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Traceability challenges and heavy metal risks in commercial shrimp and prawn

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ABSTRACT

Mislabelling in the global fisheries sector raises concerns about the identity, safety, and sustainability of seafood products. It puts human health at risk when substitute species are contaminated with heavy metals that may cause chronic diseases and cancer. The aim of this work was to analyse mislabelling in shrimps and prawns sold in the Spanish market and possible risks of heavy metal ingestion. Analysis of labels, DNA barcoding for species identification and quantification of heavy metals were performed on 100 market samples, and health risk was calculated from standard indices. More than one half of individuals did not comply with European regulations about labels, principally for the absence of mandatory elements in the label. One third of the analysed shrimps were substitute species (not mentioned on the label), and a 10% did not comply with the legal European limits for heavy metals. The prawns *Penaeus indicus* and *Penaeus latisulcatus* from West Indian and Central/East Atlantic waters exhibited the highest heavy metals concentrations. Indices calculated for these two species, and for *Pandalus borealis* and *Parapenaeus longirostris*, suggest health risks if consumed daily. If those or other species from these polluted areas were employed as substitutes, mislabelling would encompass health risk.

1. Introduction

1.1. Mislabelling and fisheries sustainability

One of the major problems concerning the fisheries sector at global scale is mislabelling, which implies to sale substitute species instead of those indicated on the label (Marín et al., 2013). Mislabelling growth is a cause of public concern about the identity, safety, and sustainability of sea products (Kroetz et al., 2020). Seafood products pass through many intermediaries in the route from fishing ships to consumers, with increasing loss of traceability (Cawthorn et al., 2012); thus, preventing mislabelling is not easy because it can occur in any point of the supply chain (Cawthorn et al., 2012; Marín et al., 2018). The European Parliament and INTERPOL/EUROPOL have identified seafood fraud as one of the main concerns related to the food supply (Paolacci et al., 2021). The high mislabelling levels show a clear need for more regulation and control measures to achieve more transparency and avoid undermining consumer confidence in food safety (Muñoz-Colmenero et al., 2017). A correct labelling is essential not only to know the nutritional value of the product, its ingredients, its price, and its allergenic capacity, but also for the consumers to make a conscious and informed choice of what they are consuming and how to use the food safely for them and the environment (Cawthorn et al., 2018). Nevertheless, despite the legislation and the rise in consumer demand for transparency (Lu et al., 2016; Rodriguez-Salvador & Dopico, 2020; Verbeke, 2001), mislabelling has become a major issue for consumers, especially in fishery products (Galal-Khallaf et al., 2016; Guardone et al., 2017; Luque & Donlan, 2019).

The European Regulation (EU) No 1379/2013 establishes, among others, the obligation to disclose the commercial and scientific name of the product, the geographical area of production or catch, and the processing method or fishing gear (Tinacci, Stratev, et al., 2018). However, seafood labelling in Europe often lacks some mandatory information, with the capture method and the scientific name being most frequently missing from the label (Feldmann et al., 2021; Paolacci et al., 2021), so the consumer is not able to choose a safe and ecologically responsible choice of seafood. According to the annual report of the European Union Food Fraud Network, published on May 18, 2020, 47% of food fraud is the result of omissions in the labelling, or the substitution of one species for another (Rasmussen & Morrissey, 2008; Spencer

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& Bruno, 2019). These substitutions may be due to accidental recording errors occurring, for example, in species caught from mixed fisheries (Blanco-Fernandez et al., 2022; Garcia-Vazquez et al., 2012); or due the lack of information on specific traits for identification (Ardura et al., 2010). Mislabelling can be also due to intentional fraud with an economic incentive, substituting the species registered on the label for a species of lower economic value. Substitution may involve wild and aquaculture species (Muñoz-Colmenero et al., 2017), species of different geographical origin (Garcia-Vazquez et al., 2011), species coming from illegal or unregulated fishing that cannot be legally sold (Blanco-Fernandez, Garcia-Vazquez, & Machado-Schiaffino, 2021; Von der Heyden et al., 2010), and others.

Regardless of the causes, the consequences of mislabelling affect the consumer and the ecosystem. The consumer buys a species that they do not demand, violating their right to decide for reasons of health, religion and/or environmental awareness (Delpiani et al., 2020; Marín et al., 2013, 2018). In the case of the ecosystem, mislabelling impedes proper resource management (Galal-Khallaf et al., 2016; Kroetz et al., 2020; Marko et al., 2011). Inaccurate estimation of the catch rates of substitute species endangers the sustainability of fishing, the depletion and over-exploitation of resources and, in the long term, the local extinction of populations (Agnew et al., 2009).

1.2. Heavy metal risks in seafood consumers

Incorrect labelling may put human health at risk when substitute species, that are inadvertently consumed, are contaminated. Human activities such as agriculture, industry, or mining produce, among other pollutants, cause an increase in the content of heavy metals in the water (Jafarzadeh et al., 2022). Some of these metals, like cadmium, mercury and lead represent a serious environmental problem for their high toxicity and genotoxicity (e.g., Biruk et al., 2017; Faraji et al., 2023; Salem et al., 2014), causing chronic dermatitis, lung fibrosis, cardiovascular and kidney diseases (Ali & Khan, 2018) and even carcinogenic and teratogenic effects (Abd-Elghany et al., 2020). Once they enter the ecosystem, they can remain there for hundreds of years, contaminating the habitat and accumulating in plant and animal tissues. The concentration of some heavy metals in organisms increases along the food chain (Adel et al., 2016, 2018; Ali & Khan, 2018; Ali & Khan, 2019; Mazrouh & Mourad, 2019), and may reach humans through consumption of contaminated specimens. The concentration of heavy metals depends on the species and the capture area (Steinhaunsen et al., 2022). For example, tuna caught from West African waters are more contaminated than those caught in South African and European waters, associated with the respective levels of metal pollution in these regions (Garcia-Vazquez et al., 2021). Following this example, selling West African species as substitutes of South African ones would entail an added risk of mercury intake.

The risk of heavy metal ingestion through fish consumption has been widely studied (Garcia-Vazquez et al., 2021; Gbogbo et al., 2018; Liu et al., 2019; Steinhausen et al., 2022; Traina et al., 2019). Other groups of seafood species are much less investigated. Shrimps and prawns, for example, are widely consumed worldwide accounting for 15% of the main groups of species in the fish trade in 2018 (FAO, 2020). These crustaceans are found all over the globe, from the equator to the polar region. Most marine species occupy shallow to moderately deep waters, commercial shrimps being caught mainly on continental shelves at depths shallower than 100 m. Some are pelagic, but by far the majority are benthic and live on a wide variety of bottoms, Penaeidae and Pandalidae being commercially very important (Fransen, 2014). Prawn aquaculture is also remarkable. *Penaeus vannamei* and *Penaeus monodon* are the most farmed prawn worldwide accounting for 63.5% of global prawn production in 2018 (6,000,000 tonnes), which means that more prawns are farmed than fished (Boyd et al., 2022). The world's leading producers of prawns are China (2,051,921 tonnes in 2018), Indonesia (907,988 tonnes) and India (682,300 tonnes) (Boyd et al., 2022).

Compared to other species, shrimps are highly susceptible to environmental pollution for their way of feeding, picking up from sediments organic matter that may have accumulated heavy metals (Abd-Elghany et al., 2020; Fakhri et al., 2018). However, to date, only a few studies have analysed the risks of heavy metal ingestion through shrimp consumption (e.g., Abd-Elghany et al., 2020; Baki et al., 2018; Kato et al., 2020). The results vary among regions. Although generally shellfish accumulate more metals like arsenic than shrimp (reviewed by Kato et al., 2020), significant concentrations of arsenic were found in brown shrimp *Cragnon* from Belgium, in the Scheldt estuary, with an increasing gradient towards the sea (Van Ael et al., 2017). Abd-Elghany et al. (2020) found significant risk of heavy metal ingestion through shrimp consumption in Egypt. Green tiger shrimp *Penaeus semisulcatus* from Turkey exhibits higher concentrations of heavy metals than fish from the same waters (Kaya & Turkoglu, 2017), and the same happened for the species *Penaeus semisulcatus* with dangerous levels of cadmium and lead (Aytekin et al., 2019). In contrast with these data, Olgunoglu (2015) did not find any dangerous level of heavy metals in other three different shrimp species from Turkish waters (*Plesionika martia, Plesionika edwardsii, Aristeus antennatus*). Likewise, Baki et al. (2018) found similarly acceptable carcinogen ranges in fish and shrimp from Bangladesh, and lower lead concentrations in shrimp than in fish. Not only the environmental pollution of a region influences the level of heavy metals in shrimps, but also there are differences between species as well, due to their different way of life that will determine how they bioaccumulate heavy metals. This has been seen in different species in Bangladesh (Hossain et al., 2022).

On the other hand, mislabelling seems to be important in these crustaceans. For example, Galal-Khallaf et al. (2016) found about 16% of mislabelled shrimp and prawn in Spanish markets. If substitute species bioaccumulate naturally more metals or come from more polluted regions than those specified on the label, mislabelling would encompass an inadvertent risk of heavy metal ingestion.

