UNIVERSITAT DE BARCELONA

# Dietary exposure to neurotoxic and endocrine disruptive effects: The mercury case

Marco Capodiferro

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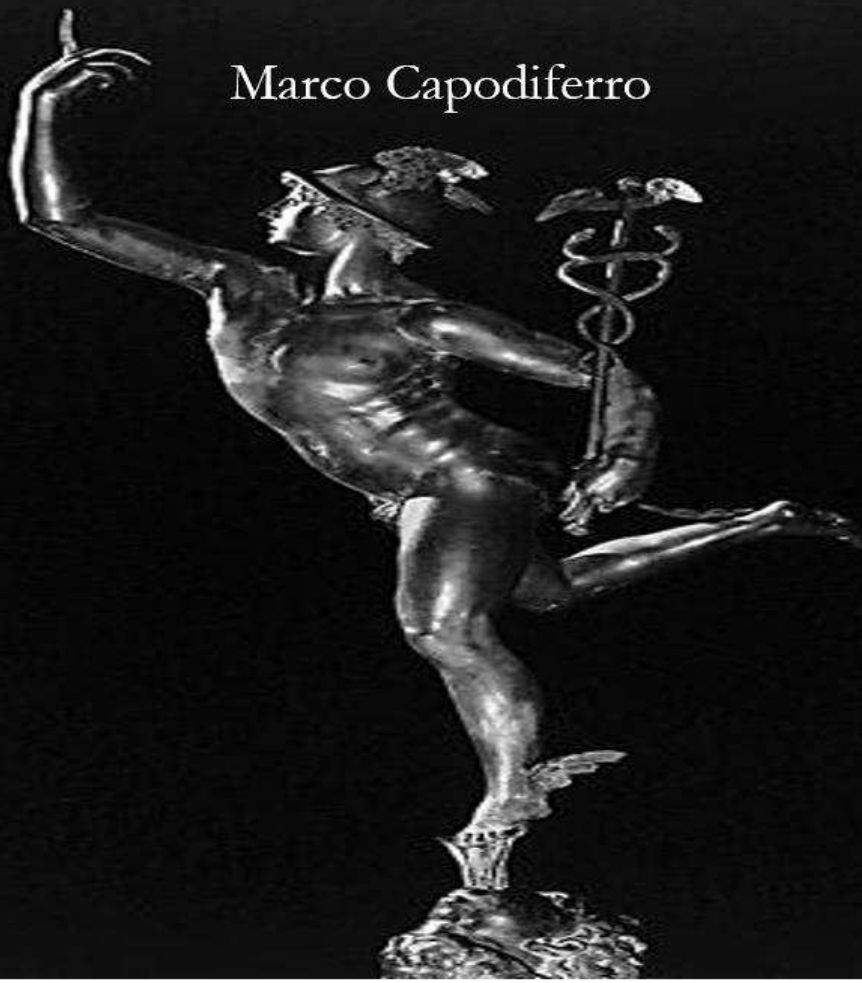

UNIVERSITAT DE BARCELONA

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# DIETARY EXPOSURE TO NEUROTOXIC AND ENDOCRINE DISRUPTIVE EFFECTS: THE MERCURY CASE

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# DIETARY EXPOSURE TO NEUROTOXIC AND ENDOCRINE DISRUPTIVE EFFECTS: THE MERCURY CASE

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El Dr. **Joan O. Grimalt Obrador**, Profesor de Investigación del Instituto de Diagnóstico Ambiental y Estudios del Agua (IDAEA), perteneciente al Consejo Superior de Investigaciones Científicas (CSIC),

**Certifica:**

Que la presente memoria presentada para optar al titulo de Doctor, titulada "***Dietary exposure to neurotoxic and endocrine disruptive effects: the mercury case***", ha sido realizada bajo mi dirección por el Sr. **Marco Capodiferro** en el Instituto Instituto de Diagnóstico Ambiental y Estudios del Agua (IDAEA), perteneciente al Consejo Superior de Investigaciones Científicas (CSIC), y que todos los resultados presentados son fruto del trabajo experimental realizado pr el mencionado doctorando.

Y para que así conste, expido y firmo el presente certificado.

Barcelona, 15 de noviembre de 2021

**Dr. Joan O. Grimalt**

*“Un viaggio di mille miglia comincia sempre con il primo passo”*

*“A journey of a thousand miles always begins with the first step” Lao Tzu*

*Alla mia famiglia...*

*Dedicated to my family...*

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*“Non c’è uomo più completo di colui che ha viaggiato, che ha cambiato venti volte la forma del suo pensiero e della sua vita” (Alphonse de Lamartine).* Con questo motto di vita e con la constante ricerca della completezza, ho intrapreso questo viaggio fantastico che sono stati questi tre anni del dottorato di ricerca. Sembra ieri, che questa avventura sia iniziata ed invece, eccoci qui, dopo tre anni. Come qualsiasi cosa che ho affrontato nella mia vita, ci ho messo tutto l’impegno e la dedizione possibile. Come disse Confucio “Ovunque tu vada, vacci con tutto il tuo cuore”, così arrivato a Barcellona ci ho lasciato un pezzo del mio cuore.

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*“Fatti non foste a viver come bruti ma per seguir virtute e canoscenza” (Dante Alighieri).* “Progressi eccezionali fluiscono dalla combinazione di colta pigrizia, pensiero intelligente ed estrema ambizione” (Richard Koch). In questo modo, ho iniziato a produrre i primi risultati. Nella mia permanenza durante questi tre anni, ho incontrato persone fantastiche (che menzionerò ad una ad una nei ringraziamenti per il supporto scientifico successivamente) che mi hanno sopportato e supportato, dando un consiglio e/o un aiuto, laddove fosse servito.

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*"Le parole di mamme e papà sono come interruttori della luce. Pronuncia una parola al momento giusto nella vita di un bambino ed è come illuminare tutta una stanza piena di possibilità" (Gary Smalley).* Consapevole del fatto che fare il genitore è il mestiere più difficile al mondo, sono immensamente grato ai miei genitori per i sacrifici che hanno fatto per permettermi di studiare senza alcuna preoccupazione e per avermi tirato su con dei sani valori che mi accompagnano in tutto quello che faccio. Grazie per il continuo sostegno che mi avete fatto arrivare sempre: la chiamata di fine giornata e il messaggino dell'ora di pranzo non mi ha mai fatto mancare il vostro calore e la vostra presenza. Se oggi sono la persona che sono, lo devo solo ed esclusivamente a voi che mi avete permesso di sviluppare la mia persona senza mai farmi dimenticare le cose importanti della vita. Quindi oggi cammino per il sentiero costruito da me, essendo la somma di voi due.

*"L'amore fraterno è il più durevole; assomiglia a una pietra preziosa che resiste ai più duri metalli e il cui valore si accresce con gli anni" (Hector Carbonneau)*. "Che comunque vada, mio fratello ci sarà" (Due su due - Articolo 31). Sentendo parlare di esperienze di persone che sono figli unici, di tanto in tanto mi sono fermato a pensare di come sarebbe stata triste la mia esistenza senza mia sorella Paola. Di sicuro avrei posseduto più cose, ma sarei stato immensamente più povero: non avrei imparato a condividere, non sarei stato empatico tanto quanto lo sono ora, non avrei apprezzato i rapporti umani tanto quanto lo faccio adesso, non avrei avuto una persona pronta a darti un consiglio al posto e al momento giusto, non avrei avuto modo di condividere gioie e dolori, non avrei avuto un abbraccio, una parola giusta, un messaggio quando ne avevo

