marine fish and crustaceans collected in the banned fishing area to reference areas and commercialized seafood markets along the Brazilian coast. The results reveal that metals detected in seafood tissue collected in the fishing ban area are not significantly different than other reference areas or from commercially available seafood. This result indicates that elevated metal concentrations in seafood are a regional problem, unrelated to the Event. Higher concentrations of metals in fish in reference areas outside of the fishing ban area as well as in commercial seafood markets demonstrates that the risk management approach of a localized fishing ban is ineffective for reducing risk to the population related to seafood consumption.

Keywords

Fish Consumption, Mine Tailings, Fundão Dam, Marine Environment,

Brazilian Coast, Fishing Ban

1. Introduction

On November 5, 2015, the Fundão Dam, located within Samarco Mineração S.A.'s Germano Industrial Complex, in the municipality of Mariana, state of Minas Gerais, breached, releasing 40 million cubic meters (m³) of tailings, water, dam construction materials, and debris to downstream watercourses and adjacent areas. The resulting wave (henceforth referred to as the "Event") traveled over 670 kilometers (km) and reached the Rio Doce estuary and nearby marine area 17 days later. Over 20 million m³ of water and fine particles from the tailings, as well as scoured soil and sediments near Fundão Dam, reached the Atlantic coastline [1].

To prevent potential impacts to human health related to the Event, the Federal Court of Espírito Santo [case No 0002571-13.2016.4.02.5004 (2016.50.04.002571-0)] banned fishing along the Atlantic coastline on February 17, 2016. This area (henceforth referred to as the "Fishing Ban Area") ranges from the mouth of the Rio Doce, east or seaward to a depth of 25 meters, south to Barra do Riacho, and north to Degredo Beach, as shown on **Figure 1**. At the time of the ban, concentrations of metals in fish from continental and marine areas were not well established.



Figure 1. Delimitation of the fishing ban area.

This article presents a statistical approach to compare concentrations of metals in samples of marine fish and crustaceans collected in the Fishing Ban Area, to samples collected in a similar reference area (Jequitinhonha River Mouth) and a control area (Abrolhos Archipelago Region). Statistical comparisons were also made to fish in commercial markets along the Brazilian coast.

2. Methodology

2.1. Key Parameter Selection

In June 2019, the Brazilian health regulatory agency (Anvisa) published a human health risk assessment for fish consumption in the Rio Doce basin and coastal area [2]. Anvisa identified mercury and lead as the metals that pose the greatest concern to human health related to the consumption of fish. Anvisa recommended restricting the daily consumption of fish to 200 grams (g) for adults and 50 g for children for both continental and coastal areas of the Rio Doce basin.

Several articles addressed direct and indirect water quality impacts from the Event to both fresh [3] [4] and marine environments [5]. The statistically determined parameters from these articles were aluminum, arsenic, barium, iron, lead, manganese, and nickel. The Brazilian Ministry of Health also identified arsenic, cadmium, mercury and lead to be of particular importance based on maximum allowable concentrations in fish [6].

The statistical evaluation presented assesses these, combined, nine key metals in both fish and crustacean samples collected from marine and coastal regions after the Event.

2.2. Data Compilation

As shown in **Table 1**, the statistical analyses performed by the authors used a dataset of 22,659 analytical results, 2976 samples (1995 fish and 981 crustaceans) of 73 unique species of marine fish and 10 unique species of crustaceans collected between 2018 and 2020 in both wet (October to March) and dry (April to September) seasons. All chemical analysis results are from muscle tissue measured on a wet weight basis.

The dataset includes samples compiled by five research groups consisting of universities, institutes, and consultants. The dataset used in this article is available for download at a public repository (<u>https://osf.io/u7fk6/</u>). Figure 2 presents the spatial distribution of the samples.

The data usability requirements were verified according to Brazilian and United States Environmental Protection Agency (US EPA) guidelines [7] [8] and data were validated prior to analysis. Outlier analyses were performed for marine fish and crustaceans, which excluded five crustacean results from the assessment, representing 0.02% of the dataset. Results that were greater than three standard deviations from the mean were considered outliers. Details on the outlier analyses are provided, separately, in **Table S1** as supporting information.





Figure 2. Spatial distribution of the samples used in the statistical analysis.

Fable 1. Fish and crustacea	n sample counts within each	location, year, and season.
-----------------------------	-----------------------------	-----------------------------

-	Fish									
Location		2018			2019			2020		Total
	Dry ^a	Wet ^a	Total	Dry	Wet	Total	Dry	Wet	Total	TOtal
Abrolhos—Nature Reserve		61	61		74	74				135
Commercially Available—Markets	96		96		362	362	96	12	108	566
Jequitinhonha River Mouth—Anthropogenic Reference Area		84	84	45	80	125		63	63	272
Within Fishing Ban Area		179	179	671	20	691		152	152	1022
Total	96	324	420	716	536	1252	96	227	323	1995
		Crustaceans								
Location		2018		2019				2020		
	Dry ^a	Wet*	Total	Dry	Wet	Total	Dry	Wet	Total	Total
Abrolhos—Nature Reserve		16	16		10	10				26
Commercially Available—Markets	48		48		169	169	42	12	54	271
Jequitinhonha River Mouth—Anthropogenic Reference Area		45	45	75	40	115		47	47	207
Within Fishing Ban Area		75	75	357	7	364		38	38	477
Total	48	136	184	432	226	658	42	97	139	981

a. Note: Dry (April to September) and Wet (October to March) seasons.

The dataset was divided into two groups based on those reported in the Brazilian Family Budget Survey [9]: marine fish and crustaceans. Fish comprise 67% of samples and crustaceans comprise 33% (see supporting information **Table S2** and **Table S3**). A total of 73 species of marine fish were sampled. The most sampled species (70% of total fish samples), consolidated by common name, were: croaker (32%), weakfish (15%), sea catfish (14%), and stardrum fish (9%). Ten species of crustaceans were sampled and included: shrimp (92%), crab (6.0%), and lobster (2%).

Half of the reporting limit was used for results below laboratory reporting limits. Average values were calculated for individual fish samples that were split and analyzed by different labs.

2.3. Statistical Analysis

Comparative analyses of the nine key metal concentrations in marine fish and crustaceans between samples collected in the Fishing Ban Area to other locations were performed. The locations used for these comparisons are described below:

- Abrolhos—Nature Reserve Control Area located 200 km north of the mouth of the Rio Doce and outside of the influence of the Event [10];
- Jequitinhonha River—Anthropogenic Reference Area located 400 km north in a similar urban and estuarine environment as the mouth of the Rio Doce [11];
- Commercialized Fish (Interstate), and Commercialized Fish (Regional) —Commercialized Fish include fish and crustacean samples from markets located along the Brazilian coast.

Samples collected from fish were heterogeneous and did not present a balanced spatial and temporal distribution between seasons and locations for each group. Therefore, a statistical approach was used to evaluate potential seasonal differences in metal concentrations of marine fish and crustacean samples using the Mann-Whitney U (MWU) test. Comparison of metal concentrations in different marine locations used the Games-Howell post-hoc analyses.

Previous studies show that mercury can bioaccumulate within trophic levels, especially in piscivore fish species [12] [13] [14] [15]. Therefore, mercury analyses were limited to piscivores collected during the wet season when the highest mercury concentrations were observed.

As demonstrated by Rodrigues *et al.* (2010) [16], different fish species may present different behaviors in relation to bioaccumulation, which may be related to their feeding and migratory habits, changes in metabolism throughout life, among other reasons. Additionally, morphometric measurements (sample length and weight) generally correlate with the age of a given specimen [17], therefore indicating the specimen's exposure time to the environment. Thus, the correlation between the size of the fish and the concentration of a certain contaminant is a common occurrence for elements that bioaccumulate [17] [18] [19]. Therefore, fish genus/species-specific analyses for the genera that had the best sample

coverage among the locations investigated were also performed using metals concentrations normalized by fish length to address potential bioaccumulation differences related to size and/or age. To provide sufficient information for confirmatory analyses, fish genus/species specific analyses were performed for the genera that had the best sample coverage among the locations (**Table S2** and **Table S3** for fish and crustacean species, respectively). Five genera provided at least ten samples in the Fishing Ban Area, the commercially available markets, and at least one reference area: *Conodon (Conodon nobilis*—Grunt Fish), *Genidens (Genidens barbus* and *genidens*—Sea Catfish), *Macrodon (Macrodon ancylodon* and *atricauda*—Weakfish), and *Paralonchurus (Paralonchurus brasiliensis*—Croaker). Of these five genera, *Macrodon* is among the most captured genera of fish in Espírito Santo's marine area [20]. Tissue metal concentrations from these five genera were normalized by length to account for potential bioaccumulation differences related to size and/or age [17].