1.3. Objectives and departure hypothesis

The objectives of this work were two-fold. First, to analyse mislabelling in commercial shrimps and prawns from different regions sold in the Spanish market, where there is a considerable level of mislabelling (Galal-Khallaf et al., 2016); second, to infer if it encompasses additional risk of heavy metal intake, from heavy metal analysis of correctly and incorrectly labelled samples.

The expectation was that the profile of heavy metal concentrations would be different depending on the capture region and the species. DNA barcoding was employed to authenticate the species for comparison with label information, as in many other seafood studies (Adibah et al., 2020; Teletchea et al., 2005; Tinacci, Guidi, et al., 2018).

2. Material and methods

2.1. Sampling and labels analysis

The terms "shrimp" and "prawn" do not refer currently to specific taxonomic groups, rather to the size being "shrimp" generally applied to smaller individuals. In the EU the terms "shrimp" and "prawn" may be applied to the same species depending on the country (https://fish-commercial-names.ec.europa.eu/fish-names/species_es?sn=27761#commdes). In this study will be used the category that appears on the label. A total of 20 packages of frozen shrimps and prawn (cooked or raw; 10 packages of shrimps and 10 of prawns) commercialized in Spain by 13 different brands were randomly obtained from five local supermarkets in Asturias, northern Spain, between November and March 2022. Five individuals were analysed per package, representing a total of 100 samples of different species and genera. The information disclosed in the labels was photographed and digitally recorded. According to the European Commission Directorate-General for Maritime Affairs and Fisheries (2015) and Regulation (EU) No 1379/2013 of the European Parliament and of the Council of December 11, 2013, the following items must be displayed on the label: commercial designation and scientific name, production method (aquaculture or extractive fishing), fishing gear (trawls, bottom longlines, gillnets, hook and line …), catch area, and identification mark (code indicating the name of the country, the approval number of the establishment where production takes place, and the abbreviation CE, which must appear when the product is manufactured in the EU) (European Commission Directorate-General for Maritime Affairs and Fisheries, 2015).

2.2. DNA extraction and barcoding

DNA was extracted from the tail muscle of the shrimps and prawns, far from the digestive tract to avoid contamination from the gut content. DNA extraction and PCR procedures followed Ghalal-Khallaf et al. (2016), based on Chelex® resin (Bio-Rad Laboratories) extraction and PCR amplification of two barcodes for species authentication. The mitochondrial cytochrome *c* oxidase subunit I (COI) gene was amplified by polymerase chain reaction (PCR) using the primers jgLCO1490 (5’ - TIT CIA CIA AYC AYA ARG AYA TTG G – 3′) and jgHCO2198 (5’ - TAI ACY TCI GGR TGI CCR AAR AAY CA – 3′) from Geller et al. (2013). The mitochondrial 16 S rRNA gene was PCR-amplified using the primers 16Sar (5’ - CGC CTG TTT ATC AAA AAC AT – 3′) and 16Sbr (5’ - CCG GTC TGA ACT CAG ATC ACG T – 3’) from Palumbi (1996). The PCR mix for both genes amplification contained 2 μl of 10 μM primers, 2 μl of 2.5 mM dNTPs, 2 μl of 25 mM $MgCl_2$, 4 μl of 5 × Buffer GoTaq® Promega, 0.15 μL of GoTaq® Polymerase (5 u/μl) and 2 μl of DNA, in a final volume of 20 μl. PCRs were run in a thermal cycler from Applied Biosystems, model 2720. For COI gen with an initial denaturation step at 95 °C for 5 min followed by 35 cycles of denaturation at 95 °C for 1 min, annealing at 48 °C for 1 min, elongation at 72 °C for 1 min, and final extension at 72 °C for 5 min. For 16 S rRNA gene with an initial denaturation step at 95 °C for 5 min then 35 cycles of denaturation at 94 °C for 1 min, annealing at 45–55 °C for 1 min, elongation at 72 °C for 2 min, and final extension at 72 °C for 7 min.

PCR products were separated and visualized using 2% agarose gel stained with 10 mg/μL SympleSafe™ (EURx, Gdansk, Poland). Amplicons were sequenced using standard Sanger sequencing method at Macrogen Spain, Inc. (Madrid, Spain). Sequences were manually checked and trimmed using the bioinformatic software BioEdit, and then BLASTed on NCBI (https://blast.ncbi.nlm.nih.gov/Blast.cgi) for species identification. The best match was chosen considering the highest identity (always >97%).

2.3. Heavy metals analysis

A small piece (0.2–0.5 g) of muscle tissue was removed from each sample from the tail (edible part of these species) for the heavy metal analysis, using only plastic materials to manipulate the tissue to avoid external metal contamination. All samples were kept in sealed plastic bags, labelled, and frozen at the laboratory until processing.

Muscle tissue was digested with nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) in a temperature-controlled microwave (Ethos One) heating in closed TFM vessels (Teflon tubes). The concentration of eight heavy metals (Cr, Ni, Cu, Zn, As, Cd, Hg and Pb) were obtained using Inductively Coupled Plasma Mass Spectrometry technology (ICP-MS, Agilent 7700x series spectrometer with autosampler). The measurements were repeated three times per sample and the mean was calculated. Concentrations were obtained in μg kg^{-1} wet weight (w/w), with the corresponding relative standard deviation of less than 10%. Limits of detection (LOD) were 0.01396 μg kg^{-1} for Cr; 0.01132 μg kg^{-1} for Ni; 0.003072 μg kg^{-1} for Cu; 0.08793 μg kg^{-1} for Zn; 0.001794 μg kg^{-1} for As; 0.003026 μg kg^{-1} for Cd; 0.0113 μg kg^{-1} for Hg and 0.001073 μg kg^{-1} for Pb.

To strengthen the validation of the analytical method’s precision, a certified reference sample was employed (European Reference Material ERM® BB422 Fish muscle) (Diop et al., 2016; Gbogbo et al., 2018; Steinhausen et al., 2022). In mg kg^{-1} results were: 1.69 measured (1.67 certified) for Cu, 14.21 measured (16 certified) for Zn, 12.56 measured (12.7 certified) for As, 0.01 measured (0.01 certified) for Cd and 0.6 measured (0.6 certified) for Hg. Therefore, the analysis can be considered reliable.

All this process of analysis of heavy metals was done in the Scientific and Technical Services (SCTs) of the University of Oviedo (Spain).

2.4. Health risk assessment

This assessment was done for general European adult populations and separately, for pregnant women, following Abd-Elghany et al. (2020), Steinhausen et al. (2022) and Traina et al. (2019). Estimates could not be done for children in absence of data about their daily consumption rate of crustaceans in Europe. The potential effect of cooking on contaminants was not considered, as in prior studies (Copat et al., 2012; Miri et al., 2017; Steinhausen et al., 2022). Precautionary approach was adopted, whereby metals are assumed to be in their specific harmful forms (Ackah, 2019; Liu et al., 2019; Steinhausen et al., 2022). These harmful forms are inorganic arsenic, organic mercury (methylmercury) and chromium VI.

The concentration of heavy metals of each sample was compared with the European regulations, in order to check if the samples comply with the limitations (Commission Regulation (EC) No 1881/2006 of December 19, 2006). These regulations indicate the limit of mercury, lead, and cadmium in 0.5 mg kg $^{-1}$ (w/w).

2.4.1. Estimated weekly intake (EWI) of heavy metals

EWI (mg kg^{-1}) was calculated following:

$$EWI = \frac{C_m \times CR}{BW} \times 7$$

Where EWI = weekly exposure to metal *m* through ingestion of contaminated shrimps and prawns (mg kg^{-1} BW per week), C_m = mean metal concentration in shrimp and prawn tissue (mg kg^{-1} w/w), CR = mean daily consumption rate of crustaceans (0.041 kg day^{-1}) (Abd-Elghany et al., 2020), BW = body weight of the individual consumer, considering 70.08 kg as average European adult and 64 kg for pregnant women (Steinhausen et al., 2022; Walpole et al., 2012).

Provisional Tolerable Weekly Intake values (PTWI) (in mg kg^{-1} BW) reported by Garcia-Vazquez et al. (2021) and Steinhausen et al. (2022) were used for comparing with EWI values; if EWI values are higher than PTWI, they are not tolerable. When a PTWI range was specified, the precautionary approach was adopted by selecting the lowest value (Steinhausen et al., 2022).

2.4.2. Target hazard quotient (THQ) and total target hazard quotient (TTHQ)

THQ provides an indication of the possible hazard linked to exposure to a specific pollutant and values above one exceeds the reference dose, thus indicating possible non-carcinogenic effects and the possibility of experiencing a significant health risk from metal ingestion (Steinhausen et al., 2022; Traina et al., 2019). Following earlier studies on risk assessment, the ingestion dose is assumed to be equal to the absorbed contaminant dose (Traina et al., 2019).