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*"Testimoni del passato, garanzia del presente ed eredi del futuro: i nonni." (Carmine De Masi).* Sono stato molto fortunato nell'aver conosciuto tutti e quattro i nonni ed addirittura una bis-nonna. In punta di piedi e con la più grande sensibilità, hanno sempre visto muovere i miei passi con occhio vigile ed amorevole, vedendomi crescere. Custodi di un'altra età, sono stati sempre presenti nella mia vita, rallegrandosi delle mie conquiste. Ora due di loro non ci sono più, ma mi hanno dato tantissime cose che custodisco gelosamente dentro di me. Proprio durante il periodo del dottorato, ho perso nonna Italia; questa dannata pandemia e la lontananza non mi ha permesso di salutarla per l'ultima volta. La tua eredità vive in me, sempre. Non mi siederò alla tavola di casa tua per il caffè, ma avrai sempre un posto alla tavola del mio cuore. Spero che ovunque tu sia ora, tu sia orgogliosa di me. Ad ogni modo, ora mi sto godendo la presenza di nonna Maria Rosaria e di nonno Michele. Instancabili miei supporter, non hanno perso occasione per farmi arrivare la loro vicinanza ed il loro affetto in tutti i modi possibili durante questi tre anni. Non dimenticherò mai i loro occhi durante i vari rientri dalle vacanze. Se dovessi dipingere la felicità con un'immagine, la farei con i vostri occhi nonni miei! Il nostro tempo assieme è inestimabile ed è una benedizione avervi vicino. Grazie per tutto quello che avete fatto e che fate per me.

*"Quale la pianta, tali i frutti" (proverbio cinese)*. Avendo vissuto in una famiglia numerosa e variegata, ho trovato qualche difficoltà all'inizio della mia avventura qui a Barcellona in cui per forza di cose mi sono trovato da solo. Tutti i componenti della mia famiglia mi hanno comunque dimostrato la loro vicinanza e il loro supporto. Un grazie speciale va ad Alessandra, Emanuele, Gianmaria, Valeria, pepa Patrizia, pipo Andrea, zio Gianni, zia Barbara, zio Paolo, zia Urania, zia Adriana, per il continuo appoggio. Un grazie speciale va a Maurizio, Mariassunta e Barbara che sono stati sempre vicino durante questo percorso. Nel sostenere me ed Alessia nelle scelte di tutti i giorni, non ci hanno fatto mancare mai tutto il loro affetto. Sempre forieri di parole di conforto nei nostri confronti, hanno provato sempre a farci sentire in famiglia, nonostante la lontananza. Un grazie speciale va a Stefano, che durante questo periodo ha mostrato la sua vicinanaza sempre, tanto da venirmi a trovare qui a Barcellona a metà del mio percorso.

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*"E quindi uscimmo a riveder le stelle" (Dante Alighieri).* Così finisce l'Inferno di Dante. Così finisce la mia prima esperienza accademica fuori dall'Italia. La mia prima esperienza all'estero mi ha dato moltissimo e mi ritrovo con il bagaglio culturale notevolmente arricchito e con la consapevolezza di aver lavorato al meglio delle mie capacità. Ma come Dante, subito dopo l'uscita dall'Inferno, riparte alla volta del Purgatorio, così io mi proietto verso una nuova avventura. Come un pirata romantico alla costante ricerca di nuovi lidi inesplorati, non so ancora cosa mi riserva il futuro, quindi scruto l'orizzonte sperando di vedere subito una nuova terra e una nuova avventura. Guardando indietro a questa esperienza del dottorato, al pensiero delle tante cose fatte e alle persone incontrate, senza alcuna remora mi accingo ad affrontare nuove sfide e nuove possibilità, consapevole del fatto che se la nuova esperienza sarà bella anche solo la metà di come è stata quella del dottorato, ne sarà valsa la pena.

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*"There is no man more complete than one who has travelled, who has changed the form of his thought and his life twenty times" (Alphonse de Lamartine).* With this motto and with the constant search for completeness, I have embarked on this fantastic journey that these three years of the PhD have been. It seems like yesterday that this adventure has begun and instead, here we are, with three years behind us. I've put all my efforts and dedication into it. As Confucius said, “Wherever you go, go there with all your heart”, so when I arrived in Barcelona, I left a piece of my heart there.

*"If you follow your star, you will access in a glorious harbour" (Dante Alighieri).* Faced with a challenge that I had never faced, at first everything seemed new and impervious to me. However, as for Dante it was Virgil in the Divina Commedia, I had my guide in this path too. Joan O. Grimalt was my Virgil accompanying me in all the stages of this path, explaining in detail how things had to be done, enriching my cultural background, making me grow as a person and as a scientist, teaching me methods to be able to do the work consistently and in a proper way, always leaving me the space to be myself. In short, he was always close step by step, like a second father who lovingly and firmness let me move my first steps in this new context. Along with the best guidance, I began this journey with the aim of finding explanations for unsolved phenomena, trying to imitate the model of person that is Dr. Grimalt.

*"You were not made to live like brutes but to follow virtue and knowledge" (Dante Alighieri).* “Outstanding progress flows from the combination of cultured laziness, intelligent thinking and extreme ambition" (Richard Koch). Using this method, I started producing the first results. During these three years, I have met fantastic people (whom I will mention one by one in thanks for the scientific support later) who have endured and supported me, giving advice and / or help, where needed.

*"The universe only makes sense when we have someone to share our emotions with" (Paulo Coelho).* Luck wanted not to undertake this journey alone, but in the company of the woman who most of all saw inside me, Alessia. We have always found in each other a safe harbour where we can stop, recharge our being and leave better than we were. Accomplice of a lifetime and in life, she has solved many knots of me with her witty and pungent intelligence. Certainly, during our stay in Barcelona, we shared good moments and others less so. Let us remember that there have been no easy times: a virus

has imprisoned us at home, made us breathe little fresh air and forced us to see more Netflix than the people we love. I can only tell you that it was nt so tragic having you by my side all this time. I thank you for who you are and it is also thanks to you, if today I am reaching this milestone.

*"The words of moms and dads are like light switches. Say a word at the right moment in a child's life and it's like lighting up a whole room full of possibilities" (Gary Smalley).* Aware of the fact that parenting is the hardest job in the world, I just have to thank my parents for the sacrifices they made for allowing me to study without any worries and for raising me with healthy values that accompany me in everything I do. Thank you for the continuous support that you always gave me: the end of day phone call and the lunchtime message never made me lack your warmth and nearness. If today I am the person I am, I owe it solely and exclusively to you who have allowed me to develop my person without ever making me forget the important things in life. So today I am walking on the path I am building, being the sum of my mom and dad.

*"Brotherly love is the most lasting; it resembles a precious stone that resists the hardest metals and whose value increases over the years" (Hector Carbonneau).* "Whatever happens, my brother will be there" (Due su due - Articolo 31). Hearing about the experiences of people who are only child, I was thinking from time to time about how sad my existence would have been without my sister Paola. For sure, I would have possessed more things, but I would have been immensely poorer: I would not have learned to share, I would not have been as empathetic as I am now, I would not have appreciated human relationships as much as I do now, I would not have had a person ready to give you advice in the right place and at the right time, I would not have had the opportunity to share joys and sorrows, I would not have had a hug, a right word, a message when I needed it. Knowing that whatever happens, my sister will be there is one of the most beautiful things this life could give.

*"Witnesses of the past, guarantee of the present and heirs of the future: grandparents." (Carmine De Masi).* I was very lucky to have met all four grandparents and even a great-grandmother. On tiptoe and with the greatest sensitivity, they have always seen my steps move with a watchful and loving eye, watching me grow. Guardians of another age, they have always been present in my life, rejoicing in my achievements. Now two of them are gone, but they have given me so many things that I jealously guard inside of me. Just during the period of my doctorate, I lost grandmother Italia; this damned

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*"Like the plant, like the fruits" (Chinese proverb).* Having lived in a large and varied family, I found some difficulties at the beginning of my adventure here in Barcelona where I inevitably found myself alone. However, all the members of my family have shown me their closeness and their support. A special thanks goes to Alessandra, Emanuele, Gianmaria, Valeria, pepa Patrizia, pipo Andrea, uncle Gianni, aunt Barbara, uncle Paolo, aunt Urania, aunt Adriana, for their continued support. A special thanks goes to Maurizio, Mariassunta and Barbara, who have always been close during this journey. In supporting me and Alessia in our everyday choices, they never made us lack all their affection. Always harbingers of comfort words towards us, they have always tried to make us feel at home, despite the distance. A special thanks goes to Stefano, who has always shown his closeness during this period, so much so that he came to visit me here in Barcelona in the middle of my journey.