3. Results

For marine fish, all nine of the metals analyzed presented higher mean concentrations in the wet season, with six of them presenting statistically significant differences (**Table S4**). For crustaceans, the wet season had higher mean concentrations for all metals except arsenic and nickel, which had higher concentrations in the dry season, of which, only arsenic was significantly higher. Since wet season samples resulted in higher concentration of metals in almost all scenarios, these samples were selected for comparative location analysis. Commercialized Fish (Regional) were sampled in wet and dry seasons, however, all fish samples within Commercialized Fish (Interstate) were collected only in the dry season. Consequently, these data were not compared to wet season samples to avoid bias. Crustaceans were sampled in both wet and dry seasons in both interstate and regional markets.

Figure 3 presents average marine fish concentration and standard error bar graphs for each metal and location. Figure 4 presents the same graphs for crustaceans. Both graphs are limited to wet season samples. The Games Howell post hoc test results are shown in Table 2 and Table 3 for fish and crustaceans, respectively. As shown by Figure 3 and Figure 4, in all cases evaluated, at least one or more reference areas and/or commercially available seafood market presented higher average metal concentrations than samples from the Fishing Ban Area. Results that were statistically significantly higher than the Fishing Ban Area are denoted with an asterisk.

Figure 5 presents average mercury concentrations with standard error bars of piscivores samples collected in the wet season for each location. Games Howell post hoc test results for these samples are shown in **Table 4**. These analyses show that the Abrolhos control area (nature reserve) and commercially available fish have statistically significantly higher mercury concentrations compared to the Fishing Ban Area.

Analyte	Group 1	Group 2	p-Value	Significant	Direction
Aluminum	Fishing Ban Area	Abrolhos	6.96E-13	Significant	Fishing Ban Area > Abrolhos
Aluminum	Fishing Ban Area	Jequitinhonha	2.25E-01	Not Significant	
Aluminum	Abrolhos	Jequitinhonha	1.45E-01	Not Significant	
Arsenic	Fishing Ban Area	Abrolhos	5.85E-01	Not Significant	
Arsenic	Fishing Ban Area	Jequitinhonha	5.91E-13	Significant	Jequitinhonha > Fishing Ban Area
Arsenic	Fishing Ban Area	Market-Regional	8.44E-01	Not Significant	
Arsenic	Abrolhos	Jequitinhonha	0.00E+00	Significant	Jequitinhonha > Abrolhos
Arsenic	Abrolhos	Market-Regional	8.78E-01	Not Significant	
Arsenic	Jequitinhonha	Market-Regional	1.10E-12	Significant	Jequitinhonha > Market-Regional
Barium	Fishing Ban Area	Abrolhos	2.60E-02	Significant	Fishing Ban Area > Abrolhos
Barium	Fishing Ban Area	Jequitinhonha	8.48E-01	Not Significant	
Barium	Abrolhos	Jequitinhonha	3.40E-02	Significant	Jequitinhonha > Abrolhos
Cadmium	Fishing Ban Area	Abrolhos	3.27E-01	Not Significant	
Cadmium	Fishing Ban Area	Jequitinhonha	3.90E-02	Significant	Fishing Ban Area > Jequitinhonha
Cadmium	Fishing Ban Area	Market-Regional	5.00E-03	Significant	Market-Regional > Fishing Ban Area
Cadmium	Abrolhos	Jequitinhonha	2.29E-01	Not Significant	
Cadmium	Abrolhos	Market-Regional	1.92E-08	Significant	Market-Regional > Abrolhos
Cadmium	Jequitinhonha	Market-Regional	9.16E-12	Significant	Market-Regional > Jequitinhonha
Iron	Fishing Ban Area	Abrolhos	0.00E+00	Significant	Fishing Ban Area > Abrolhos
Iron	Fishing Ban Area	Jequitinhonha	8.90E-02	Not Significant	
Iron	Fishing Ban Area	Market-Regional	1.00E+00	Not Significant	
Iron	Abrolhos	Jequitinhonha	2.00E-02	Significant	Jequitinhonha > Abrolhos
Iron	Abrolhos	Market-Regional	6.22E-06	Significant	Market-Regional > Abrolhos
Iron	Jequitinhonha	Market-Regional	9.30E-02	Not Significant	
Lead	Fishing Ban Area	Abrolhos	9.99E-01	Not Significant	
Lead	Fishing Ban Area	Jequitinhonha	7.99E-01	Not Significant	
Lead	Fishing Ban Area	Market-Regional	0.00E+00	Significant	Market-Regional > Fishing Ban Area
Lead	Abrolhos	Jequitinhonha	8.56E-01	Not Significant	
Lead	Abrolhos	Market-Regional	0.00E+00	Significant	Market-Regional > Abrolhos
Lead	Jequitinhonha	Market-Regional	2.24E-10	Significant	Market-Regional > Jequitinhonha
Manganese	Fishing Ban Area	Abrolhos	7.05E-09	Significant	Fishing Ban Area > Abrolhos
Manganese	Fishing Ban Area	Jequitinhonha	5.40E-02	Not Significant	
Manganese	Fishing Ban Area	Market-Regional	0.00E+00	Significant	Fishing Ban Area > Market-Regional

 Table 2. Fish metal concentrations, games/howell post hoc results.

Continuea					
Manganese	Abrolhos	Jequitinhonha	8.53E-05	Significant	Jequitinhonha > Abrolhos
Manganese	Abrolhos	Market-Regional	1.53E-01	Not Significant	
Manganese	Jequitinhonha	Market-Regional	1.27E-05	Significant	Jequitinhonha > Market-Regional
Mercury	Fishing Ban Area	Abrolhos	2.54E-08	Significant	Abrolhos > Fishing Ban Area
Mercury	Fishing Ban Area	Jequitinhonha	2.15E-01	Not Significant	
Mercury	Fishing Ban Area	Market-Regional	0.00E+00	Significant	Market-Regional > Fishing Ban Area
Mercury	Abrolhos	Jequitinhonha	1.80E-09	Significant	Abrolhos > Jequitinhonha
Mercury	Abrolhos	Market-Regional	3.70E-02	Significant	Market-Regional > Abrolhos
Mercury	Jequitinhonha	Market-Regional	0.00E+00	Significant	Market-Regional > Jequitinhonha
Nickel	Fishing Ban Area	Abrolhos	1.43E-01	Not Significant	
Nickel	Fishing Ban Area	Jequitinhonha	3.87E-04	Significant	Jequitinhonha > Fishing Ban Area
Nickel	Fishing Ban Area	Market-Regional	4.00E-03	Significant	Market-Regional > Fishing Ban Area
Nickel	Abrolhos	Jequitinhonha	1.06E-05	Significant	Jequitinhonha > Abrolhos
Nickel	Abrolhos	Market-Regional	8.57E-04	Significant	Market-Regional > Abrolhos
Nickel	Jequitinhonha	Market-Regional	6.80E-01	Not Significant	



Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)

Figure 3. Location-specific concentrations of each metal in marine fish samples collected in the wet season.