THQ was calculated following this equation:

$$THQ = \frac{EF \times ED \times CR \times C_m}{RfDo \times BW \times AT}$$

Where EF = exposure frequency of consumption (365 days $year^{-1}$), ED = exposure duration total (44 years for Europeans adults (https://www.ine.es/prodyser/demografia_UE/bloc-1c.html?lang=en) and 29.5 years for average pregnant women in Europe (https://www.idescat.cat/indic ados/?id=ue&n=10752&lang=es), CR = mean daily consumption rate of crustaceans, C_m = concentration of heavy metal m in the sample (mg kg^{-1} w/w), RfDo = oral reference dose for non-carcinogenic effects (inorganic As = 0.003 mg kg^{-1} day^{-1}, Cd = 0.001 mg kg^{-1} day^{-1}, MethylHg = 0.001 mg kg^{-1} day^{-1}) (Steinhausen et al., 2022), (Cr (VI) = 0.005 mg kg^{-1} day^{-1}) (Copat et al., 2012), (Cu = 0.0371 mg kg^{-1} day^{-1}, Ni = 0.02 mg kg^{-1} day^{-1}, Zn = 0.3 mg kg^{-1} day^{-1} (Steinhausen et al., 2022), (Pb = 0.004 kg^{-1} day^{-1}) (Traina et al., 2019), BW = body weight of the individual consumer, AT = time of exposure (365 days $year^{-1}$ × ED).

The TTHQ or Hazard Index (HI) was calculated as the sum of the individual THQs of each metal, as the exposure is simultaneous and the effects of the contaminants may be combined and the risk therefore higher (Abd-Elghany et al., 2020; Steinhausen et al., 2022; Traina et al., 2019).

2.4.3. Carcinogenic risks (CR_{lim})

CR_{lim} indicates the maximum daily lifetime intake rate that is expected not to cause adverse carcinogenic effects; it is the acceptable limit for health (in kg day^{-1}) (Miri et al., 2017; Steinhausen et al., 2022; Traina et al., 2019). There are certain heavy metal compounds (inorganic arsenic, chromium VI, and inorganic lead compounds), which have the potential to induce cancer, after many years of exposure and certain amounts of consumption (Miri et al., 2017; Steinhausen et al., 2022).

CR_{lim} was calculated following this equation:

$$CR_{lim} = \frac{ARL \times BW}{CSF \times C_m}$$

Where ARL = maximum acceptable lifetime risk level (10^{-5}); CSF = cancer slope factor, for inorganic lead (0.0085 mg kg^{-1} day^{-1}), for Arsenic (1.5 mg kg^{-1} day^{-1}), and for Chromium (VI) (0.5 mg kg^{-1} day^{-1}) (Miri et al., 2017; Steinhausen et al., 2022).

The CR_{lim} was compared to the European average daily intake (CR: 0.041 kg day^{-1}). If this intake exceeds the CR_{lim}, carcinogenic effects could develop in the consumer after a lifetime of exposure.

2.5. Statistics

Comparison between packs for all the heavy metals analysed (contents of the eight heavy metals) was performed using PERMANOVA with 9999 permutations, followed by post-hoc pairwise F test in case of significance. Multivariate rank tests (ANOSIM) were done to test for higher content of all metals in a group versus other/s, also with 9999 permutations.

For comparisons of single metal concentration among sample groups (species or catch regions) the Kruskal-Wallis test was employed, and in cases of significant difference, the post hoc Mann-Whitney test. Only those species with at least n = 5 were considered for the statistical comparison between species. The comparison of the level of mislabelling between products sold as shrimp and prawn was done using contingency Chi-square.

All statistical analyses were performed in R software (https://www.R-project.org/

3. Results

3.1. Labels completeness

Detailed information about the species indicated in the labels, production method, and catch area is shown in Supplementary Table 1.

Table 1

Summary of labels compliance. Label completeness: ScN, PM, FG, CA, IM are scientific name, production method, fishing gear, catch area and identification mark, respectively. Correct ScN should be the species name, not only the genus. Substitutes, % of substitutes identified from DNA barcoding in each pack. Note that the species *Litopenaeus vannamei* is currently named *Penaeus vannamei*. Regarding the code, "L" refers to packages of prawns and "G" to packages of shrimps. The second two-letter part of the code represents the selling point (first letter) and commercial brand (second letter). The presentation (whole or peeled) is also given.

Pack Code	Commercial designation/ presentation	Production method and area on the label	Scientific name on the label	Substitutes	ScN	PM	FG	CA	IM
L1-AA	Prawn/whole	Aquaculture in Nicaragua	*Penaeus vannamei*	**0%**	✓	✓	✓	✓	✓
L2-AD	Prawn/peeled	Aquaculture in Ecuador	*Penaeus* spp. *(Penaeus vannamei ….)*	**0%**	×	✓	✓	✓	✓
L3-AD	Prawn/peeled	No information	*Penaeus* spp. *(Penaeus vannamei ….)*	**0%**	×	×	×	×	✓
L4-CD	Prawn/whole	Aquaculture in Cuba	*Penaeus* spp. *(Penaeus vannamei ….)*	**0%**	×	✓	✓	✓	✓
L5-CE	Prawn/peeled	Trawls in Southwest Atlantic Ocean (FAO n°41)	*Pleoticus muelleri*	**0%**	✓	✓	✓	✓	✓
L6-CC	Wild Prawn/whole	Trawls in Indian Ocean (FAO n° 51)	*Penaeus indicus*	**0%**	✓	✓	✓	✓	✓
L7-CF	Wild ivory prawn/ whole	Trawls in West Indian Ocean (FAO n° 51)	*Penaeus latisulcatus*	**0%**	✓	✓	✓	✓	✓
L8-BG	Austral prawn/whole	Trawls in Southwest Atlantic Ocean (FAO n°41)	*Pleoticus muelleri*	**0%**	✓	✓	✓	✓	×
L9-BH	Prawn/peeled	Trawls in Southwest Atlantic Ocean (FAO n°41)	*Pleoticus muelleri*	**0%**	✓	✓	✓	✓	✓
L10-BG	Prawn/whole	Aquaculture in Panama	*Litopenaeus vannamei*	**0%**	✓	✓	✓	✓	×
G1-AI	Boreal shrimp/peeled	Trawls in Northwest Atlantic Ocean	*Pandalus borealis*	**0%**	✓	✓	✓	✓	✓
G2-AJ	White shrimp/whole	Trawls in East Central Atlantic Ocean (FAO n° 34)	*Parapenaeus longirostris*	**0%**	✓	✓	✓	✓	✓
G3-CD	Shrimp/peeled	Trawls in West Indian Ocean	*Parapenaeopsis* spp., *Solenocera crassicornis; Parapenaeus longirostis; Trachypenaeus* spp.	**0%**	×	✓	✓	✓	✓
G4-CF	Shrimp/peeled	Trawls in Northwest Pacific Ocean (FAO n° 61)	*Trachypenaeus* spp.	**100%**	×	✓	✓	✓	✓
G5-CC	Shrimp/peeled	Trawls in East Indian Ocean	*Solenocera* spp.	**100%**	×	✓	✓	✓	✓
G6-BB	Shrimp/peeled	Trawls in Northwest Pacific Ocean (FAO n° 61)	*Solenocera melantho*	**100%**	✓	✓	✓	✓	×
G7-CC	Shrimp/peeled	Trawls in East Indian Ocean	*Parapenaeopsis* spp.	**100%**	×	✓	✓	✓	✓
G8-BK	Shrimp/peeled	Trawls in Northwest Pacific Ocean (FAO n° 61)	*Solenocera melantho*	**60%**	✓	✓	✓	✓	✓
G9-DL	Shrimp/peeled	Trawls in Northwest Pacific Ocean (FAO n° 61)	*Solenocera melantho*	**100%**	✓	✓	✓	✓	✓
G10-EM	Shrimp/peeled	Trawls in West Indian Ocean (FAO n° 51) and East Indian Ocean (FAO n° 57)	*Parapenaeopsis* spp., *Parapenaeus* spp.	**100%**	×	✓	✓	✓	✓

According to the labels (Table 1), some products were from aquaculture in Central America and others from extractive fisheries in different marine regions (Fig. 1). The FAO and catch areas displayed on the labels were checked for concordance with the natural distribution of the species or genus declared. No inconsistencies were found (Supplementary Table 1).

Labels were analysed to determine their compliance with European Commission Directorate-General for Maritime Affairs and Fisheries (2015) and Regulation (EU) No 1379/2013 of the European Parliament and of the Council of December 11, 2013. Only 45% of the packs analysed exhibited complete labels (Table 1). Eight out of 20 (40%) indicated only the genus on the label, while it is mandatory to display the complete scientific name of all the species of the package according to EU regulations. The label of one of those packs (L3) was very incomplete, displaying only the genus name (in parentheses a species) and the identification mark, while another, G3, displayed a mixture of species and genera in the label. Three labels (15%) did not show the identification mark.