*"Friends are the family you can choose" (Audrey Hepburn).* Last but not least, a special thanks goes to friends. Naming them all would be impossible. Anyway, I would like to thank them all in private, the ones who have always been there and the recent ones. Thanks to the "Infognati" for giving me so many moments of light-heartedness, laughter and support. One of the things that I have missed the most is certainly our outgoes. Sometimes, I would have needed to have a beer with a long-time friend to disconnect from the routine or to just have a talk and share with you my thoughts. However, I hope that we will be able to reply soon. Thanks also to all the others who have been able to stay close to me during this period. Among all, I wanted to thank two people, Davide and Marco. Your messages and your words did not make me feel distant and thanks to you I felt much less the fact of being far from home.

*"And finally we got out to see again the stars" (Dante Alighieri).* Using this sentence, Dante ends his Inferno. Thus, it ends my first academic experience outside Italy. My first experience abroad greatly enriched my cultural background, with the awareness of having worked to the best of my possibilities. But like Dante, immediately after leaving Inferno (Hell), he leaves for Purgatorio (Purgatory), so I am projected towards a new adventure. Like a romantic pirate heading for new unexplored shores, I still do not know what the future holds for me, so I scan the horizon hoping to see a new land and a new experience. Looking back at this PhD times, at the thought of the many things done and the people met, without any hesitation I am going to face new challenges and new possibilities, aware of the fact that if the new experience is only half as good as it was that of the PhD, it will be worth it.

*"Let's learn to say thank you. We teach it to children, but then we forget it!" (Pope Francesco).* I hope I have never forgotten anyone! I also believe I never forgotten to thank people during my PhD experience. But once more, thank you, from the bottom of my heart!

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SUMMARY

Mercury is a strong neurotoxin. It has adverse effects on the nervous and cardiovascular systems, it is also deleterious for several organs, and may act as immunotoxic and hormonal disruptor. This compound is released to the environment by several processes which have generated a widespread contamination throughout the whole planet. However, because of the physical-chemical properties of this metal, semi-volatility and water insolubility, the sink of most of it are the sediments of water bodies, namely marine and lacustrine systems. Bacterial processes transform mercury into methylmercury which is much more deleterious for the organisms and human beings. Methylmercury enters into the food chain and accumulates in fish and from fish into humans. Fish consumption is the main source of mercury for the general population.

The Mediterranean Sea is a hot spot of mercury pollution, but the origin of this metal has not been elucidated. The present PhD dissertation is devoted assessing what is the origin of this metal and which human pollution burden is involved in fish consumption.

To assess the impact of a chlor-alkali plant on a confined environment, fish, sediment and a mammal were collected downstream from the location of a chlor-alkali plant that used a technology that directly releases mercury. Mercury and other metals were examined in the area surrounding the Ebro River and in the marine area located near the mouth of this river. The study has shown that the effects of metal contamination are visible nearby the industrial complex, downriver and in the fish of the Mediterranean area located nearby the Ebro mouth, more than 110 kilometres downriver. This study has also allowed to characterize the specific mercury isotopic composition of chlor-alkali plants.

Subsequently, the study of the fish devoted to human consumption in L'Ampolla, Ametlla de Mar, Alacant, Mallorca, Menorca, Eivissa, Marseille, Genoa, Civitavecchia and Alghero has allowed the contribution characterization of the atmospheric deposition plus the background level and chlor-alkali plants in the fish from Western Mediterranean. The inputs from the chlor-alkali plants represented about 63-100% of total mercury while the inputs from background + atmospheric fallout ranged between 0% and 37%.

The study of 1345 specimens of commercial value sold for human consumption in the above-mentioned locations allowed the characterization of the mercury

concentrations in 58 species of lean and cartilaginous fish. 316 samples (23.5% of the total) showed Hg concentrations above the EU recommended limits for human consumption, 0.5 mg $kg^{-1}$ wet weight (ww) or 1 mg $kg^{-1}$ ww.

The extrapolation of the mercury concentrations observed in Mediterranean fish to the tolerable provisional weekly intakes of methylmercury showed intakes above the thresholds recommended by EFSA for the three countries where the fish was collected, 152%, 151% and 144% for the populations of Spain, France and Italy, respectively.

Comparison of the mercury content in the different species allowed to define one group of twelve species whose specimens always fulfilled the EU recommended values for human consumption: sardine, anchovie, squid, surmullet, painted comber, blackspot seabream, blue whiting, salema, brown meagre, picarel, pearly razorfish and common dolphinfish. On the contrary, the species showing a high percentage of individuals not fulfilling the EU recommendations were dusky grouper, european barracuda, common dentex, norway lobster, greater forkbeard, common seabream, porbeagle and thornback ray.

# RESUM

El mercuri és una neurotoxina forta. Té efectes adversos sobre els sistemes nerviós i cardiovascular, també és perjudicial per a diversos òrgans i pot actuar com a disruptor immunotòxic i endocrí. Aquest compost s'allibera al medi ambient per diversos processos que han generat una contaminació generalitzada per tot el planeta. No obstant això, a causa de les seves propietats físico-químiques, semivolatilitat i insolubilitat en aigua, l'embornal de la major part d'aquest metall es troba en els sediments de les masses d'aigua, és a dir, els sistemes marins i lacustres. Els processos bacterians transformen el mercuri en metilmercuri que és molt més nociu per als organismes i els éssers humans. El metilmercuri entra a la cadena alimentària i s'acumula en els peixos i dels peixos als humans. El consum de peix és la principal font de mercuri per a la població en general.

La mar Mediterrània és un punt calent de contaminació per mercuri, però l'origen d'aquest metall en aquest mar està pendent d'esbrinar. La present tesi doctoral es dedica a avaluar quin és l'origen d'aquest metall i quina càrrega de contaminació humana implica el consum de peix.

Per avaluar l'impacte d'una planta de clor-àlcali en un entorn confinat, es van recollir peixos, sediments i un mamífer aigües avall de la ubicació d'una aquestes plantes que utilitzava una tecnologia que alliberava directament mercuri. El mercuri i altres metalls es van examinar a l'entorn del riu Ebre i a la zona marina situada prop de la seva desembocadura. L'estudi ha mostrat que els efectes de la contaminació per metalls són visibles a prop del complex industrial, riu avall i en els peixos de la zona mediterrània situats prop de la desembocadura de l'Ebre, a més de 110 quilòmetres riu avall. Aquest estudi també ha permès caracteritzar la composició isotòpica específica del mercuri de les plantes clor-àlcali.

Posteriorment, l'estudi del peix dedicat al consum humà a l'Ampolla, Ametlla de Mar, Alacant, Mallorca, Menorca, Eivissa, Marsella, Gènova, Civitavecchia i l'Alguer ha permès caracteritzar les aportacions de la deposició atmosfèrica conjuntament amb el nivell de fons i de les plantes cloràlcali en els peixos de la Mediterrània occidental. Els inputs de les plantes de clor-àlcali representen al voltant del 63-100% del mercuri total, mentre que els del nivell de fons + deposició atmosfèrica oscil·len entre el 0% i el 37%.

L'estudi de 1.345 exemplars de valor comercial venuts per al consum humà a les localitzacions esmentades anteriorment va permetre caracteritzar les concentracions de

mercuri en 58 espècies de peix blanc i cartilaginós. 316 mostres (23,5% del total) tenien concentracions de Hg per sobre dels límits recomanats de la UE per al consum humà, 0.5 mg $kg^{-1}$ ww o 1 mg $kg^{-1}$ ww.