Analyte	Group 1	Group 2	p-Value	Significant	Direction
Aluminum	Fishing Ban Area	Abrolhos	1.46E-01	Not Significant	
Aluminum	Fishing Ban Area	Jequitinhonha	2.00E-03	Significant	Jequitinhonha > Fishing Ban Area
Aluminum	Abrolhos	Jequitinhonha	9.24E-01	Not Significant	
Arsenic	Fishing Ban Area	Abrolhos	7.81E-10	Significant	Fishing Ban Area > Abrolhos
Arsenic	Fishing Ban Area	Jequitinhonha	6.25E-11	Significant	Jequitinhonha > Fishing Ban Area
Arsenic	Fishing Ban Area	Market-Regional	8.60E-02	Not Significant	
Arsenic	Fishing Ban Area	Market-Intrastate	3.13E-14	Significant	Fishing Ban Area > Market-Intrastate
Arsenic	Abrolhos	Jequitinhonha	4.10E-14	Significant	Jequitinhonha > Abrolhos
Arsenic	Abrolhos	Market-Regional	1.20E-09	Significant	Market-Regional > Abrolhos
Arsenic	Abrolhos	Market-Intrastate	9.92E-01	Not Significant	
Arsenic	Jequitinhonha	Market-Regional	1.00E-03	Significant	Jequitinhonha > Market-Regional
Arsenic	Jequitinhonha	Market-Intrastate	0.00E+00	Significant	Jequitinhonha > Market-Intrastate
Arsenic	Market-Regional	Market-Intrastate	1.20E-10	Significant	Market-Regional > Market-Intrastate
Barium	Fishing Ban Area	Abrolhos	2.28E-01	Not Significant	
Barium	Fishing Ban Area	Jequitinhonha	9.65E-01	Not Significant	
Barium	Abrolhos	Jequitinhonha	2.33E-01	Not Significant	
Cadmium	Fishing Ban Area	Abrolhos	9.92E-01	Not Significant	
Cadmium	Fishing Ban Area	Jequitinhonha	4.76E-06	Significant	Fishing Ban Area > Jequitinhonha
Cadmium	Fishing Ban Area	Market-Regional	2.55E-04	Significant	Market-Regional > Fishing Ban Area
Cadmium	Fishing Ban Area	Market-Intrastate	7.38E-04	Significant	Fishing Ban Area > Market-Intrastate
Cadmium	Abrolhos	Jequitinhonha	9.51E-01	Not Significant	
Cadmium	Abrolhos	Market-Regional	2.00E-03	Significant	Market-Regional > Abrolhos
Cadmium	Abrolhos	Market-Intrastate	9.93E-01	Not Significant	
Cadmium	Jequitinhonha	Market-Regional	6.88E-06	Significant	Market-Regional > Jequitinhonha
Cadmium	Jequitinhonha	Market-Intrastate	3.24E-01	Not Significant	
Cadmium	Market-Regional	Market-Intrastate	1.68E-05	Significant	Market-Regional > Market-Intrastate
Iron	Fishing Ban Area	Abrolhos	2.58E-01	Not Significant	
Iron	Fishing Ban Area	Jequitinhonha	3.27E-05	Significant	Jequitinhonha > Fishing Ban Area
Iron	Fishing Ban Area	Market-Regional	8.00E-03	Significant	Market-Regional > Fishing Ban Area
Iron	Abrolhos	Jequitinhonha	3.87E-01	Not Significant	
Iron	Abrolhos	Market-Regional	6.03E-01	Not Significant	
Iron	Jequitinhonha	Market-Regional	7.88E-04	Significant	Jequitinhonha > Market-Regional

 Table 3. Crustacean metal concentrations, Games/Howell post hoc results.

Continued					
Lead	Fishing Ban Area	Abrolhos	7.97E-01	Not Significant	
Lead	Fishing Ban Area	Jequitinhonha	1.79E-01	Not Significant	
Lead	Fishing Ban Area	Market-Regional	0.00E+00	Significant	Market-Regional > Fishing Ban Area
Lead	Fishing Ban Area	Market-Intrastate	2.61E-01	Not Significant	
Lead	Abrolhos	Jequitinhonha	1.00E+00	Not Significant	
Lead	Abrolhos	Market-Regional	6.87E-08	Significant	Market-Regional > Abrolhos
Lead	Abrolhos	Market-Intrastate	4.10E-01	Not Significant	
Lead	Jequitinhonha	Market-Regional	8.71E-10	Significant	Market-Regional > Jequitinhonha
Lead	Jequitinhonha	Market-Intrastate	3.73E-01	Not Significant	
Lead	Market-Regional	Market-Intrastate	5.01E-01	Not Significant	
Manganese	Fishing Ban Area	Abrolhos	7.90E-02	Not Significant	
Manganese	Fishing Ban Area	Jequitinhonha	1.75E-08	Significant	Jequitinhonha > Fishing Ban Area
Manganese	Fishing Ban Area	Market-Regional	3.65E-07	Significant	Market-Regional > Fishing Ban Area
Manganese	Abrolhos	Jequitinhonha	9.92E-01	Not Significant	
Manganese	Abrolhos	Market-Regional	8.06E-01	Not Significant	
Manganese	Jequitinhonha	Market-Regional	4.61E-01	Not Significant	
Mercury	Fishing Ban Area	Abrolhos	5.10E-02	Not Significant	
Mercury	Fishing Ban Area	Jequitinhonha	3.28E-01	Not Significant	
Mercury	Fishing Ban Area	Market-Regional	1.61E-01	Not Significant	
Mercury	Fishing Ban Area	Market-Intrastate	1.81E-14	Significant	Fishing Ban Area > Market-Intrastate
Mercury	Abrolhos	Jequitinhonha	5.00E-03	Significant	Abrolhos > Jequitinhonha
Mercury	Abrolhos	Market-Regional	3.00E-03	Significant	Abrolhos > Market-Regional
Mercury	Abrolhos	Market-Intrastate	2.50E-07	Significant	Abrolhos > Market-Intrastate
Mercury	Jequitinhonha	Market-Regional	9.60E-01	Not Significant	
Mercury	Jequitinhonha	Market-Intrastate	0.00E+00	Significant	Jequitinhonha > Market-Intrastate
Mercury	Market-Regional	Market-Intrastate	0.00E+00	Significant	Market-Regional > Market-Intrastate
Nickel	Fishing Ban Area	Abrolhos	2.80E-01	Not Significant	
Nickel	Fishing Ban Area	Jequitinhonha	1.40E-02	Significant	Jequitinhonha > Fishing Ban Area
Nickel	Fishing Ban Area	Market-Regional	9.19E-01	Not Significant	
Nickel	Abrolhos	Jequitinhonha	5.57E-01	Not Significant	
Nickel	Abrolhos	Market-Regional	3.51E-01	Not Significant	
Nickel	Jequitinhonha	Market-Regional	3.30E-01	Not Significant	



Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)

Figure 4. Location-specific concentrations of each metal in crustacean samples collected in the wet season.



Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)

Figure 5. Location-specific concentrations of mercury in samples of piscivorous fish collected in the wet season.

Analyte	Group 1	Group 2	p-Value	Significant	Direction
Mercury	Fishing Ban Area	Abrolhos	4.40E-06	Significant	Abrolhos > Fishing Ban Area
Mercury	Fishing Ban Area	Jequitinhonha	9.90E-02	Not Significant	
Mercury	Fishing Ban Area	Market-Regional	9.33E-15	Significant	Market-Regional > Fishing Ban Area
Mercury	Abrolhos	Jequitinhonha	2.78E-07	Significant	Abrolhos > Jequitinhonha
Mercury	Abrolhos	Market-Regional	7.50E-02	Not Significant	
Mercury	Jequitinhonha	Market-Regional	0.00E+00	Significant	Market-Regional > Jequitinhonha

Table 4. Mercury concentrations in piscivore fish, Games/Howell post hoc results.

For confirmatory analyses, Figures 6-9 presents graphs similar to those above but normalized by length for Conodon nobilis (Grunt Fish), Genidens barbus/genidens (Sea Catfish), Macrodon ancylodon/atricauda (Weakfish), and Paralonchurus brasiliensis (Croaker), respectively. The Games Howell post hoc test results for the confirmatory analyses are shown in Table S5. The Conodon results show that at least one or more reference area and/or commercially available seafood market have a higher average metal concentrations than samples from the Fishing Ban Area for arsenic, lead, manganese, and mercury. Cadmium and iron had higher concentrations in the Fishing Ban Area, but they were not statistically significant. In all cases evaluated for Genidens and Paralonchurus, at least one or more reference area and/or commercially available seafood market presented higher concentrations than samples from the Fishing Ban Area, with aluminum, arsenic, iron, manganese, mercury, and nickel statistically significantly higher. Macrodon had statistically significantly higher concentrations for aluminum, arsenic, barium, iron, and manganese in the Fishing Ban Area compared to the reference areas and/or commercially available seafood market. These metals, however, did not pose a human health risk associated with the consumption of marine seafood [2]. Macrodon samples had higher lead, mercury, and nickel concentrations in the commercially available seafood markets compared to the Fishing Ban Area and other reference areas, with lead and mercury being statistically significant. Table 5 provides a comparison of the statistical evaluation for each of the four genus/species reviewed. The genus/species analysis displays differences in results between the analytes.