3.2. *Species substitution detected from DNA*

Most of the individuals were correctly identified with COI gene, except for three cases (L92, L94 and G52 individuals, Supplementary Table 1) where 16 S gene was used. The haplotypes of the sequences obtained in this study were deposited in GenBank, under the accession numbers OQ980339-OQ980371, OR004713, OR004825, OR005425, OR016042, OR018123, OR018124, OR018126, OR018136, OR018138, OR018139, OR018140 and OR018141 for COI and OQ979140, OQ979141 and OQ980394 for 16S rRNA gene.

In total 20 species from three families (Pandalidae, Penaeidae and Solenoceridae) were found from DNA (Table 2). The species identified from DNA barcoding was compared with the information displayed on the label. When labels indicated only the genus, if the species identified from the barcode coincided with the stated genus, it was not considered as species substitution. No substitution was found in products marketed as prawns, either from aquaculture or from extractive fisheries (Table 1). In contrast, species substitution occurred in 66% of the analysed individuals marketed as shrimp, all of them peeled, as the genetic barcodes did not match the species indicated on the label. This happened in 70% of the shrimp packages (Table 1). Moreover, six of the 10 shrimp packages analysed contained a mixture of substitute species: G4, G5, G6, G7, G9 and G10 (Supplementary Table 1). The difference in the proportion of individuals with species substitution between products sold as shrimp (66%) and those sold as prawn (0%, excluding L3 where the species was not disclosed on the label) was statistically highly significant (contingency $\chi^2 = 29.46$, 1 d. f., $p < 0.001$). From this study, labelling of imported shrimp was highly inaccurate and also imprecise, since four of the seven packages with species substitution exhibited only genus or genera on the label (Table 1), and even the genus was wrong.

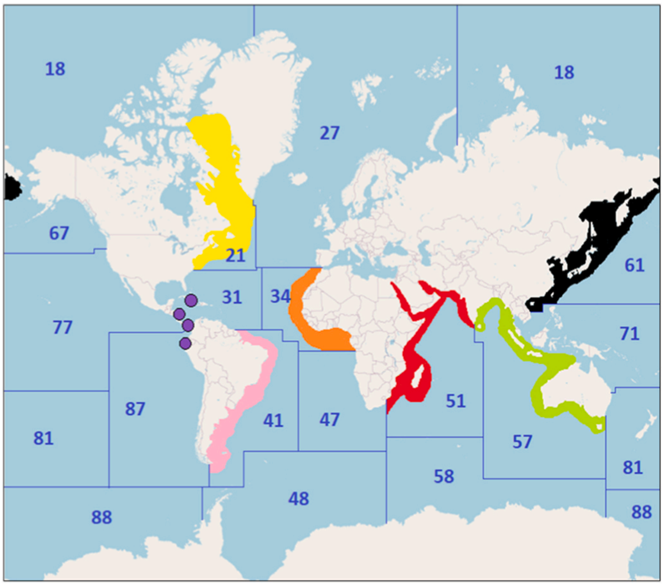

Fig. 1. Map showing the production regions retrieved from the information reported on the labels for all the products analysed in this study. Origin of products are shown as follows: yellow, Northwest Atlantic Ocean; pink, Southwest Atlantic Ocean; orange, Central/East Atlantic Ocean; black, Northwest Pacific Ocean; red, West Indian Ocean; green, East Indian Ocean. Purple dots indicate aquaculture products from Cuba, Ecuador, Nicaragua, and Panama. Map modified from OSM Standard Map, QGIS. FAO major fishing areas are marked in blue lines.

Twelve species (60% of the 20 DNA-identified species) were not declared in any of the labels here analysed (see Table 2): *Heterocarpus calmani, Metapenaeopsis andamanensis, Metapenaeopsis barbata, Metapenaeus affinis, Metapenaeus ensis, Metapenaeus monoceros, Parapenaeus fissuroides, Penaeopsis jerryi, Penaeus merguiensis, Pleosionika quasigrandi, Solenocera koelbeli, Trachysalambria longipes.* Some species declared on the label (Table 1) did not appear in any of the shrimp packs, like the genera *Trachypenaeus* and *Parapenaeopsis*. The species found in this study belong to three families: Penaidae, Pandalidae, and Solenoceridae. The genus *Solenocera* was the most frequently employed as a substitute (Table 2).

At least some substitutions found in this study could be either deliberate or due to careless manufacturing (species sorting before peeling, or mixture of peeled shrimp while packaging), because the species disclosed in the labels and those employed as substitutes were morphologically very different in some cases. Examples are the substitution of *Solenocera* for *Heterocarpus*, *Plesionika* or *Metapenaeopsis* that were found in the package G5 (Fig. 2, Supplementary Table 1).

3.3. *Overview of heavy metals concentration*

Heavy metals concentrations for the analysed samples are in Supplementary Table 2. Mean heavy metal concentrations for the 20 analysed packs are shown in Table 3; some values show a large standard deviation, which is normal especially in mislabelled products when there is a mixture of species. The contents of the ensemble of eight metals were significantly different among packs (9999 permutations PERMANOVA with $F = 17.82$, $p < 0.001$).

Post-hoc pairwise comparisons allowed to identify several groups whose members (packs) were not significantly different to each other, represented in Table 4 by shaded vertical cells. Five distinct groups were found: one containing L4 and L6 packs; a large group with 15 packs; another of three overlapping with the former that containing G5, G7 and G6; and two groups represented by single packs: L7; G2. The two single-pack groups contained species declared on the label that were not found in any other pack: *Penaeus latisulcatus* from the West Indian Ocean in L7, and *Parapenaeus longirostris* from the East Atlantic in G2. Except for these two, the rest of the groups were not clearly distributed by species or region, containing different species from different regions.

Ten individuals (10%) distributed in five packs (Table 4) did not accomplish European regulations (maximum tolerable limit of mercury, lead, and cadmium being 0.5 mg kg^{-1}, Commission Regulation (EC) No 1881/2006 of December 19, 2006). For cadmium, five *Pleoticus muelleri* from Southwest Atlantic Ocean (FAO 41) and one *Penaeus latisulcatus* from West Indian Ocean (FAO 51) were over tolerable limits. Three samples were over the maximum limit for lead: one *Metapenaeus ensis* (Western Indian Ocean, FAO 51), one *Metapenaeus affinis* (Eastern Indian Ocean, FAO 57), and one *Penaeus indicus* from the West Indian Ocean (FAO 51). One *Penaeus vannamei* specimen from Nicaragua (aquaculture) surpassed the mercury limits (Table 4, Supplementary Table 2). Summarizing, two individuals sold as shrimp and eight sold as prawn should not be marketed in Europe.

Table 2
Species detected from DNA barcoding in the analysed samples, and number of individuals of each species declared on product labels or employed as substitutes. Suborder, family, and the acronym employed in this study are given.

Suborder	Family	Species	Declared on label	Substitute	Acronym
Pleocyemata	Pandalidae	*Heterocarpus calmani*	–	1	H.c
Dendrobranchiata	Penaeidae	*Metapenaeopsis andamanensis*	–	1	Mpp.a
Dendrobranchiata	Penaeidae	*Metapenaeopsis barbata*	–	2	Mpp.b
Dendrobranchiata	Penaeidae	*Metapenaeus ensis*	–	2	Mp.e
Dendrobranchiata	Penaeidae	*Metapenaeus monoceros*	–	3	Mp.m
Dendrobranchiata	Penaeidae	*Metapenaeus affinis*	–	2	Mp.a
Pleocyemata	Pandalidae	*Pandalus borealis*	5		Pa.b
Dendrobranchiata	Penaeidae	*Parapenaeus fissuroides*	–	3	Pap.f
Dendrobranchiata	Penaeidae	*Parapenaeus longirostris*	5		Pap.l
Dendrobranchiata	Penaeidae	*Penaeopsis jerryi*	–	1	Pp.j
Dendrobranchiata	Penaeidae	*Penaeus indicus*	5	2	P.i
Dendrobranchiata	Penaeidae	*Penaeus latisulcatus*	5		P.l
Dendrobranchiata	Penaeidae	*Penaeus merguiensis*	–	1	P.m
Dendrobranchiata	Penaeidae	*Penaeus vannamei*	25	–	P.v
Dendrobranchiata	Solenoceridae	*Pleoticus muelleri*	15	–	Pl.m
Pleocyemata	Pandalidae	*Plesionika quasigrandis*	–	1	Pk.q
Dendrobranchiata	Solenoceridae	*Solenocera crassicornis*	5	4	S.c
Dendrobranchiata	Solenoceridae	*Solenocera koelbeli*	–	9	S.k
Dendrobranchiata	Solenoceridae	*Solenocera melantho*	2	–	S.m
Dendrobranchiata	Penaeidae	*Trachysalambria longipes*	–	1	T.l