L'extrapolació de les concentracions de mercuri observades en el peix mediterrani a les ingestes setmanals provisionals tolerables de metilmercuri va mostrar ingestes per sobre dels llindars recomanats per l'EFSA per als tres països on es va recollir el peix, 152%, 151% i 144% per a les poblacions d'Espanya, França i Itàlia, respectivament.

La comparació del contingut de mercuri en les diferents espècies va permetre definir un grup de dotze en què els exemplars sempre complien amb els valors recomanats per la Unió Europea per al consum humà de la UE: sardina, aladroc (seitò), calamar, moll de roca, vaca (serrà), goràs, llúcera (maire), salpa, càntera, gerret, raor i llampuga. Per contra, les espècies que van mostrar un percentatge elevat d'individus que no complien les recomanacions de la UE van ser l'anfós (nero), espet, déntol, escamarlà, mòllera de fang, pagre, marraix i clavellada (rajada).

ABBREVIATIONS AND ACRONYMS

| | |
|---|---|
| AESAN | Agencìa Española de Seguridad Alimentaria |
| AL | Alacant |
| ALG | Alghero |
| AMA | Advanced Mercury Analyzer |
| BI | Balearic Islands |
| bw | Body Weight |
| CI | Civitavecchia |
| DMHg | Dimethylmercury |
| ED | Ebro Delta littoral |
| EEA | European Environment Agency |
| EFSA | European Food Safety Authority |
| EPA | Environmental Protection Agency |
| ERM | European Reference Material |
| EU | European Union |
| EUMOFA | European Market Observatory for Fisheries and Aquaculture Products |
| EWG | Environmental Working Group |
| EWI | Estimated Weekly Intake |
| FAO | Food and Agriculture Organization |
| FDA | Food and Drug Administration |
| GSH | Glutathione |
| ha | Hectares |
| IARC | International Agency for Research on Cancer |
| ICP-AES | Inductively Coupled Plasma-Atomic Emission Spectroscopy |
| ICP-MS | Inductivelt Coupled Plasma-Mass Spectroscopy |
| ID | Identity |
| ISMEA | Institute of Service for the Agricultural Food Market (Italian Institution) |
| KMO | Kaiser-Meyer-Olkin |
| L | Length |

| | |
|---|---|
| M | Marseille |
| Max | Maximum |
| MC-ICP-MS | Multi Collector-Inductively Coupled Plasma-Mass Spectrometer |
| MDF | Mass-Dependent Fractionation |
| MeHg | Monomethylmercury |
| MESS-3 | Marine Sediment Reference Material for Trace Metals |
| MIE | Magnetic Isotope Effect |
| MIF | Mass-Independent Fractionation |
| Min | Minimum |
| N | Number |
| NIH | National Institute of Health |
| NIST | National Institute of Standards and Technology |
| NIVA | Norwegian Institute for Water Research (NIWA) |
| NVE | Nuclear Volume Effect |
| OC | Organochlorine compounds |
| OJEU | Official Journal of the European Union |
| P | P-Value, Probability Value |
| PACS-2 | Marine Sediment Reference Material for Trace Metals |
| PC | Principal Component |
| PCA | Principal Component Analysis |
| ppb | Part per billion |
| PTWI | Provisional Tolerable Weekly Intake |
| $R^2$ | Coefficient of Determination |
| rev. | Revolutions |
| SI | Similarity Index |
| SRM | Standard Reference Material |
| T-Hg | Total mercury |
| TOC | Total organic matter |
| UNEP | United Nation Environment Programme |
| UV | Ultraviolet |

| | |
|---|---|
| W | Weight |
| WHO | World Health Organization |
| ww | Wet weight |
| yr | Year |

# OBJECTIVES AND THESIS STRUCTURE

Mercury is a powerful pollutant released in high quantities from both industrial and natural sources. It is found in all environmental compartments, but it tends to accumulate in the aquatic systems because of its physical-chemical properties. The primary source of mercury in humans is the consumption of fish and seafood, which contains a consistent amount of mercury. The greatest concern with mercury uptake in humans is due to adverse neuronal, endocrinal and immunological effects it causes. Despite the information available, several gaps need to be filled in terms of assessing the extent of the effects of this neuro-pollutant, predicting the contribution of the sources that release it and considering the situation of mercury in a context such as that of the western Mediterranean because it is found in high abundance in this marine environment. Several steps have been addressed in the present PhD study to fulfill these gaps. They can be summarized according to the objectives and sub-objectives listed as follows:

- Description of the distribution of mercury in a highly contaminated setting because of the release of mercury from a chlor-alkali plant using a technology that produces mercury-rich wastes.
- Evaluation of the extent of diffusion of mercury effluents from chlor-alkali plant discharges.
- Assessment whether these industrial mercury spills can be transported downriver along a stretch of river water (specifically the Ebro river) and affect the coasts of the Mediterranean Sea, where its mouth is located.
- Characterization of the mercury isotopic composition in different types of samples contaminated by the emissions of chlor-alkali plants and tracing the geographical extent of the contamination using the mercury isotope signatures of different contaminated media.
- Tracing the origin of mercury in fish samples in the Western Mediterranean Sea using the mercury isotopic composition. That is, discriminating between different sources, and, particularly, between chlor-alkali sources and atmospheric deposition plus background.
- Development of a method for estimation of the contribution of mercury from different sources, which may be useful for designing strategies of reduction of the impacts of mercury.

- Description of the overall mercury load in the western Mediterranean fish.
- Calculation of the tolerable provisional weekly intake estimates of methylmercury in the populations of Spain, France and Italy. These estimates provide preliminary information for a future assessment of the health risks associated with local fish consumption.
- Assessment of the fish and seafood species from the Mediterranean Sea that are risky or safe for human consumption.
- Elaboration of a specific mercury assessment of the local fish species for each Western Mediterranean area, to identify the species that have a high level of mercury in a given environment and, eventually, implementing adequate measures to lower the amount of mercury absorbed by the population.

To achieve these objectives, this PhD dissertation is structured in five chapters.

- Chapter 1 introduces the state of the art on the analysis of mercury. The general information about mercury, its presence in different chemical forms and environments and its implications on human health are described. Subsequently, the isotopic signatures of mercury are described: what they are, in which samples they are analyzed and their uses. The structure of the study with the intended objectives and a scheme of the locations where the samples were collected are reported.
- Chapter 2 describes the distribution of mercury and metals in sediments and biota in an environment under the influence of a chlor-alkali plant, as it is the Catalan Ebro river stretch. This chapter is concerned with the evaluation of the impact of such installation in a water reservoir and a river section in terms of the inputs of mercury and other metals concentrations. To achieve this, samples of fish, sediment and a mammal were collected downstream from the location of a chlor-alkali plant that used mercury in the cathode. This metal and others were analyzed starting from the Sebes reservoir down to the sea to assess the extent to which the waste spills of this installation were observed. Another

important aspect covered by this chapter is the characterization of the mercury isotopic composition in the diverse types of samples contaminated by the chlor-alkali plant emissions.