These analyses demonstrate that, for all metals, there were statistically significant higher concentrations or statistically indistinguishable levels found in at least one reference area or a commercially available seafood market when compared to fish and crustaceans collected in the Fishing Ban Area. These results also corroborate those of Kananizadeh *et al.* (2023) [4] which determined that, after 2017, water quality results collected in marine waters near the Rio Doce mouth were statistically indistinguishable from those near other rivers.

The overall results reveal that metals concentrations in seafood tissue are regionally distributed with no evidence of a particular metal having a statistically enriched concentration in fish collected in the Rio Doce Mouth region associated



Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)

Figure 6. Location-specific concentrations of each metal normalized by length in samples of *Conodon Nobilis (Grunt Fish)* collected in the wet season.



Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)





Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p< 0.05); + Fishing Ban Area is significantly higher (p < 0.05)





Notes: Error bars represent standard error of mean; * Location is significantly higher than Fishing Ban Area (p < 0.05)

Figure 9. Location-specific concentrations of each metal normalized by length in samples of *Paralonchurus brasiliensis* (*Croaker*) collected in the wet season.

Genus/Species	Aluminum	Arsenic	Barium	Cadmium	Iron	Lead	Manganese	Mercury	Nickel
Conodon nobilis	NA	-1	NA	1	1	0	0	-1	0
Genidens barbus and genidens	-1	-1	0	0	-1	0	-1	-1	-1
Macrodon ancylodon and atricauda	2	2	2	0	2	-1	2	-1	0
Paralonchurus brasiliensis	0	-1	0	-1	0	0	0	0	-1

 Table 5. Comparison of the statistical evaluation for each of the four genus/species reviewed.

Note: -1 = average concentration lower in Fishing Ban Area compared to at least 1 reference area or commercial markets and at least one location with statistically significantly higher than Fishing Ban Area. 0 = average concentration lower in fishing ban compared to at least 1 reference area or commercial markets and no locations statistically significantly higher than fishing ban area. 1 = average concentration higher in fishing ban compared to at least 1 reference area or commercial markets and no locations statistically significantly lower than fishing ban area. 2 = average concentration higher in fishing ban compared to at least 1 reference area or commercial markets and no locations statistically significantly lower than fishing ban area. 2 = average concentration higher in fishing ban compared to at least 1 reference area or commercial markets and no locations statistically significantly lower than fishing ban area. 2 = average concentration higher in fishing ban area.

with the Event. Such findings align with a general scientific consensus of exceedances of metals in seafood throughout Brazil. For example, Morgano et al. (2011) [21] detected high concentrations of arsenic, chromium, and lead in several species of fish being sold in São Paulo. The same authors also found exceedances of regulatory limits for arsenic, cadmium, mercury, and lead in seafood restaurants. Lima et al. (2015) [22] found exceedances of cadmium, chromium, and mercury in a river in the north of Brazil and directly linked these high concentrations to artisanal or irregular mining. Porto & Ethur (2009) [23] found exceeding levels of aluminum, cadmium, manganese, and nickel in fish in southern Brazil. Santos (2014) [24] describes increased levels of cadmium and lead in fish along the Bahia coast. More recently, Trevisani (2019) [25] found exceedances of arsenic, chromium, and selenium for fish in three estuaries in southeast Brazil. Exceeding levels of arsenic, mercury, and chromium in commercialized fish in Brazil led Oliveira (2020) to recommend monitoring programs in Brazil to prevent the ingestion of fish meat with high levels of toxic metals [26]. Da Silva *et al.* (2021) [27] detected high levels of arsenic in a majority of marine fish species marketed in Bahia (northeastern Brazil), which indicated a potential risk of consumption of these species.

The results reveal that metals detected in the seafood tissue collected in the Fishing Ban Area at the mouth of Rio Doce are generally not significantly different than other reference areas and/or commercially available seafood markets. Having higher concentrations of metals in fish in reference areas outside of the Fishing Ban Area as well as in commercial seafood markets demonstrates that the risk management approach of a localized fishing ban is ineffective for reducing risk to the population related to seafood consumption. A comprehensive risk management approach such as placing limits on seafood consumption rates of certain species for sensitive populations (women of childbearing age, pregnant and lactating women, and children) can be more effective. Such consumption limits are already recommended by Anvisa [2].

Risk management measures should also recognize the health benefits of seafood consumption, which means no consumption at all can also represent a nutrition impairment. This is corroborated by the European Food Safety Authority (EFSA) [28] which points out that, if measures are considered to reduce exposure, the beneficial effects of fish consumption should also be considered. Similar approaches that consider both the benefits and risks of seafood consumption have also been implemented by the Ministry of Health, Labour and Welfare of Japan (MHLW) [29] and the Department of Health and Social Services of the state of Alaska in the USA (DHSS-Alaska) [30] [31].

4. Conclusions

Comparative analyses were performed between metal concentrations in marine fish and crustaceans collected in the fishing ban area, in reference areas unimpacted by the event, and commercialized seafood markets along the Brazilian coast. These analyses show that, for each metal evaluated, at least one or more reference area or seafood from commercially available markets had statistically significant higher concentrations or were statistically indistinguishable from fish and crustaceans collected in the fishing ban area.

Detailed analyses performed on *Conodon nobilis, Genidens barbus/genidens, Macrodon ancylodon/atricauda*, and *Paralonchurus brasiliensis* confirmed these results for all cases except for aluminum, arsenic, barium, iron, and manganese in *Macrodon* samples. For these metals, *Macrodon* samples showed statistically significantly higher concentrations in the Fishing Ban Area. These metals, however, according to Anvisa, did not pose a human health risk related to the consumption of fish collected from the Rio Doce basin and adjacent marine area affected by the Fundão Dam event.

Therefore, the analyses show that the risk management approach of a localized fishing ban is ineffective in reducing risk to the population related to seafood consumption. A more effective approach is to place limits on seafood consumption rates of certain species for sensitive populations, such as women of childbearing age, pregnant and lactating women, and children. A comprehensive risk management approach should also recognize the health benefits of seafood consumption, which means low or no consumption could impair nutrition.

Acknowledgements

The authors wish to acknowledge the following individuals for their contribution to this article: Pedro Henrique Guerra Alves of NewFields Brasil and Shahrokh Rouhani of NewFields Atlanta. The development of this article was supported by BHP Billiton Brasil, NewFields Atlanta LLC, and NewFields Brasil Consultoria Ambiental LTDA ("NewFields Brasil").

Conflicts of Interest

The authors declare no conflicts of interest.