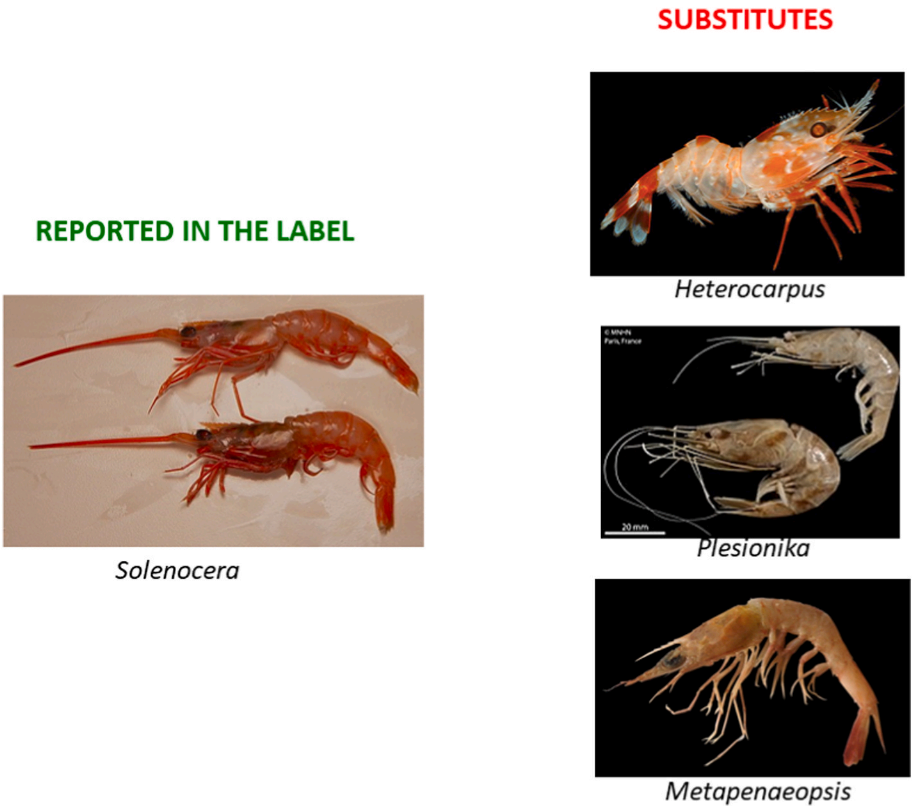

Fig. 2. Pictures of specimens of the genus reported in the label of the package G5 (*Solenocera* sp.) and of substitute genera (*Heterocarpus*, *Plesionika*, *Metapenaeopsis*) found from DNA. Images of public domain taken from the National Oceanic and Atmospheric Administration (https://www.noaa.gov/) and the World Register of Marine Species (https://www.marinespecies.org/).

3.4. Heavy metals concentration according to production region and species

The mean concentration of heavy metals in the individuals from the different production areas is represented in Fig. 3. Significant differences between the individuals from different areas were found (Supplementary Table 3). Significant post hoc tests by pairs taking as reference the area with the most contaminated individuals for each metal are indicated in Fig. 3, where individuals caught from West Indian Ocean contained more chromium, zinc, lead, and nickel, while those from the Central/East Atlantic Ocean contained more arsenic. These two areas, West Indian and Central/East Atlantic Oceans, did not show any significant difference to each other for any heavy metal. Other areas to highlight are the Southwest Atlantic Ocean, with the highest concentration of cadmium, and shrimps and prawns from Central America aquaculture for mercury and copper. On the other hand, East Indian Ocean was the area with lower concentrations of all the heavy metals analysed except chromium (Fig. 3).

Table 3

Mean (SD) content of the heavy metal analysed, in each package. Products sold as prawn and shrimp are coded with L and G respectively.

Pack	Substitutes %	Cr	Ni	Cu	Zn	As	Cd	Hg	Pb
L1	0	147.2 (88.0)	133.9 (69.0)	14,274.5 (3698.0)	13,356.0 (1024.3)	524.4 (53.1)	2.8 (1.3)	169.0 (191.5)	69.4 (61.4)
L2	0	204.8 (299.0)	116.3 (133.7)	3388.0 (1866.6)	8936.8 (1394.9)	321.6 (57.2)	1.9 (3.7)	32.0 (7.1)	57.2 (66.7)
L3	0	148.1 (90.8)	77.4 (51.3)	2398.7 (703.7)	7317.9 (1144.7)	141.4 (26.3)	1.1 (0.7)	17.8 (4.8)	35.3 (16.0)
L4	0	130.7 (141.2)	75.8 (36.6)	26,683.2 (10,875.1)	16,476.5 (1316.4)	995.5 (177.9)	1.0 (0.9)	23.2 (3.1)	142. (11.7)
L5	0	16.5 (6.7)	22.2 (9.9)	914.7 (71.2)	8441.3 (397.3)	1356.4 (194.6)	65.8 (19.3)	15.1 (2.6)	4.1 (1.5)
L6	0	1632.4 (648.4)	860.1 (306.0)	22,372.9 (5739.6)	17,307.1 (3593.1)	9464.4 (1218.9)	104.6 (46.3)	44.9 (9.8)	349.0 (163.6)
L7	0	109.7 (92.4)	139.0 (75.2)	4370.2 (1432.8)	20,162.4 (6836.2)	19,013.8 (4101.1)	408.9 (307.8)	20.5 (14.0)	23.7 (20.8)
L8	0	989.2 (593.9)	648.4 (404.5)	10,252.8 (3475.4)	15,221.5 (1220.1)	2564.5 (838.4)	777.3 (137.9)	35.4 (20.3)	303.6 (170.6)
L9	0	12.9 (3.4)	40.9 (17.7)	1376.1 (280.4)	8321.2 (418.4)	2033.7 (653.9)	95.4 (41.4)	25.4 (6.5)	2.0 (0.9)
L10	0	343.5 (736.7)	212.5 (405.8)	13,402.8 (3570.4)	1,3842.6 (1164.3)	332.4 (201.5)	3.1 (3.4)	18.0 (3.8)	46.7 (100.3)
G1	0	14.4 (5.3)	83.0 (59.7)	1961.8 (798.9)	7748.4 (939.5)	8695.7 (4682.1)	8.4 (3.4)	22.7 (4.8)	3.2 (1.0)
G2	0	184.6 (90.1)	231.7 (139.9)	10,992.3 (3694.3)	14,054.1 (1668.5)	17,351.3 (5144.3)	193.1 (126.6)	37.5 (9.0)	20.3 (9.9)
G3	0	262.0 (205.6)	159.4 (113.0)	874.1 (170.9)	5947.3 (707.5)	835.1 (113.4)	12.2 (9.5)	10.7 (3.2)	78.6 (60.0)
G4	100	124.0 (133.5)	88.3 (69.1)	873.1 (136.5)	5915.9 (555.8)	2883.4 (728.2)	18.6 (17.8)	22.6 (13.1)	107.1 (158.6)
G5	100	191.9 (295.1)	62.5 (63.2)	499.1 (149.3)	5211.6 (1428.3)	560.4 (173.1)	25.7 (15.9)	21.1 (6.6)	13.6 (16.3)
G6	100	49.8 (33.2)	43.9 (19.7)	526.5 (167.4)	4228.8 (545.7)	350.9 (38.9)	7.8 (3.5)	5.2 (0.7)	14.3 (12.7)
G7	100	253.1 (182.1)	197.6 (124.3)	1704.5 (960.9)	5377.1 (319.1)	738.8 (122.5)	15.9 (13.3)	10.3 (5.0)	49.2 (42.3)
G8	60	570.2 (376.1)	414.1 (269.8)	1344.8 (373.9)	6283.0 (1013.5)	3422.9 (811.3)	29.3 (17.6)	55.6 (30.6)	239.1 (123.9)
G9	100	236.7 (198.2)	204.7 (151.5)	1138.7 (471.4)	5032.9 (1314.0)	4575.6 (1212.6)	27.2 (25.3)	28.8 (9.6)	89.3 (82.8)
G10	100	1577.5 (2476.1)	1640.6 (2438.0)	4421.4 (4475.5)	16,014.1 (15,747.9)	1687.6 (1754.6)	143.4 (174.8)	19.7 (16.8)	355.4 (433.9)

Overall, prawns of the genus *Penaeus* were the most polluted (Fig. 4). *Penaeus indicus* (all individuals caught at West Indian Ocean) was the species with the highest levels of copper, nickel, chromium, and lead; while *Penaeus vannamei* (from aquaculture in Central America) exhibited the highest levels of mercury, and *Penaeus latisulcatus* (caught at West Indian Ocean) the highest levels of arsenic, zinc, and cadmium (Fig. 4).

As in the comparison by production area, significant differences between species were found for every metal analysed (Supplementary Table 4). Post-hoc pairwise tests (significant ones marked as asterisks in Fig. 4) showed that *Penaeus indicus* and *Penaeus latisulcatus*, the most polluted species in this study, were quite similar, exhibiting significant differences to each other only for arsenic and cadmium. In the other extreme, *Solenocera crassicronis* (caught from Northwest Pacific Ocean and West Indian Ocean) had the lowest concentration of heavy metals differing significantly from the most polluted species.