- Despite the large number of studies on mercury and its isotopic composition in fish, no one has found a specific answer for the origin of the mercury found in the Mediterranean Sea. Chapter 3 has been designed to solve this problem. Primarily, it describes the predominant mercury sources in fish from the Mediterranean Sea. Knowing how individual sources contribute to fish samples may provide unvaluable data to design adequate remediation strategies. The proposed methodology, involving the study of the mercury isotopic signature in two specific fish species distributed throughout the western Mediterranean area will provide the necessary knowledge when compared with global scale isotopic effects related with insolation. Specifically, the proposed approach will afford to distinguish between the contributions of chlor-alkali plants and atmospheric deposition plus background. Accordingly, total mercury levels and isotopic composition of mercury were examined in fish recovered throughout the western Mediterranean. The results provided information on the main mercury sources of the samples and the mercury concentrations that could be attributed to them.
- The results obtained from the isotopic signatures in chapter three have made it possible to trace the contribution of the sources acting on the samples, while the values of the mercury concentrations in chapter 4 have described the mercury levels present in the Mediterranean area. The beneficial effects of fish consumption are known, but the deleterious effects that mercury may have on human health should be monitored. Chapter 4 firstly describes the overall mercury amounts in the western Mediterranean fish and seafood. To achieve this approach, the analysis of total mercury in 1345 specimens of commercial value have been performed which has provided an appraisal of the health significance of this consumption. Fish and seafood specimens devoted to human consumption in L'Ampolla, Ametlla de Mar, Alacant, Mallorca, Menorca, Eivissa, Marseille, Genoa, Civitavecchia and Alghero have

been collected and mercury concentrations in 58 different species characterized. This information combined with the eating habits of the populations in which the fish was collected (Spain, France and Italy) have allowed us to evaluate the tolerable provisional weekly intake estimates of methylmercury for the different countries. Despite the wide variety of fish species consumed in the Mediterranean area, no previous study has assessed which species can be considered entirely safe for human consumption. Chapter four is also devoted to suggest which fish species in the Mediterranean Sea are risky and safe for human consumption. Consuming the species considered safe for human health could minimize the risks due to the intake of mercury in the diet. The database generated has also provided a detailed description of the mercury concentration of the local fish and seafood species for each area.

- Chapter 5 reports the general conclusions of the results of the previous chapters.
- Two appendices are included which describe the names of the fish and seafood species covered in the study in English, Catalan and Italian and all information regarding the samples considered in the present study, including mercury concentrations.

# CHAPTER 1

GENERAL INTRODUCTION

# CHAPTER 1

# GENERAL INTRODUCTION

## 1.1 Mercury: general information

Mercury is a silvery-colored element. It is represented by the acronym Hg, from its ancient Greek name "Hydrargyrum", formed by ὕδωρ, (iudor, water) and ἄργυρος (árgyros, silver). It is found in liquid form at ambient temperature and pressure conditions. The atomic number is 80 and the atomic mass is 200.59 g $mol^{-1}$. Mercury owns seven stable isotopes in nature $^{196}Hg$ (0.16%), $^{198}Hg$ (10.04%), $^{199}Hg$ (16.94%), $^{200}Hg$ (23.14%), $^{201}Hg$ (13.17%), $^{202}Hg$ (29.73%), $^{204}Hg$ (6.83%).

Mercury is ubiquitous. It can be present in soils, oceans, seas, lakes, rivers, and the atmosphere. The chemical forms in which mercury can be found are elemental mercury ($Hg^0$), divalent inorganic form ($Hg^{2+}$ or IHg), organic forms which include monomethylmercury (MeHg or $CH_3Hg$), and dimethylmercury (DMHg or $(CH_3)_2Hg$) (Clarkson & Magos, 2006).

Hg is a persistent pollutant. A molecule of Hg can remain in an environment for a long time, due to its high half-life (about 3000 years). The effects caused by mercury uptake on human health are very harmful. In fact, Hg is a strong neuropollutant, affecting the nervous system, but it could also provoke adverse effects on kidneys, lungs, and the cardiovascular system (Ha et al., 2017; Genchi et al., 2017; Park & Zheng, 2012; Karagas et al., 2012; Bernhoft, 2012; Grandjean & Herz, 2011; Bose-O'Reilly et al., 2010; Figure 1.1). Furthermore, Hg can cause problems at the immunological level and in the endocrine system (Gardner & Nyland, 2016; Hyman, 2004; Zhu et al., 2000). Humans are not the only ones suffering from mercury toxicity. There are animals, such as mammals, birds, seabirds, and fish, from which side effects due to Hg ingestion have been reported (Bridges et al., 2016; Tartu et al., 2013; Rutkiewicz et al., 2011; Shore et al., 2011; Wolfe et al., 1998). Its highly deleterious health potential is owing to its capability in binding biomolecules (Abbott & Nigussie, 2021; Weiyue et al., 2011).

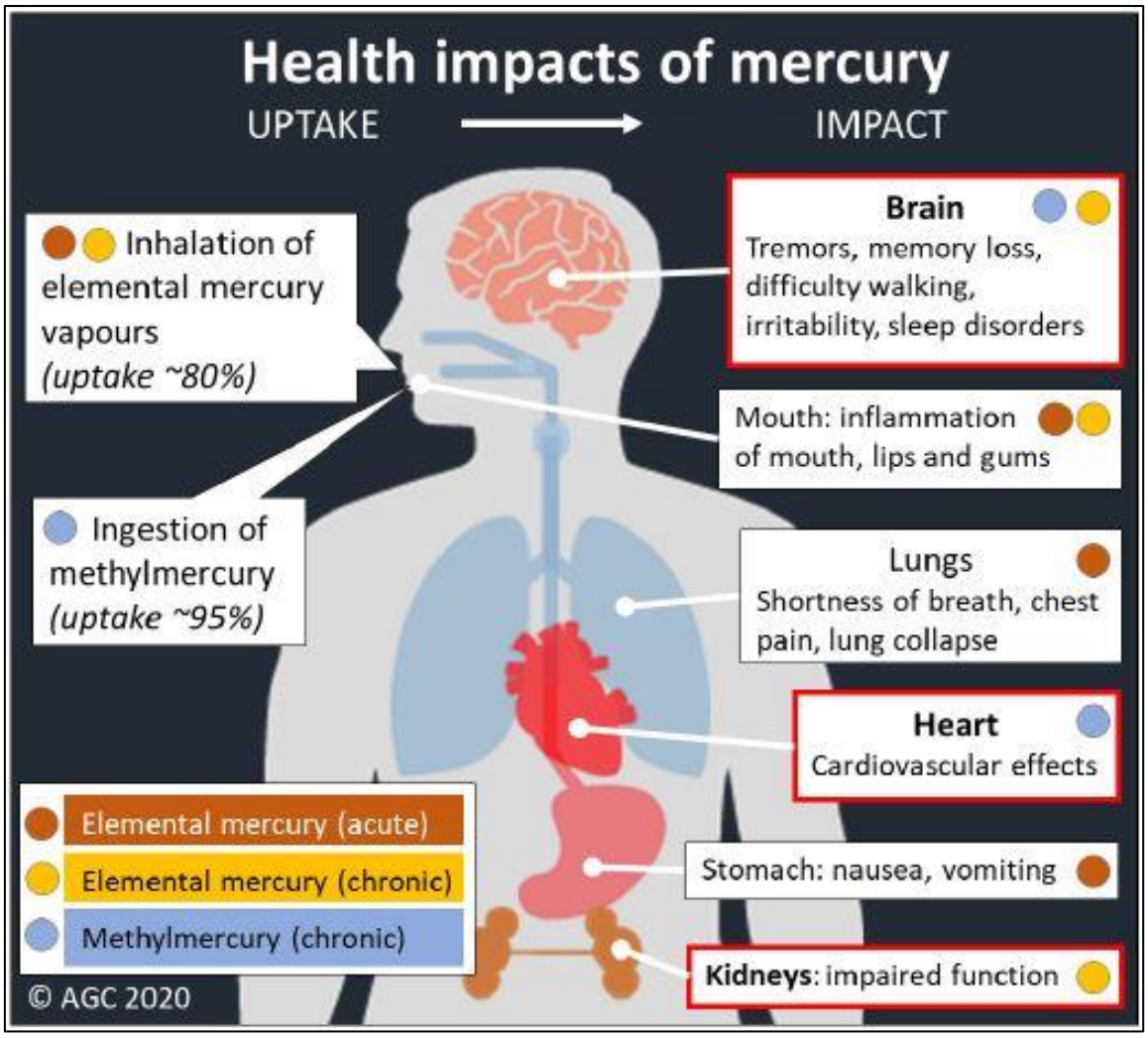


**Figure 1.1.** Major health impacts of mercury intoxication. Image adapted from Artisanal Gold Council (AGC), 2020.

The first major episode due to mercury poisoning occurred in Minamata, Kumamoto prefecture, Japan. There, the release of mercury-rich waste into the wastewater by an industry producing acetaldehyde from 1932 to 1968 caused a disaster, leading to the deaths of 1784 people and causing neurological problems to approximately 2 million people. The scale of this tragedy was so devastating that episodes of mercury poisoning took the name of Minamata disease. The main symptoms of this disease include ataxia, paraesthesia in the hands and feet, general weakness of the muscles, weakening of the visual field, hearing damage, and difficulty in articulating words (Figure 1.2). In extreme cases, it led to mental disorder, paralysis, coma, and death within a few weeks of the first symptoms. A congenital form of the disease could be transmitted to the foetus during pregnancy.