References

- ENGE Corps (2023) Atualização do Plano Integrado de Recursos Hídricos da Bacia do Rio Doce.
 <u>https://www.cbhdoce.org.br/wp-content/uploads/2023/10/1454-ANA-07-RH-RT-0</u> 001-R4 Doce.pdf
- [2] Agência Nacional de Vigilância Sanitária (2019) Avaliação de Risco: Consumo de pescado proveniente de regiões afetadas pelo rompimento da Barragem do Fundão/MG. Nota Técnica No 8/2019/SEI/GEARE/GGALI/DIRE2/ANVISA, 2019.
- [3] Wild, M., Rouhani, S., Oehrig, J., Alves, P.H.G., Odle, W. and Gaspar, D.F.A. (2023) Using Spatiotemporal Ratio Analyses to Quantitatively Estimate Water Quality Recovery of the Rio Doce. *Integrated Environmental Assessment and Management*. <u>https://doi.org/10.1002/ieam.4813</u>
- [4] Oehrig, J., Kananizadeh, N., Wild, M., Rouhani, S. and Odle, W. (2023) Applying Multivariate Techniques to Fingerprint Water Quality Impact of the Fundão Dam Breach within the Rio Doce Basin. *Integrated Environmental Assessment and Management*. <u>https://doi.org/10.1002/ieam.4820</u>
- [5] Kananizadeh, N., Wild, M., Oehrig, J., Odle, W. and Rouhani, S. (2023) Determining Recovery of Marine Water Quality of the Rio Doce Using Statistical and Temporal Comparisons with Nearby River Systems. *Integrated Environmental Assessment and Management*. <u>https://doi.org/10.1002/ieam.4818</u>
- [6] Agência Nacional de Vigilância Sanitária (2022) Instrução Normativa—IN No 160.
- [7] United States Environmental Protection Agency (1992) Guidance for Data Useability in Risk Assessment (Part A).
- [8] Associação Brasileira de Normas Técnicas (2013) Avaliação de risco a saúde humana para fins de gerenciamento de áreas contaminadas.
- [9] Instituto Brasileiro de Geografia e Estatística (IBGE) (2011) Pesquisa de Orçamentos Familiares 2008-2009—Análise do Consumo Alimentar Pessoal no Brasil.
- [10] Coppe (2020) P5—Relatório de Processos Sedimentológicos Conexos ao Rio Doce e aos Rios Adjacentes—Etapa 3—Sobre Sedimentos Depositados na Zona Costeira Adjacente à Foz do Rio Doce, após a Ruptura da Barragem da Samarco em 05/11/2015. Rio de Janeiro.
- [11] Hydrobiology (2021) Documentação do Controle de Qualidade dos Levantamentos Costeiros e Marinhos. <u>https://osf.io/tr4w8</u>
- [12] Beltran-Pedreros, S., Zuanon, J., Leite, R.G., Peleja, J.R.P., Mendonça, A.B. and Forsberg, B.R. (2011) Mercury Bioaccumulation in Fish of Commercial Importance from Different Trophic Categories in an Amazon Floodplain Lake. *Neotropical Ichthyology*, 9, 901-908. <u>https://doi.org/10.1590/S1679-62252011000400022</u>
- [13] Carvalheira, R.G., et al. (2011) Avaliação preliminar da biomagnificação de mercúrio utilizando cinco espécies de peixes na Baía de Guanabara—RJ. <u>https://www.cetem.gov.br/antigo/images/congressos/2011/CAC00560011.pdf</u>
- Kasper, D., Botaro, D., Palermo, E.F.A. and Malm, O. (2007) Mercúrio em peixes—Fontes e contaminação. *Oecologia Brasiliensis*, 11, 228-239. <u>https://doi.org/10.4257/oeco.2007.1102.07</u>
- Kehrig, H.A., Fernandes, K.W.G., Malm, O., Seixas, T.G., Di Beneditto, A.P.M. and de Souza, C.M.M. (2009) Transferência trófica de mercúrio e selênio na costa norte do Rio de Janeiro. *Química Nova*, **32**, 1822-1828.
 https://doi.org/10.1590/S0100-40422009000700026

- [16] Rodrigues, A.P.C., Carvalheira, R.G., Cesar, R.G., Bidone, E.D., Castilhos, Z.C. and Almosny, N.R.P. (2010) Bioacumulação de Mercúrio em Quatro Espécies de Peixes Tropicais Oriundos de Ecossistemas Estuarinos do Estado do Rio de Janeiro, Brasil. *Anuário do Inst. Geociências*, **33**, 54-62. <u>https://doi.org/10.11137/2010_1_54-62</u>
- [17] Suhareva, N., Aigars, J., Poikane, R. and Jansons, M. (2020) Development of Fish Age Normalization Technique for Pollution Assessment of Marine Ecosystem, Based on Concentrations of Mercury, Copper, and Zinc in Dorsal Muscles of Fish. *Environmental Monitoring and Assessment*, **192**, Article No. 279. https://doi.org/10.1007/s10661-020-08261-x
- [18] Moriarty, F. (1988) Ecotoxicology. Human & Experimental Toxicology, 7, 437-441. <u>https://doi.org/10.1177/096032718800700510</u>
- [19] Sonesten, L. (2003) Fish Mercury Levels in Lakes—Adjusting for Hg and Fish-Size Covariation. *Environmental Pollution*, **125**, 255-265. <u>https://doi.org/10.1016/S0269-7491(03)00051-4</u>
- [20] Rocha, K.S., Santos, C.T. and de Freitas, R.R. (2018) Diagnóstico da atividade pesqueira no Espírito Santo, Brasil: Um estudo sobre o segmento de peixarias. *Revista Brasileira de Engenharia de Pesca*, 11, 97-112. https://doi.org/10.18817/repesca.v11i1.1557
- [21] Morgano, M.A., Oliveira, A.P.F., Rabonato, L.C., Milani, R.F., Vasconcellos, J.P. and Martins, C.N. (2011) Avaliação de contaminantes inorgânicos (As, Cd, Cr, Hg e Pb) em espécies de peixes. *Revista do Instituto Adolfo Lutz*, **70**, 497-506.
- [22] Lima, D.P.D., Santos, C., Silva, R.D.S., Yoshioka, E.T.O. and Bezerra, R.M. Contaminação por metais pesados em peixes e água da bacia do rio Cassiporé, Estado do Amapá, Brasil. *Acta Amazonica*, **45**, 405-414. https://doi.org/10.1590/1809-4392201403995
- [23] Porto, L.C.S. and Ethur, E.M. (2009) Elementos traço na água e em vísceras de peixes da Bacia Hidrográfica Butuí-Icamaquã, Rio Grande do Sul, Brasil. *Ciência Rural*, **39**, 2512-2518. <u>https://doi.org/10.1590/S0103-84782009005000213</u>
- [24] Santos, L.F.P. (2011) Avaliação dos Teores de Cádmio e Chumbo em Pescado Proveniente de São Francisco do Conde, Bahia. Master's Thesis, Universidade Federal da Bahia, Salvador. <u>https://repositorio.ufba.br/bitstream/ri/11160/1/Disserta%C3%A7%C3%A3o_Nut_%20Lu%C3%ADs%20Santos.pdf</u>
- [25] Trevizani, T.H. (2018) Bioacumulação e biomagnificação de metais pesados em teias tróficas de estuários do sul-sudeste do Brasil. Ph.D. Thesis, Universidade de São Paulo, São Paulo. https://teses.usp.br/teses/disponiveis/21/21137/tde-01022019-141450/pt-br.php
- [26] de Oliveira, J.G. and Poletto, M. (2020) Drying Characteristics and Heavy Metal Levels in Three Marine Fishes Commercialized in Brazil [Características de secagem e teor de metais pesados em três espécies de peixes marinhos comercializados no Brasil]. *Brazilian Journal of Development*, 6, 76897-76912. https://doi.org/10.34117/bjdv6n10-206
- [27] da Silva, C.A., Garcia, C.A., de Santana, H.L., de Pontes, G.C., Wasserman, J.C. and da Costa, S.S. (2021) Metal and Metalloid Concentrations in Marine Fish Marketed in Salvador, BA, Northeastern Brazil, and Associated Human Health Risks. *Regional Studies in Marine Science*, **43**, Article ID: 101716. https://doi.org/10.1016/j.rsma.2021.101716
- [28] EFSA European Food Safety Authority (2012) Scientific Opinion on the Risk for Public Health Related to the Presence of Mercury and Methylmercury in Food. Ita-

ly.

- [29] MHLW Ministry of Health Labour and Welfare of Japan (2005) Advice for Pregnant Women on Fish Consumption and Mercury.
- [30] DHSS-Alasca Department of Health and Social Services State of Alaska (2014) Fish Consumption Advice for Alaskans: A Risk Management Strategy to Optimize the Public's Health.
- [31] DHSS-Alasca Department of Health and Social Services State of Alaska (2022) Eating Fish Safely, Guidelines For Alaska Women and Children.