3.5. Health risk assessment results

The risk indices calculated are in Tables 5 and 6. THQ and TTHQ values higher than one indicates that the consumption of shrimps and prawns poses health risks if consumed daily (Steinhausen et al., 2022). For single metals, only the THQ for arsenic was higher than one, as well as the TTHQ in some species (*Penaeus indicus, Penaeus latisulcatus, Pandalus borealis* and *Parapenaeus longirostris*) (Table 5). For these species, EWI values were higher than the recommended PTWI, as it was also the arsenic EWI of *Solenocera koelbeli* for pregnant women (Table 5). All together, these results would indicate a significant risk of arsenic ingestion derived from the consumption of some imported shrimps and prawns commercialized in northern Spain.

In all the species, except for *Pandalus borealis,* CR_{lim} was smaller than the CR (0.041 kg day^{-1}) for chromium, and for arsenic all (Table 6), suggesting carcinogenic health risk if the metals were in their carcinogenic form (Steinhausen et al., 2022).

3.6. Inference of heavy metal risk by species substitution

The difference between the content of the eight heavy metals analysed for the shrimp species reported on the package label (authentic products) and the mislabelled substitutes was significant (PERMANOVA with F = 8.32, p = 0.001). Regarding the toxic metals, the content of arsenic was significantly higher in authentic products (mean = 8289.8 ppb, 95% confidence interval [4362.6, – 12,217]) than in substitutes (mean = 1957 ppb, interval [1331.8–2582.3]); t = 4.57, $p = 3.4E-5$. The mean contents of Cd (66.16 ppb, [11.71–120.61]), versus 38.85 [11.19–66.52]) and Hg (26.4 ppb, [18.77–34.02] versus 21.9 ppb [14.87–28.96]) were also higher in authentic products, but the difference was not significant (t = 1.04 and 0.82, respectively, both not significant). In contrast, the mean content of lead was lower in the authentic (mean = 57.68 ppb, [12.74–102.62]) than in the substitute shrimp (mean = 117.26 ppb, [43.21–191.3]), although the difference was not significant (t = 1.42, $p = 0.16$). In summary, taking all the substitutes of this study as a whole, mislabelling would not encompass a significant increase of heavy metal ingestion.

However, that general result should be taken with caution because as seen above there are differences between species and regions for heavy metal contents. A significant effect of the catch region was found for *Solenocera crassicornis*. This species was a substitute in two packs from Northwest Pacific (n = 4) and an authentic product (as in the label) in one pack from West Indian Ocean (n = 5). From a higher metal pollution in West Indian Ocean products (Fig. 2), in this particular case the substitutes were cleaner than the authentic product: Northwest Pacific substitute individuals exhibited significantly lower metal contents than those from West Indian Ocean (multivariate rank test with R = 0.481, $p = 0.008$), mean contents (SD) being (West Indian/Pacific): Cr, 262.0 (205.6)/42.8 (37.6); Ni, 159.4 (113.0)/45.4 (22.6); Cu, 874.1 (170.9)/583.2 (324.2); Zn, 5947.3 (707.5)/4569.8 (837.6); As, 835.1 (113.4)/950.7 (1148.6); Cd, 12.2 (9.5)/6.5 (1.1); Hg, 10.7 (3.2)/6.5 (3.1); Pb, 78.6 (60.0)/17.4 (13.4) ppb.

The effect of the species could be also important, although difficult to detect in this particular study. In all the packs with substitutions (G4 to G10; Table 1), substitutes belonged to a mixture of species including many that were not found on labels and were scarcely represented by only one or two individuals (Table 2). Thus, a comparison of metal contents between substitute and label species was not possible in most

Table 4

Summary table showing groups of packs by heavy metals concentration. Packs not significantly different to each other are marked with the same colour in vertically contiguous cells (e.g., L4 & L6). The number of specimens for each pack over tolerable metal concentration from EU legislation are indicated as "Risk samples". Species acronyms as in Table 2.

Pack code	Sold as	Pack groups		Risk samples	Production Region	Species
L7	Prawn	▥		1 (Cd)	West Indian Ocean	P.l
L6	Prawn		▤	1 (Pb)	West Indian Ocean	P.i
L4	Prawn		▤		Aquaculture Cuba	P.v
L2	Prawn	🟦			Aquaculture Ecuador	P.v
L1	Prawn	🟦		1 (Hg)	Aquaculture Nicaragua	P.v
L10	Prawn	🟦			Aquaculture Panama	P.v
L3	Prawn	🟦			No information	P.v
L9	Prawn	🟦			Southwest Atlantic Ocean	Pl.m
L5	Prawn	🟦			Southwest Atlantic Ocean	Pl.m
L8	Prawn	🟦		5 (Cd)	Southwest Atlantic Ocean	Pl.m
G1	Shrimp	🟦			Northwest Atlantic Ocean	Pa.b
G9	Shrimp	🟦			Northwest Pacific Ocean	S.k, S.m
G8	Shrimp	🟦			Northwest Pacific Ocean	Pap.f, S.c, S.k
G4	Shrimp	🟦			Northwest Pacific Ocean	Pap.f, S.k, T.l
G10	Shrimp	🟦		2 (Pb)	Indian Ocean	Mp.a, Mp.e, Mp.m, P.i, P.m
G3	Shrimp	🟦			West Indian Ocean	S.c
G5	Shrimp	🟦	▒		East Indian Ocean	H.c, Mpp.a, Pap.f, Pk.q, Pp.j
G7	Shrimp	🟦	▒		East Indian Ocean	Mp.a, Mp.e, Mp.m, P.i
G6	Shrimp		▒		Northwest Pacific Ocean	Mpp.b, S.c
G2	Shrimp	■			East Central Atlantic Ocean	Pap.l

cases. Focusing on the most frequent substitute *Solenocera koelbeli* (9 individuals), it replaced the label species in G4, G5, G8 and G9 samples (together with other species). In Table 5 it is shown that *Solenocera koelbeli* encompasses a significant risk of arsenic for pregnant women, while for *Solenocera crassicornis* (a species frequently substituted by *Solenocera koelbeli*) the risk is not significant. Thus, there would be an increased risk associated to arsenic when *Solenocera koelbeli* is a substitute.

4. Discussion

4.1. Mislabelling and resource sustainability

This study revealed important new data regarding mislabelling of shrimps and prawns in Europe. A high proportion of labels were non-compliant with European regulations for labelling of fishery products. The scientific name was the most frequently missing piece of information on labels, as pointed out by other authors (Feldmann et al., 2021; Paolacci et al., 2021). A previous study in Spain published seven years ago showed as many as 63% products with incomplete scientific names (Galal-Khallaf et al., 2016). A 40% of labels with the same mistake found in the present study would represent an improvement in comparison, but the level of incomplete labelling is still very high, which hampers the possibility of informed consumers to select the products they really want (Armani et al., 2015; Paolacci et al., 2021).

In this study, species substitution happened only in products peeled (thus unrecognizable) and labelled as shrimp, not in prawn. This points to mislabelling in products containing specimens of small size. It may be accidental, as in many other cases of mixed fisheries (Blanco-Fernandez et al., 2022; Garcia-Vazquez et al., 2012), because relatively small species will be difficult to distinguish from each other. In some cases of substitute species morphologically different (Fig. 2), the error could be produced during manufacturing. The deliberate fraud cannot be ruled out in those cases, although in principle it would be less probable. Shrimp are generally cheaper than bigger prawns, and fraud is generally associated to more appreciated, expensive species that are replaced by cheaper ones (Blanco-Fernandez, Ardura, et al., 2021), thus fraud in shrimp would be economically less advantageous than in prawn.

Even if it comes from unintentional errors during species sorting, the mislabelling detected here could encompass a serious conservation risk. Some substitute species like *Solenocera crassicornis, Metapenaeopsis barbata, Penaeus merguiensis, Penaeus indicus and Metapenaeus* spp., are overexploited in different fishing areas (Abbas et al., 2020; Galal-Khallaf et al., 2016; Jayawardane et al., 2002). This is a major concern for the

Fig. 3. Heavy metal concentration depending on the origin of the product. Mean concentration of each heavy metal in the samples of each catch area, with standard deviation as capped bars. Significant differences from the sample with the highest concentration are shown with an asterisk of the same colour.

ecosystem. Shrimps and prawns generally suffer from overexploitation in the countries where they are produced, due to a higher domestic consumption added to fishing for exportations (Sharawy et al., 2017). The use of overexploited stocks as substitute species - thus undeclared in official reports - leads to underestimates of real catch, hindering any conservation and sustainable exploitation plans for natural populations (El-Chichakli, et al., 2016; Marko et al., 2004).