**Figure 1.2.** People with Minamata disease.

Since that time, many other episodes of lesser scope than Minamata related to mercury toxicity have occurred in Japan (Maruyama et al., 2012) and elsewhere (Bonsignore et al., 2016; Kim et al., 2016; Garì et al., 2013; Brown, 2001; Maurice-Bourgoin et al., 2000; Andersen et al., 1987), giving relevance to a problem that has not yet been solved and that can lead to catastrophic results if not kept under control.

Mercury can be released from both natural and anthropogenic sources. Although some natural sources, such as volcano activities (Bagnato et al., 2007; Pyle & Mather, 2003; Ferrara et al., 2000), rock erosion (Kwasigroch et al., 2018; Beldowska et al., 2016; Craw et al., 2000), fires (Melendez-Perez et al., 2014; Friedli et al., 2009; Wiedinmyer & Friedli, 2007), and glacierized watersheds (Nagorski et al., 2021) may play a role in the re-mobilization of mercury at a global scale, the main source of mercury emission comes from anthropogenic activities. The main anthropogenic sources that cause the release of mercury are coal combustion (Hu et al., 2020; Yang et al., 2020; Streets et al., 2018), artisanal gold mining (Gyamfi et al., 2021; Gerson et al., 2018; Zolnikov & Ortiz, 2018), chlor-alkali plants emissions (Fantozzi et al., 2021; Navràtil et al., 2021; Biester et al., 2002), cement and steel production (Kogut et al., 2021; Cai et al., 2020; Han et al., 2019; Bhave & Shrestha, 2018; Wu et al., 2017), non-ferrous metal smelting (Liao et al., 2019; Wang et al., 2006), and waste disposal and incineration (Sun et al., 2020; Li et al., 2017). There are several estimates of mercury releases into the atmosphere. Mason and Sheu (2002) computed that 64% originated from anthropogenic sources and 36% from natural sources. The global anthropogenic Hg emission into the atmosphere every year is estimated to be about 2200 tons (Yin et al., 2010; Pacyna et al., 2006). The gaseous released Hg is in the form $Hg^0$, which is oxidized to $Hg^{2+}$ in the atmosphere and transported in all compartments of every environment, reaching the most remote sites of the planet (Morel et al., 1998; Figure 1.3).

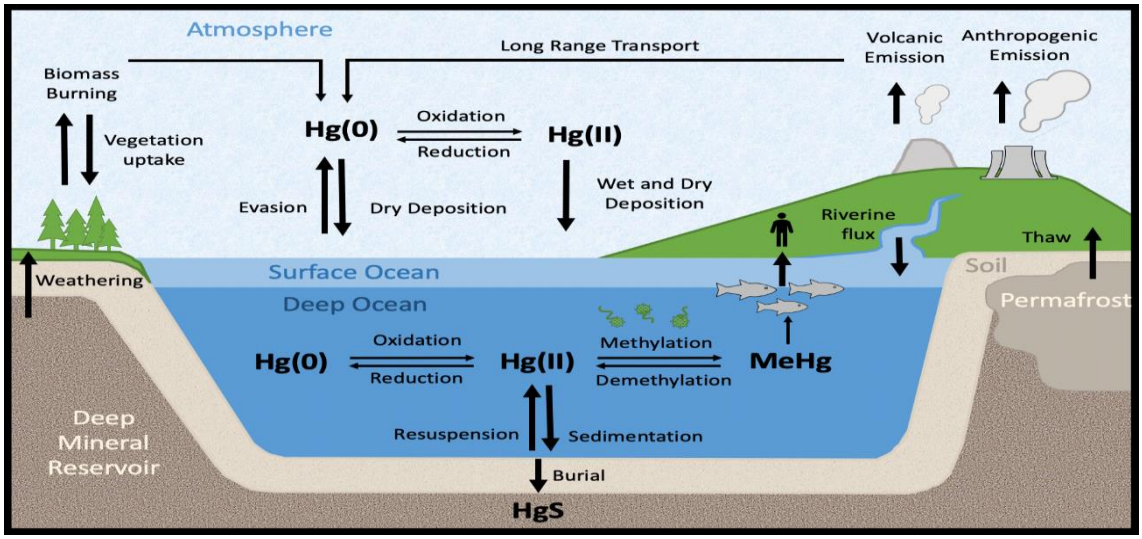


**Figure 1.3.** Mercury cycle. Reprinted from MacKenzieEJewell (2020; https://en.wikipedia.org/wiki/Mercury_cycle#/media/File:PhysicalMercuryCycle.png)

The flows of mercury into the atmosphere from different sources varies. The same also happens for the mercury flows that fall from the atmosphere towards the different environments. These are summarized in Figure 1.4 which provides an estimate of the mercury exchanges between environmental compartments (Mason et al., 2012).

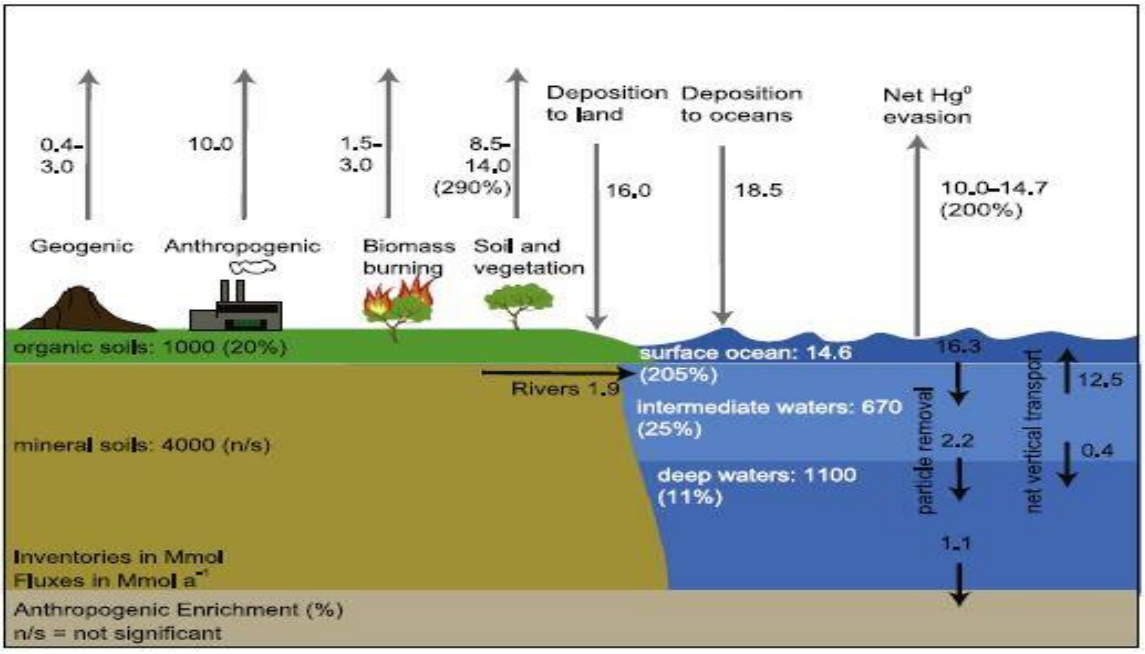


**Figure 1.4.** Estimation of mercury fluxes at the Earth's surface based on a simulation of global mercury emission model. The percentage values in brackets are the estimated increases in concentration and fluxes in the last century due to anthropogenic activities. Fluxes are in Mmol year$^{-1}$. Image adapted from Mason et al., 2012.