Supplements

Identification of Samples and	Replica Sample Results by Laboratory (mg/kg of wet weight)					
Respective Replica Samples	Hidroquímica/Oceanus	Merieux Nutrisciences	Tommasi			
PT6 Sample 312, results of	Mercury in shrimp sample "camarã	o sete barbas" (Xiphopenaeus kroy	veri(a))			
Replica 1	0.06					
Replica 2	0.06					
Replica 3		<0.05				
Replica 4			68.68			
PT6 Sample 317, results of	Mercury in shrimp sample "camarã	o sete barbas" (Xiphopenaeus kroy	veri(a))			
Replica 1	0.04					
Replica 2	0.04					
Replica 3		<0.05				
Replica 4			88.42			
PT8 Sample 474, 1	results of Lead in crab sample "caran	guejo uça" (Ucides cordatus(b))				
Replica 1	19.16					
Replica 2	19.02					
Replica 3		0.07				
Replica 4			0.02			
PT1 Sample 32,	results of Nickel in crab sample "sir	i azul" (Callinectes sapidus(c))				
Replica 1	0.07					
Replica 2	0.07					
Replica 3		<0.05				
Replica 4			235.81			

Table S1. Mercury, lead, and nickel results in which outliers were identified.

Table S2. Fish species summary by sample counts for each location.

	Sample Classification			Location						
Genus	Species	Common Name	Piscivore (Y/N)	Within Fishing Ban Area	Commercially Available- Markets	Abrolhos- Nature Reserve	Jequitinhonha River Mouth- Anthropogenic Reference Area	Total		
Anchovia	Anchovia clupeoides	Anchovy	Ν	15		13		28		
Anchoviella	Anchoviella lepidentostole	Anchovy	Ν	20				20		
Anisotremus	Anisotremus surinamensis	Dogfish	Ν		5			5		

Continued								
Aspistor	Aspistor luniscutis	Catfish	N		9			9
Dagua	Bagre bagre	Catfish	Y		1			1
Bagre	Bagre marinus	Catfish	Y				6	6
Balistes	Balistes capriscus	Triggerfish	Ν		41	6		47
	Balistes sp.	Triggerfish	Ν			2		2
	Caranx crysos	Mackerel	Y	1	5			6
Caranx	Caranx hippos	Crevalle Jack	Ν		6			6
	Caranx latus	Jack	Y	1				1
	Cathorops agassizii	Catfish	Y		2			2
Cathorops	Cathorops arenatus	Catfish	Ν		3			3
	Cathorops spixii	Catfish	Y	25	1	8	41	75
Centropomus	Centropomus parallelus	Snook	Y	6	16	5		27
	Centropomus undecimalis	Snook	Y	12	16			28
Chaetodipterus	Chaetodipterus faber	Hoe	Ν		22			22
Chloroscombrus	Chloroscombrus chrysurus	Bumper	Ν	5				5
Conodon	Conodon nobilis	Grunt	Y	15	11	10	5	41
<u> </u>	Cynoscion acoupa	Weakfish	Y		16		1	17
Cynoscion	Cynoscion guatucupa	White Mullet	Y		15			15
	Cynoscion jamaicensis	Weakfish	Y	24	3			27
	Cynoscion leiarchus	Weakfish	Y		19	1		20
	Cynoscion steindachneri	Weakfish	Y		7			7
	Cynoscion virescens	Hake	Ν		5			5
Diapterus	Diapterus rhombeus	Perch	N	11	11			22

Continued								
Epinephelus	Epinephelus marginatus	Garoupa/ Yellowbelly Grouper	N			1		1
Eugerres	Eugerres brasilianus	Perch	N		5			5
Conidons	Genidens barbus	Catfish	Y	21	27		17	65
Genidens	Genidens genidens	Catfish	Y	84	5	6	19	114
Haamulan	Haemulon aurolineatum	Biquara Tomtate Grunt	Ν		5			5
Haemulon	Haemulon plumierii	White Grunt	Ν		8			8
Harengula	Harengula clupeola	Herring	N	5				5
Isopisthus	Isopisthus parvipinnis	Pescadinha	Y	14	20	4		38
Lagocephalus	Lagocephalus laevigatus	Baiacu/ Smooth Puffer	Y			4		4
Larimus	Larimus breviceps	Drum	Y	40			1	41
Lutianus	Lutjanus cyanopterus	Snapper	Y	9				9
Lutjanus	Lutjanus synagris	Lane snapper	Y			4		4
Magaalan	Macrodon ancylodon	Weakfish	Y	11	66	5		82
Macrodon	Macrodon atricauda	Weakfish	Y	112	30	9		151
Megalops	Megalops atlanticus	Tarpoon	Y	10				10
	Menticirrhus americanus	Kingfish	Ν		9		1	10
Menticirrnus	Menticirrhus littoralis	Croaker	Y	1				1
Micropogonias	Micropogonias furnieri	Croaker	Y		90			90
Mugil	Mugil brasiliensis	Mullet	N	2				2
	Mugil curema	Muller	Ν	7	8			15
Nebris	Nebris microps	Croaker	Y	47	11			58

W. Odle *et al.*

DOI: 10.4236/jep.2023.1410048

Journal of Environmental Protection

Continued								
Notarius	Notarius grandicassis	Catfish	N	5		1		6
Oligoplites	Oligoplites saliens	Leatherjacket	Ν	15	5			20
Ophioscion	Ophioscion punctatissimus	Croaker	Ν	118				118
Pagrus	Pagrus pagrus	Porgy/ Seabream	Y			9		9
Paralichthys	Paralichthys patagonicus	Linguado Flounder	Y	12				12
Paralonchurus	Paralonchurus brasiliensis	Croaker	Ν	80	16	5	6	107
Peprilus	Peprilus paru	Harvestfish	Ν		9	4		13
Doludoctuluo	Polydactylus brasiliensis	Thredfin	Ν				5	5
Polydactylus	Polydactylus oligodon	Perch	Y				11	11
Pomadasys	Pomadasys crocro	Corcorocal Grunt	Y	2				2
Prepilus	Prepilus paru	Gordinho/ Butterfish	Y	8		2		10
Priacanthus	Priacanthus arenatus	Vermelho	Y	3				3
Pseudupeneus	Pseudupeneus maculatus	Perch	N		6			6
Rhomboplites	Rhomboplites aurorubens	Vermilion snapper	Ν		7			7
Scomberomorus	Scomberomorus brasiliensis	Mackerel	Y		25	6		31
Stellifer	Stellifer brasiliensis	Croaker	N	60			56	116
	Stellifer naso	Stardrum	Ν	25			13	38
	Stellifer rastrifer	Stardrum	Ν	116			16	132
	Stellifer stellifer	Croaker	Ν	78			61	139
Syacium	Syacium papillosum	Linguado/ Flounder	Y			6		6
Symphurus	Symphurus trewavasae	Tongue Fish	Ν				3	3
Thalassoma	Thalassoma pavo	Wrasse	Ν			10		10
111111111111111111111111111111111111111	Thalassoma sp.	Wrasse	Ν			10		10
Trachinotus	Trachinotus falcatus	Sernambiguara	Ν	1				1

Continued								
Trinectes	Trinectes paulistanus	Sole	N			4	10	14
Zapteryx	Zapteryx brevirostris	Guitarfish	Ν	1				1
				1022	566	135	272	1995

Table S3. Crustacean species summary by sample counts in each location.

Sam	ple Classification		Location				
Genus	Species	Common Name	Within Fishing Ban Area	Commercially Available — Markets	Abrolhos — Nature Reserve	Jequitinhonha River Mouth— Anthropogenic Reference Area	Total
Callinactor	Callinectes danae	Crab			5		5
Canniectes	Callinectes sapidus	Crab	8				8
Cardisoma	Cardisoma guanhumi	Crab		12			12
	Farfantepenaeus brasiliensis	Shrimp		38		15	53
Farfantepenaeus	Farfantepenaeus paulensis	Shrimp	5	25			30
	Farfantepenaeus sp.	Shrimp		45			45
Litopenaeus	Litopenaeus schmitti	Shrimp		28	14		42
Panulirus	Panulirus laevicauda	Lagosta		17			17
Ucides	Ucides cordatus	Crab	17	12	5		34
Xiphopenaeus	Xiphopenaeus kroyeri	Shrimp	447	94	2	192	735
	Grand Total		477	271	26	207	981

Table S4. Comparison of wet and dry season results for fish and crustaceans.