4.2. Heavy metal exposure through consumption of imported shrimp and prawn

The heavy metal pollution detected in this study in prawn of the genera *Penaeus* and *Parapenaeus*, and the shrimp *Pandalus borealis* suggests a possible risk for consumers as indicated from their TTHQ values (Table 5). Moreover, a few individuals of these species exceeding the tolerable EU limits for commercialization in cadmium (one *Penaeus latisulcatus*), lead (one *Penaeus indicus*) and mercury (one *Penaeus vannamei*). The risks were calculated for an average daily intake of 41 g/day reported by Abd-Elghany et al. (2020) for crustaceans, but in some countries, adults consume more seafood and shellfish than in others. For example, Spain and Portugal are the European countries with the highest consumption of fisheries and aquaculture products (https://oceans-and-fisheries.ec.europa.eu/facts-and-figures/facts-and-figures-common-fisheries-policy/consumption_en). Therefore, the risk would be higher there than in other European countries. From higher heavy metal content in these species, health risk assessment indices pointed at *Penaeus indicus* and *Penaeus latisulcatus* as the least safe (Fig. 3). Their daily consumption may pose health risks, also applicable to *Pandalus borealis* and *Parapenaeus longirostris* for their arsenic content. High concentrations of total arsenic in pregnancy are associated with many adverse outcomes in mother and child (Ashley-Martin, Fisher, Belanger, Cirtiu, & Arbuckle, 2022). If confirmed from further studies, a lower consumption of these shrimps and prawns would be recommended for pregnant women. Indeed, this should be taken with caution avoiding creating consumer's alarm, because the number of samples analysed in this study for the species most commonly used as substitutes was somewhat limited. Shrimps and prawns can absorb metals from the surrounding environment, as other crustaceans do (Abd-Elghany et al., 2020). In addition to regional differences in metal pollution, there are differences between species for heavy metal intake and accumulation. An example is the species *Penaeus vannamei* that would accumulate more heavy metals than other species. In their review of Persian Gulf prawns, Shahsavani et al. (2017) indicated that *Penaeus vannamei* contained the highest concentration of lead. Ruelas-Inzunza et al. (2004) compared species of wild prawns from Mexico, finding that *Penaeus vannamei* had the highest concentration of mercury in the hepatopancreas. Delgado-Alvarez et al. (2015) found more mercury in *Penaeus vannamei* farmed in Mexico than in other wild and farmed shrimps. The results of *Penaeus vannamei*, imported from Central America aquaculture and being the one with the highest mercury content, would be consistent with those results. Prawn farms are in coastal areas that receive mercury from terrestrial environments, and through estuaries and coastal water bodies used for pond stocking; moreover, aquaculture feeds and chemicals used during production processes are also important sources of

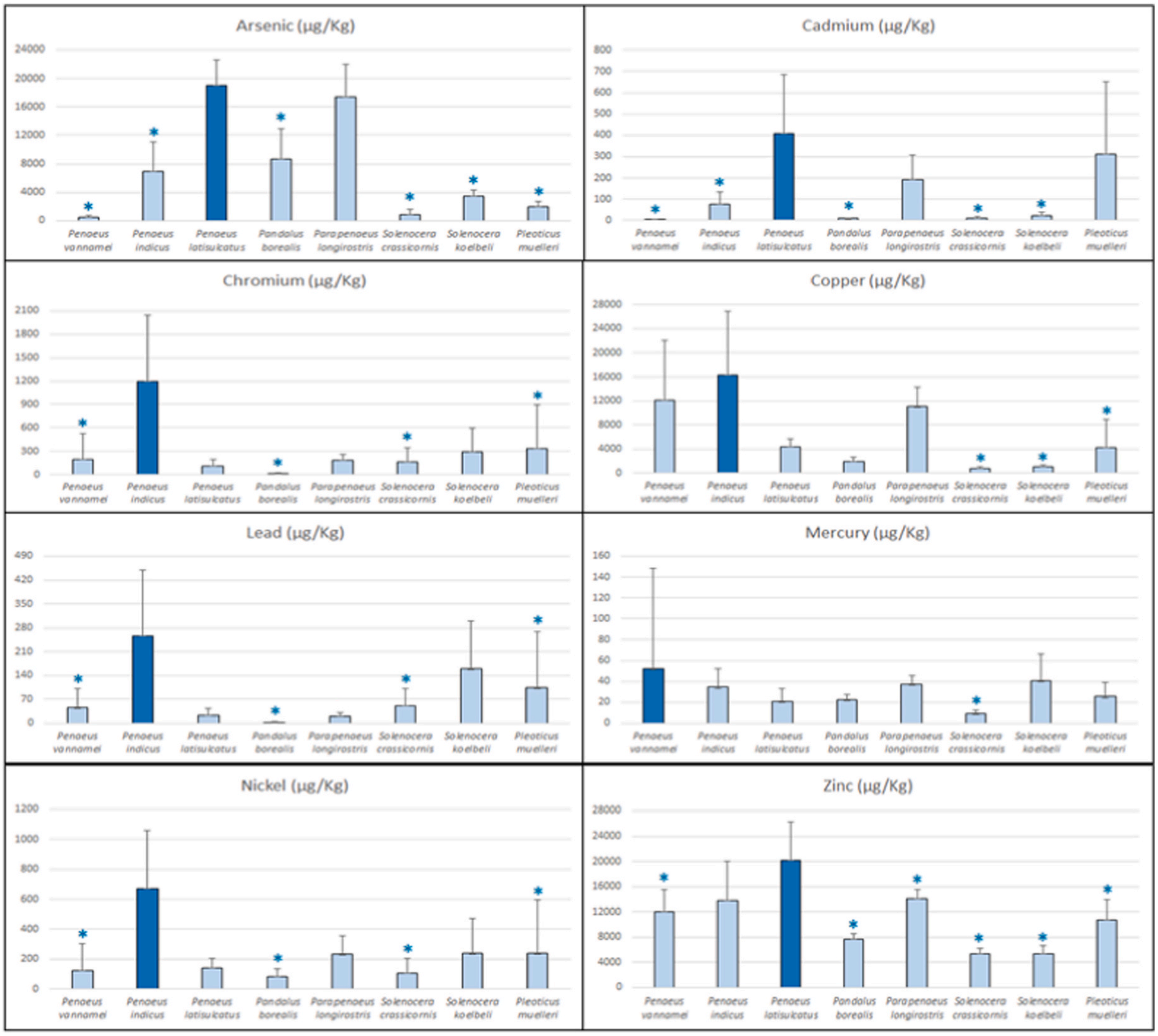

Fig. 4. Heavy metal concentration by species. Mean concentration and standard deviation of each heavy metal are shown for each species. Those species exhibiting significant differences from the species with the highest concentration (dark blue) are depicted with an asterisk.

mercury in ponds (Delgado-Alvarez et al., 2015; Lacerda et al., 2011). All this could explain high mercury concentration of *Penaeus vannamei* in this study.

Another example of apparent higher accumulation of metals than in other species is *Penaeus indicus*, although in this case not all the studies were consistent. Mitra et al. (2010, 2012) found that *Penaeus indicus* had some of the highest concentrations of lead and cadmium compared to other prawns. However, in other studies (Fakhri et al., 2018) *Penaeus indicus* did not stand out for any high concentration of heavy metals. The results of this study would support high levels of heavy metals in this species, at least in some exports to Europe, but not higher than those of *Penaeus latisulcatus* from the same region (West Indian Ocean). This should be studied further.

In the present study, it is difficult to discern the effect of the species from that of the catch area because, except for *Solenocera crassicornis*, most of species were caught from only one region. That said, the results of this study point at the West Indian Ocean as a hotspot of heavy metal pollution (Fig. 2). *Penaeus latisulcatus* (from that zone) stood out for arsenic, zinc, and cadmium in the present work, and *Penaeus indicus* exhibited the highest amounts of nickel, chromium, copper, and lead (Fig. 3). *Solenocera crassicornis* had very low metal concentrations in our study, as in others (Wan et al., 2022), but those from West Indian Ocean were also the most polluted. There are many references of high heavy metal pollution therein, with hotspots of arsenic, cadmium, mercury, and lead, probably derived from mining activities in Africa (Garcia-Vazquez et al., 2021). Heavy metal contamination in this area can also be due to volcanic activity (Sen Gupta & Singbal, 1988). Some islands of the West Indian Ocean such as Reunion, Mascarene and Comoros Islands have volcanic activity. Dœlsch et al. (2006) showed that the natural pedogeochemical background could explain the chromium, copper, nickel, and zinc concentrations in the soils of Reunion Island. Furthermore, the oil industry also contributes significantly to heavy metals pollution (e.g., Vane et al., 2020). This type of industry is a chronic problem in the northern Indian Ocean, as two oil tanker routes from Middle Eastern countries pass through the Arabian Sea, one of them heading to the western hemisphere through the Mozambique Channel (Sen Gupta & Singbal, 1988). The next polluted region would be West African waters. Shrimp from the Central/East Atlantic Ocean (FAO area 34) joined the West Indian Ocean individuals as the most polluted ones, being consistent with high metal pollution also in that zone (Garcia-Vazquez et al., 2021). Therefore, the results of this study can be principally explained from the pollution of the fishing area.

From these results, heavy metal risks would increase in mislabelled products when the substitutes come from polluted regions, as it would be the case of West Indian Ocean in this study. Substituting a species by others coming from less polluted areas may even decrease the risk of heavy metal pollution, as happened here with *Solenocera crassicornis*. Only in one case of substitution would the risk of heavy metal ingestion

Table 5

Heavy metal risk indices for adults and for pregnant women, calculated for the most abundant species found in this study. For each species, the values of EWI (Estimated Weekly Intake)/THQ (Target Hazard Quotient) of the analysed metals (mg/kg (w/w) are presented. Values encompassing a possible health risk are highlighted in bold. PTWI (Values over Provisional Weekly Tolerable Intake) and TTHQ (Total Target Hazard Quotient) are also presented. When a PTWI range was specified, the precautionary approach was adopted by selecting the lowest value (Steinhausen et al., 2022).