## 1.2 Mercury in the aquatic environment

In the aquatic environment, a molecule of $Hg^{2+}$ deposited from the atmosphere undergoes a series of processes (Harris et al., 2007). Microorganisms from water and sediment operate a series of oxidation-reduction, methylation-demethylation transformations, and photochemical reactions (Perrot et al., 2013; Celo et al., 2006; Barkay et al., 2003; Amyot et al., 1994). The microorganisms that transform $Hg^{2+}$ into MeHg are anaerobic bacteria. The bacteria that perform these reactions are sulphate-reducing bacteria, iron-reducing bacteria, methanogenic archaea, and the phylum *firmicutes* (Gilmour et al., 2013; Yu et al., 2013). The *hgcA* and *hgcB* are the genes responsible of the production of MeHg. The main mechanism that converts the $Hg^{2+}$ into MeHg requires the transfer of a methyl group from a molecule of methylcobalamin, as follow:

$$CH_3CoB_{12} + H_2O + Hg^{2+} \rightarrow CH_3Hg^{+} + H_2OCoB_{12}$$

The amount of MeHg in an environment is affected by several variables, such as temperature, pH, redox conditions, the type and the number of methylator bacteria, dissolved organic species and sulphates availability.

The MeHg quantity in a single environment is balanced by the demethylation processes, accomplished by ultraviolet radiation and microbes (Lehnherr & St. Louis, 2009; Marvin-Dipasquale & Oremland, 1998; Figure 1.5).

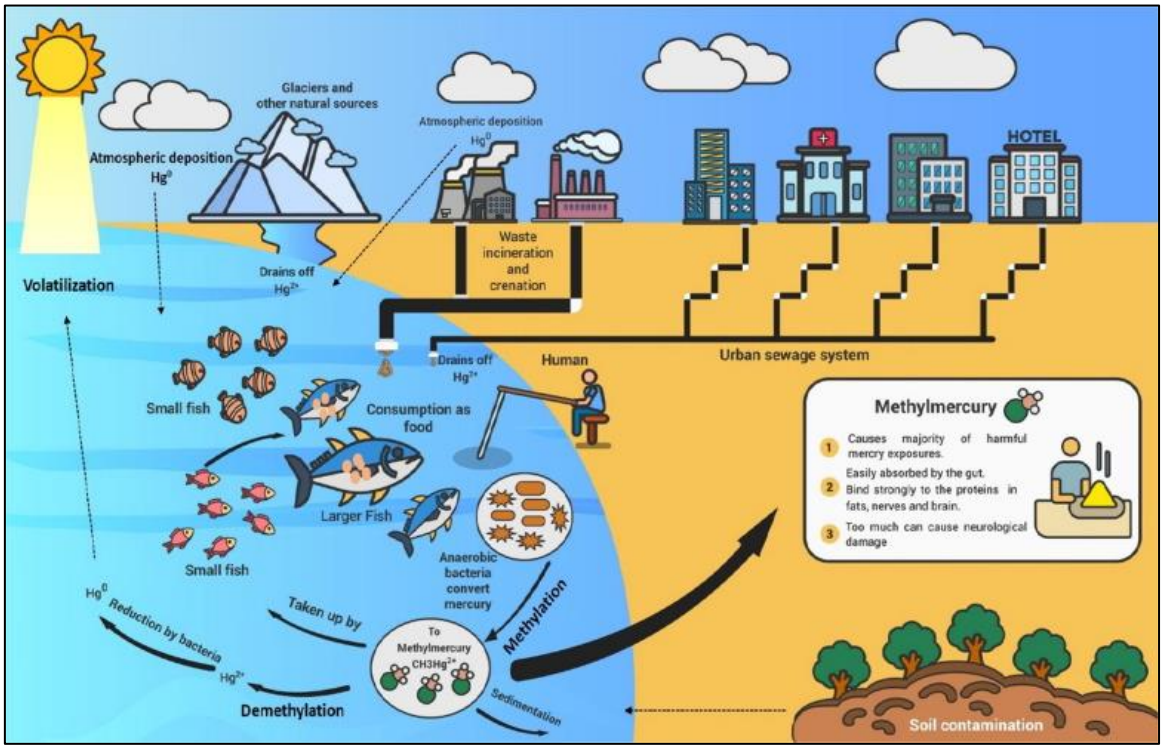


**Figure 1.5.** Transformation of Hg (methylation-demethylation) in the aquatic environment. Image reprinted from Duan et al., 2020.

The MeHg is lipophilic. It enters the vascular system of the organism, is transported through the blood, and binds to organs and tissues throughout the body. Almost the totality of the mercury present in an organism belongs to the MeHg form (Salazar-Camacho et al., 2021; Lescord et al., 2018; Perrot et al., 2012).

Due to its adverse effects on human health, mercury is considered a pollutant of global concern. The organometallic forms are the most harmful, having MeHg as the main actor, which can move through the food web. This form of mercury can be accumulated

during the entire life of a living being, due to bioaccumulation, and can be transferred from one organism to another, in that process called biomagnification.

The main neurotoxic effects due to mercury exposure depend on the age of the exposed subjects. Children and fetuses of pregnant women are those who suffer the most (Reuben et al., 2020; Santos-Lima et al., 2020; Stratakis et al., 2020; Gonzalez et al., 2019; Gump et al., 2017). However, adults are not free from adverse effects and episodes of cognitive function impairment have been reported (Bernhoft, 2012; Grandjean & Herz, 2011; Bose-O'Reilly et al., 2010). In animals, mercury intake leads to reduced fertility, reduced breeding frequency, impaired development of embryos, changes in behavior, negative effects on blood chemistry, brain damage (Tartu et al., 2013; Rutkiewicz et al., 2011). Fish also suffer mercury toxicity which is reflected in different hatching times and decreased survival rates of offspring (Bridges et al., 2016).

The most prevalent mercury exposure for humans is due to the consumption of fish (Junquè et al., 2017; Garì et al., 2013). Maternal age, smoking during pregnancy, gestational time, breastfeeding, place of residence, parity, passive smoking at 4 years, maternal and paternal occupation and educational level, child's sex, fish, and other seafood consumption are the factors considered when investigating the main socio-economic characteristics influencing mercury uptake. A significant positive correlation has been reported between the level of mercury and the consumption of fish and seafood (Garì et al., 2013; Figure 1.6).

In 2017, FAO estimated that the per capita world consumption of fish was 20.3 kg. This consumption was showing an upward trend (FAO, 2020; Figure 1.7).

Muscles are the edible part of a fish specimen and are considered critical sites of Hg bio-accumulation due to the high storage capacity and low purification rates. Fish muscles are also used to detect contaminant concentration (Copat et al., 2012) and play a critical role in assessing the risk of metal exposure, including Hg, in humans (La Colla et al., 2021; Sabo et al., 2021; Milanov et al., 2016).

The problem of mercury poisoning is certainly not something out of date. In fact, worrying levels of mercury have been found in the hair of children. A substantial part of the samples from the Menorca cohort shows a quantity of mercury above the limits imposed by the World Health Organization threshold value, set at 2.0 $\mu g\ g^{-1}$ (Garì et al., 2013; Figure 1.7).