Class	Analyte	Statistic	p-Value	Significant	Direction
Crustacean	Aluminum	28757.5	6.26E-21	Significant	Wet Season Higher
Crustacean	Arsenic	146,072	5.34E-10	Significant	Dry Season Higher
Crustacean	Barium	39,148	3.54E-07	Significant	Wet Season Higher
Crustacean	Cadmium	93225.5	8.75E-01	Not significant	
Crustacean	Iron	86,428	6.70E-05	Significant	Wet Season Higher
Crustacean	Lead	85583.5	5.37E-02	Not significant	
Crustacean	Manganese	56,796	1.82E-08	Significant	Wet Season Higher
Crustacean	Mercury	61521.5	2.54E-23	Significant	Wet Season Higher
Crustacean	Nickel	63226.5	4.74E-01	Not significant	
Fish	Aluminum	120414.5	1.69E-37	Significant	Wet Season Higher
Fish	Arsenic	440,592	4.19E-05	Significant	Wet Season Higher

Continued					
Fish	Barium	128250.5	1.12E-31	Significant	Wet Season Higher
Fish	Cadmium	394,587	7.90E-10	Significant	Wet Season Higher
Fish	Iron	356,565	3.48E-01	Not significant	
Fish	Lead	396407.5	2.16E-09	Significant	Wet Season Higher
Fish	Manganese	347,405	6.74E-02	Not significant	
Fish	Mercury	233,787	3.97E-65	Significant	Wet Season Higher
Fish	Nickel	309,031	4.14E-01	Not significant	

Table S5. Games/Howell post hoc results, fish species specific metal concentrations, normalized by length.

Analyte	Genus	Group 1	Group 2	p-Value	Significant	Direction
Arsenic	Conodon	Fishing Ban Area	Abrolhos	3.37E-04	Significant	Abrolhos > Fishing Ban Area
Arsenic	Conodon	Fishing Ban Area	Market-Regional	1.45E-04	Significant	Market-Regional > Fishing Ban Area
Arsenic	Conodon	Abrolhos	Market-Regional	1.00E-03	Significant	Abrolhos > Market-Regional
Cadmium	Conodon	Fishing Ban Area	Abrolhos	4.00E-03	Significant	Fishing Ban Area > Abrolhos
Cadmium	Conodon	Fishing Ban Area	Market-Regional	8.65E-01	Not Significant	
Cadmium	Conodon	Abrolhos	Market-Regional	3.29E-01	Not Significant	
Iron	Conodon	Fishing Ban Area	Abrolhos	3.93E-01	Not Significant	
Iron	Conodon	Fishing Ban Area	Market-Regional	9.34E-01	Not Significant	
Iron	Conodon	Abrolhos	Market-Regional	7.60E-02	Not Significant	
Lead	Conodon	Fishing Ban Area	Abrolhos	8.89E-01	Not Significant	
Lead	Conodon	Fishing Ban Area	Market-Regional	1.33E-01	Not Significant	
Lead	Conodon	Abrolhos	Market-Regional	1.52E-01	Not Significant	
Manganese	Conodon	Fishing Ban Area	Abrolhos	7.00E-03	Significant	Fishing Ban Area > Abrolhos
Manganese	Conodon	Fishing Ban Area	Market-Regional	6.18E-01	Not Significant	
Manganese	Conodon	Abrolhos	Market-Regional	9.50E-02	Not Significant	
Mercury	Conodon	Fishing Ban Area	Abrolhos	1.60E-04	Significant	Abrolhos > Fishing Ban Area
Mercury	Conodon	Fishing Ban Area	Market-Regional	2.00E-03	Significant	Market-Regional > Fishing Ban Area
Mercury	Conodon	Abrolhos	Market-Regional	9.34E-01	Not Significant	
Nickel	Conodon	Abrolhos	Market-Regional	2.50E-02	Significant	Market-Regional > Abrolhos

DOI: 10.4236/jep.2023.1410048

Journal of Environmental Protection

Continued						
Aluminum	Genidens	Fishing Ban Area	Abrolhos	3.00E-03	Significant	Fishing Ban Area > Abrolhos
Aluminum	Genidens	Fishing Ban Area	Jequi	4.50E-02	Significant	Jequi > Fishing Ban Area
Aluminum	Genidens	Abrolhos	Jequi	3.20E-02	Significant	Jequi > Abrolhos
Arsenic	Genidens	Fishing Ban Area	Abrolhos	1.32E-06	Significant	Fishing Ban Area > Abrolhos
Arsenic	Genidens	Fishing Ban Area	Jequi	3.55E-08	Significant	Jequi > Fishing Ban Area
Arsenic	Genidens	Fishing Ban Area	Market-Regional	3.34E-01	Not Significant	
Arsenic	Genidens	Abrolhos	Jequi	3.10E-10	Significant	Jequi > Abrolhos
Arsenic	Genidens	Abrolhos	Market-Regional	2.99E-05	Significant	Market-Regional > Abrolhos
Arsenic	Genidens	Jequi	Market-Regional	6.73E-07	Significant	Jequi > Market-Regional
Barium	Genidens	Fishing Ban Area	Abrolhos	3.21E-04	Significant	Fishing Ban Area > Abrolhos
Barium	Genidens	Fishing Ban Area	Jequi	9.03E-01	Not Significant	
Barium	Genidens	Abrolhos	Jequi	1.50E-02	Significant	Jequi > Abrolhos
Cadmium	Genidens	Fishing Ban Area	Abrolhos	9.11E-09	Significant	Fishing Ban Area > Abrolhos
Cadmium	Genidens	Fishing Ban Area	Jequi	3.83E-06	Significant	Fishing Ban Area > Jequi
Cadmium	Genidens	Fishing Ban Area	Market-Regional	9.66E-01	Not Significant	
Cadmium	Genidens	Abrolhos	Jequi	4.40E-02	Significant	Jequi > Abrolhos
Cadmium	Genidens	Abrolhos	Market-Regional	1.49E-01	Not Significant	
Cadmium	Genidens	Jequi	Market-Regional	2.77E-01	Not Significant	
Iron	Genidens	Fishing Ban Area	Abrolhos	4.93E-04	Significant	Fishing Ban Area > Abrolhos
Iron	Genidens	Fishing Ban Area	Jequi	1.10E-02	Significant	Jequi > Fishing Ban Area
Iron	Genidens	Fishing Ban Area	Market-Regional	1.00E+00	Not Significant	
Iron	Genidens	Abrolhos	Jequi	5.00E-03	Significant	Jequi > Abrolhos
Iron	Genidens	Abrolhos	Market-Regional	3.94E-01	Not Significant	
Iron	Genidens	Jequi	Market-Regional	1.30E-02	Significant	Jequi > Market-Regional
Lead	Genidens	Fishing Ban Area	Abrolhos	5.93E-05	Significant	Fishing Ban Area > Abrolhos
Lead	Genidens	Fishing Ban Area	Jequi	9.91E-01	Not Significant	
Lead	Genidens	Fishing Ban Area	Market-Regional	7.49E-01	Not Significant	
Lead	Genidens	Abrolhos	Jequi	5.00E-03	Significant	Jequi > Abrolhos
Lead	Genidens	Abrolhos	Market-Regional	1.20E-04	Significant	Market-Regional > Abrolhos