Metal	PTWI		Penaeus vannamei	Pleoticus muelleri	Penaeus indicus	Penaeus latisulcatus	Pandalus borealis	Parapenaeus longirostris	Solenocera crassicornis	Solenocera koelbeli
Adults										
Cr	0.023		0.0008/0.0228	0.0014/0.0397	0.0049/0.1390	0.0004/0.0128	5.91E-05/0.0017	0.0008/0.0216	0.0007/0.0193	0.0012/0.0339
Ni	0.035		0.0005/0.0036	0.0010/0.0069	0.0027/0.0196	0.0006/0.0041	0.0003/0.0024	0.0009/0.0068	0.0004/0.0032	0.0010/0.0069
Cu	0.35–3.5		0.0493/0.1870	0.0171/0.0659	0.0667/0.2567	0.0179/0.0689	0.0080/0.0309	0.0450/0.1733	0.0031/0.0117	0.0041/0.0156
Zn	2.1–7		0.0491/0.0234	0.0437/0.0208	0.0564/0.0268	0.0826/0.0393	0.0317/0.0151	0.0576/0.0274	0.0218/0.0104	0.0222/0.0106
As	0.015		0.0019/0.0903	0.0081/0.3871	**0.0283**/**1.3473**	**0.0779**/**3.7080**	**0.0356**/**1.6958**	**0.0711**/**3.3838**	0.0036/0.1729	0.0140/0.6666
Cd	0.007		8.03E-06/0.0011	0.0013/0.1830	0.0003/0.0450	0.0017/0.2392	3.34E-05/0.0049	0.0008/0.1130	3.96E-05/0.0057	8.56E-05/0.0122
Hg	0.005		0.0002/0.0304	0.0001/0.0148	0.0001/0.0201	8.38E-05/0.120	9.30E-05/0.0133	0.0002/0.0220	3.63E-05/0.0052	0.0002/0.0236
Pb	0.025		0.0002/0.0065	0.0004/0.0151	0.0010/0.0375	9.72E-05/0.0035	1.31E-05/0.0005	8.32E-05/0.0030	0.0002/0.0075	0.0006/0.0232
		TTHQ	0.37	0.73	**1.89**	**4.09**	**1.76**	**3.75**	0.24	0.79
Pregnant women										
Cr	0.023		0.0009/0.0250	0.0015/0.0435	0.0053/0.1528	0.0005/0.0140	6.47E-05/0.0018	0.0008/0.0237	0.0007/0.0211	0.0013/0.0371
Ni	0.035		0.0006/0.0039	0.0011/0.0076	0.0030/0.0214	0.0006/0.0045	0.0004/0.0027	0.0010/0.0074	0.0005/0.0035	0.0011/0.0076
Cu	0.35–3.5		0.0539/0.2056	0.0188/0.0722	0.0730/0.2811	0.0196/0.0755	0.0088/0.0339	0.0493/0.1898	0.0033/0.0129	0.0044/0.0171
Zn	2.1–7		0.0537/0.0256	0.0478/0.0228	0.0617/0.0294	0.0904/0.0431	0.0347/0.0165	0.0630/0.0300	0.0239/0.0114	0.0243/0.0116
As	0.015		0.0021/0.0989	0.0089/0.4239	**0.0310**/**1.4753**	**0.0853**/**4.0602**	**0.0390**/**1.8569**	**0.0778**/**3.7052**	0.0040/0.1893	**0.0153**/0.7299
Cd	0.007		8.80E-06/0.0013	0.0014/0.2004	0.0003/0.0493	0.0018/0.2620	3.75E-05/0.0054	0.0009/0.1237	4.34E-05/0.0062	9.38E-05/0.0134
Hg	0.005		0.0002/0.0333	0.0001/0.0162	0.0002/0.0220	9.19 E−05/0.0131	0.0001/0.0146	0.0002/0.0240	3.98E-05/0.0057	0.0002/0.0259
Pb	0.025		0.0002/0.0071	0.0005/0.0165	0.0011/0.0410	0.0001/0.0038	1.43E-05/0.0005	9.12E-05/0.0033	0.0002/0.0082	0.0007/0.0254
		TTHQ	0.40	0.80	**2.07**	**4.48**	**1.93**	**4.11**	0.26	0.87

(arsenic) increase in the present study: when *Solenocera koelbeli* is sold as a substitute of *Solenocera crassicornis*. Therefore, it is recommended to consider the geographical origin of imports, emphasizing the quality of controls in products coming from polluted regions.

4.3. Study limitations

The main limitation of this study was a limited number of samples analysed. Although sufficient to detect and quantify mislabelling, the dispersion of substitute species hindered the power of statistical tests for the differences between species for heavy metal contents. Larger sample sizes should be considered in further studies on shrimp and prawn.

5. Conclusions and recommendations

Non-compliance of European regulations was observed in most of the analysed labels of shrimp and prawn products imported in Spain, principally due to the absence of the complete scientific names.

A total of 20 species, from which 12 were substitutes, were identified from DNA barcoding. A 66% of the shrimps analysed were genetically identified as a species not disclosed in the label. Many of these species are overexploited, therefore, their use as substitutes likely contributes to such overexploitation that may cause subsequent population declines. Species substitution was not detected in prawns in this study.

Regarding heavy metal content, 10% of the analysed individuals did not comply with the European limits for heavy metals (Commission Regulation (EC) No 1881/2006 of December 19, 2006). Individuals fished in the West Indian Ocean and the Central/East Atlantic Ocean exhibited higher concentrations of heavy metals than those from other zones, having *Penaeus indicus* and *Penaeus latisulcatus* from West Indian Ocean the highest levels. These regions are polluted by mining and other anthropogenic disturbances that should be controlled to reduce heavy metal burdens in aquatic species.

According to the calculated indices, the following species could cause health risks if consumed daily by adults and pregnant women: *Pandalus borealis, Parapenaeus longirostris, Penaeus indicus* and *Penaeus latisulcatus.* If confirmed from further analyses, it would be advisable to reduce the intake of these species so preventing chronic diseases and cancer, especially for the possible effects of high arsenic concentrations.

The relative weight of the species biology and the catch region on the heavy metal content in shrimp and prawn needs to be studied further. More research on mislabelling, heavy metals and the natural populations of shrimps and prawns is advisable to provide further guidance to consumers and help enforce regulations compliance.

CRediT authorship contribution statement

Marta Pilar Ortiz-Moriano: Methodology, Investigation, Writing – original draft, preparation, Formal analysis, Data curation, Visualization, Writing – review & editing. **Gonzalo Machado-Schiaffino:** Writing – review & editing. **Eva Garcia-Vazquez:** Conceptualization, Data curation, Formal analysis, Visualization, Supervision, Writing –

Table 6
Values of CR_{lim} (Carcinogenic risks (kg day^{-1})) for inorganic arsenic, chromium (VI) and lead in the analysed species, estimated for the general adult population and for pregnant women. Precautionary approach assuming all inorganic arsenic and chromium (VI). Values indicating significant health risk are in bold.

	Species	As (inorganic)	Cr (VI)	Pb
Adults	*Penaeus vannamei*	**0.00719**	**0.00101**	1.85130
	Penaeus indicus	**0.00118**	**0.00007**	0.32168
	Penaeus latisulcatus	**0.01278**	**0.00002**	3.47502
	Pandalus borealis	0.09717	**0.00005**	25.81588
	Parapenaeus longirostris	**0.00759**	**0.00003**	4.06033
	Solenocera crassicornis	**0.00852**	**0.00053**	1.60483
	Solenocera koelbeli	**0.00484**	**0.00014**	0.51965
	Parapenaeus fissuroides	**0.00782**	**0.00013**	1.46784
	Metapenaeus monoceros	**0.00578**	**0.00068**	1.47227
	Pleoticus muelleri	**0.00413**	**0.00024**	0.79858
Pregnant women	*Penaeus vannamei*	**0.00657**	**0.00092**	1.69068
	Penaeus indicus	**0.00107**	**0.00006**	0.29377
	Penaeus latisulcatus	**0.01167**	**0.00002**	3.17353
	Pandalus borealis	0.08874	**0.00005**	23.57615
	Parapenaeus longirostris	**0.00693**	**0.00002**	3.70806
	Solenocera crassicornis	**0.00778**	**0.00048**	1.46560
	Solenocera koelbeli	**0.00442**	**0.00012**	0.47456
	Parapenaeus fissuroides	**0.00714**	**0.00012**	1.34049
	Metapenaeus monoceros	**0.00527**	**0.00062**	1.34453
	Pleoticus muelleri	**0.00377**	**0.00021**	0.72930

review & editing, All authors have read and agreed to the published version of the manuscript. **Alba Ardura:** Supervision, Visualization, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodcont.2023.110193.

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