Continued						
Lead	Genidens	Jequi	Market-Regional	7.42E-01	Not Significant	
Manganese	Genidens	Fishing Ban Area	Abrolhos	1.30E-01	Not Significant	
Manganese	Genidens	Fishing Ban Area	Jequi	8.00E-03	Significant	Jequi > Fishing Ban Area
Manganese	Genidens	Fishing Ban Area	Market-Regional	4.31E-01	Not Significant	
Manganese	Genidens	Abrolhos	Jequi	5.00E-03	Significant	Jequi > Abrolhos
Manganese	Genidens	Abrolhos	Market-Regional	7.01E-01	Not Significant	
Manganese	Genidens	Jequi	Market-Regional	5.00E-03	Significant	Jequi > Market-Regional
Mercury	Genidens	Fishing Ban Area	Abrolhos	6.87E-01	Not Significant	
Mercury	Genidens	Fishing Ban Area	Jequi	9.28E-01	Not Significant	
Mercury	Genidens	Fishing Ban Area	Market-Regional	4.85E-04	Significant	Market-Regional > Fishing Ban Area
Mercury	Genidens	Abrolhos	Jequi	8.15E-01	Not Significant	
Mercury	Genidens	Abrolhos	Market-Regional	4.59E-01	Not Significant	
Mercury	Genidens	Jequi	Market-Regional	1.96E-04	Significant	Market-Regional > Jequi
Nickel	Genidens	Fishing Ban Area	Abrolhos	5.25E-07	Significant	Fishing Ban Area > Abrolhos
Nickel	Genidens	Fishing Ban Area	Jequi	9.13E-05	Significant	Jequi > Fishing Ban Area
Nickel	Genidens	Fishing Ban Area	Market-Regional	4.33E-01	Not Significant	
Nickel	Genidens	Abrolhos	Jequi	1.78E-05	Significant	Jequi > Abrolhos
Nickel	Genidens	Abrolhos	Market-Regional	9.98E-01	Not Significant	
Nickel	Genidens	Jequi	Market-Regional	2.73E-05	Significant	Jequi > Market-Regional
Aluminum	Macrodon	Fishing Ban Area	Abrolhos	3.05E-07	Significant	Fishing Ban Area > Abrolhos
Arsenic	Macrodon	Fishing Ban Area	Abrolhos	5.43E-09	Significant	Fishing Ban Area > Abrolhos
Arsenic	Macrodon	Fishing Ban Area	Market-Regional	1.81E-07	Significant	Fishing Ban Area > Market-Regional
Arsenic	Macrodon	Abrolhos	Market-Regional	2.00E-03	Significant	Market-Regional > Abrolhos
Barium	Macrodon	Fishing Ban Area	Abrolhos	1.00E-03	Significant	Fishing Ban Area > Abrolhos
Cadmium	Macrodon	Fishing Ban Area	Abrolhos	5.00E-03	Significant	Fishing Ban Area > Abrolhos
Cadmium	Macrodon	Fishing Ban Area	Market-Regional	6.43E-01	Not Significant	
Cadmium	Macrodon	Abrolhos	Market-Regional	7.20E-02	Not Significant	
Iron	Macrodon	Fishing Ban Area	Abrolhos	2.92E-10	Significant	Fishing Ban Area > Abrolhos

Continued						
Iron	Macrodon	Fishing Ban Area	Market-Regional	1.10E-02	Significant	Fishing Ban Area > Market-Regional
Iron	Macrodon	Abrolhos	Market-Regional	7.00E-02	Not Significant	
Lead	Macrodon	Fishing Ban Area	Abrolhos	2.70E-02	Significant	Fishing Ban Area > Abrolhos
Lead	Macrodon	Fishing Ban Area	Market-Regional	1.21E-05	Significant	Market-Regional > Fishing Ban Area
Lead	Macrodon	Abrolhos	Market-Regional	1.63E-06	Significant	Market-Regional > Abrolhos
Manganese	Macrodon	Fishing Ban Area	Abrolhos	1.67E-06	Significant	Fishing Ban Area > Abrolhos
Manganese	Macrodon	Fishing Ban Area	Market-Regional	1.91E-06	Significant	Fishing Ban Area > Market-Regional
Manganese	Macrodon	Abrolhos	Market-Regional	9.25E-01	Not Significant	
Mercury	Macrodon	Fishing Ban Area	Abrolhos	3.96E-01	Not Significant	
Mercury	Macrodon	Fishing Ban Area	Market-Regional	9.45E-05	Significant	Market-Regional > Fishing Ban Area
Mercury	Macrodon	Abrolhos	Market-Regional	4.40E-02	Significant	Market-Regional > Abrolhos
Nickel	Macrodon	Fishing Ban Area	Abrolhos	3.90E-09	Significant	Fishing Ban Area > Abrolhos
Nickel	Macrodon	Fishing Ban Area	Market-Regional	7.04E-01	Not Significant	
Nickel	Macrodon	Abrolhos	Market-Regional	1.90E-02	Significant	Market-Regional > Abrolhos
Aluminum	Paralonchurus	Fishing Ban Area	Jequi	2.37E-01	Not Significant	
Arsenic	Paralonchurus	Fishing Ban Area	Abrolhos	1.20E-02	Significant	Fishing Ban Area > Abrolhos
Arsenic	Paralonchurus	Fishing Ban Area	Jequi	4.40E-02	Significant	Jequi > Fishing Ban Area
Arsenic	Paralonchurus	Fishing Ban Area	Market-Regional	7.31E-05	Significant	Market-Regional > Fishing Ban Area
Arsenic	Paralonchurus	Abrolhos	Jequi	3.90E-02	Significant	Jequi > Abrolhos
Arsenic	Paralonchurus	Abrolhos	Market-Regional	2.44E-06	Significant	Market-Regional > Abrolhos
Arsenic	Paralonchurus	Jequi	Market-Regional	6.40E-02	Not Significant	
Barium	Paralonchurus	Fishing Ban Area	Jequi	2.83E-01	Not Significant	
Cadmium	Paralonchurus	Fishing Ban Area	Abrolhos	2.35E-01	Not Significant	
Cadmium	Paralonchurus	Fishing Ban Area	Jequi	1.17E-01	Not Significant	
Cadmium	Paralonchurus	Fishing Ban Area	Market-Regional	2.41E-11	Significant	Market-Regional > Fishing Ban Area

DOI: 10.4236/jep.2023.1410048

Continued						
Cadmium	Paralonchurus	Abrolhos	Jequi	5.22E-01	Not Significant	
Cadmium	Paralonchurus	Abrolhos	Market-Regional	1.10E-09	Significant	Market-Regional > Abrolhos
Cadmium	Paralonchurus	Jequi	Market-Regional	5.87E-07	Significant	Market-Regional > Jequi
Iron	Paralonchurus	Fishing Ban Area	Abrolhos	9.40E-02	Not Significant	
Iron	Paralonchurus	Fishing Ban Area	Jequi	5.44E-01	Not Significant	
Iron	Paralonchurus	Fishing Ban Area	Market-Regional	7.85E-01	Not Significant	
Iron	Paralonchurus	Abrolhos	Jequi	5.32E-01	Not Significant	
Iron	Paralonchurus	Abrolhos	Market-Regional	1.24E-01	Not Significant	
Iron	Paralonchurus	Jequi	Market-Regional	5.53E-01	Not Significant	
Lead	Paralonchurus	Fishing Ban Area	Abrolhos	6.74E-01	Not Significant	
Lead	Paralonchurus	Fishing Ban Area	Jequi	1.00E+00	Not Significant	
Lead	Paralonchurus	Fishing Ban Area	Market-Regional	8.93E-01	Not Significant	
Lead	Paralonchurus	Abrolhos	Jequi	5.82E-01	Not Significant	
Lead	Paralonchurus	Abrolhos	Market-Regional	1.68E-08	Significant	Market-Regional > Abrolhos
Lead	Paralonchurus	Jequi	Market-Regional	8.97E-01	Not Significant	
Manganese	Paralonchurus	Fishing Ban Area	Abrolhos	9.95E-01	Not Significant	
Manganese	Paralonchurus	Fishing Ban Area	Jequi	3.00E-01	Not Significant	
Manganese	Paralonchurus	Fishing Ban Area	Market-Regional	2.87E-01	Not Significant	
Manganese	Paralonchurus	Abrolhos	Jequi	3.06E-01	Not Significant	
Manganese	Paralonchurus	Abrolhos	Market-Regional	2.51E-01	Not Significant	
Manganese	Paralonchurus	Jequi	Market-Regional	3.49E-01	Not Significant	
Mercury	Paralonchurus	Fishing Ban Area	Abrolhos	1.33E-01	Not Significant	
Mercury	Paralonchurus	Fishing Ban Area	Jequi	4.67E-01	Not Significant	
Mercury	Paralonchurus	Fishing Ban Area	Market-Regional	2.34E-01	Not Significant	
Mercury	Paralonchurus	Abrolhos	Jequi	2.40E-02	Significant	Abrolhos > Jequi
Mercury	Paralonchurus	Abrolhos	Market-Regional	9.83E-01	Not Significant	
Mercury	Paralonchurus	Jequi	Market-Regional	1.70E-02	Significant	Market-Regional > Jequi
Nickel	Paralonchurus	Fishing Ban Area	Jequi	1.07E-01	Not Significant	
Nickel	Paralonchurus	Fishing Ban Area	Market-Regional	5.42E-06	Significant	Market-Regional > Fishing Ban Area
Nickel	Paralonchurus	Jequi	Market-Regional	3.10E-01	Not Significant	