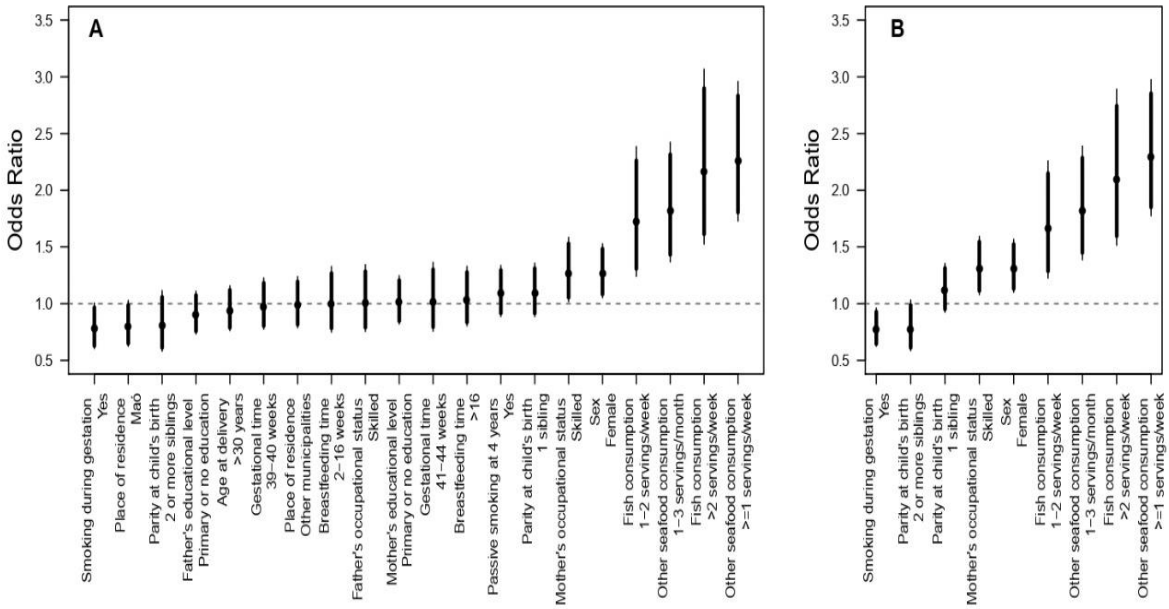


**Figure 1.6.** Main socio-economic factors of mercury accumulation in four-year-old children from Menorca. Bars represent the 90% and 95% confidence intervals, shown in thick and thin lines, respectively. A) Model adjusted for maternal age, smoking during pregnancy, gestational time, breastfeeding, place of residence, parity, passive smoking at 4 years, maternal and paternal occupation and educational level, child's sex, fish, and other seafood consumption. B) Model with the statistically significant covariates after application of the stepwise algorithm. Reprinted from Garì et al., 2013.

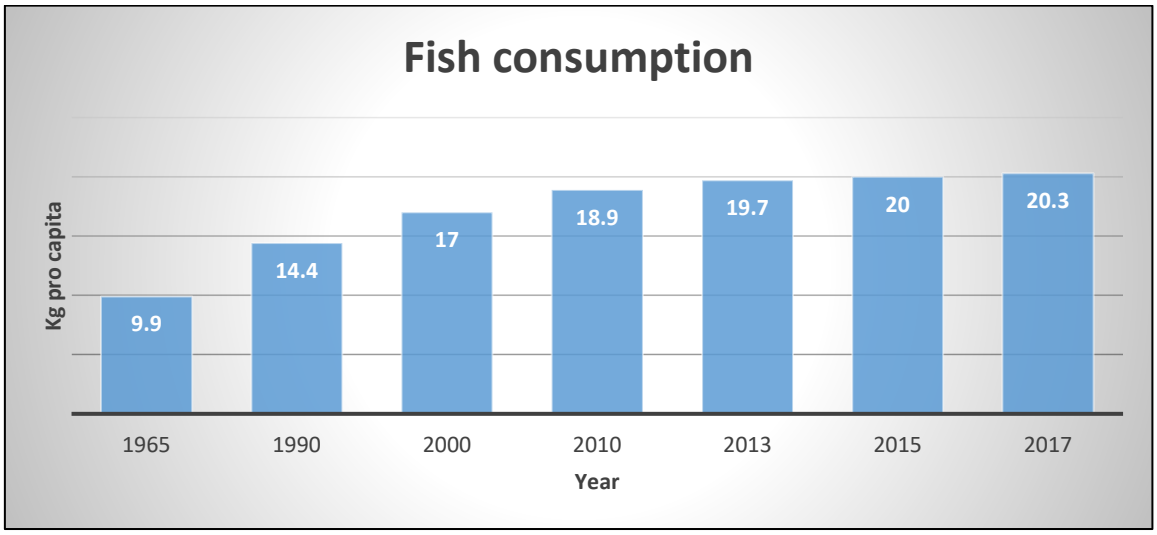


**Figure 1.7.** Fish consumption trend (in Kg per capita).

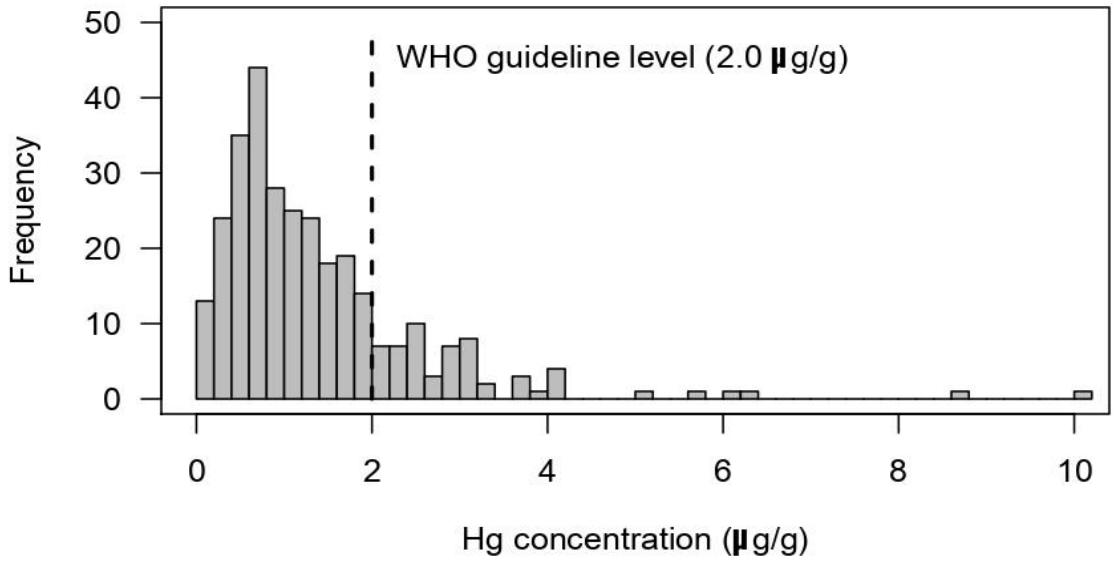


**Figure 1.8.** Mercury concentration in hair of four-year-old children from Menorca. The WHO guidelines level is considered as a reference. All samples to the right of the dotted line have mercury levels above the cutoff limit. Reprinted from Garì et al., 2013.

## 1.3 Stable isotope composition

### 1.3.1 Isotopic fractionation

Various nature processes are involved in the fractionation of isotopes. This fractionation depends on the relative mass difference among isotopes and on element characteristics that respond to physical, biological, and chemical reactions (Epov et al., 2012). The fractionation of mass-dependent and mass-independent isotopes have been considered effective tools for tracing the sources of mercury emission (Cransveld et al., 2017; Yin et al., 2014; Gehrke et al., 2011; Bergquist & Blum, 2007).

The isotope composition of many elements, including mercury, is analyzed using a Multicollector-Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS). This instrument can measure the isotopic ratio with high accuracy, even with a low amount of sample in different matrices. The high sensitivity, selectivity, and high ionization efficiency are peculiar characteristics of the instrument.

### 1.3.2 Mercury isotopic fractionation

Mercury has seven stable isotopes ($^{196}Hg$, $^{198}Hg$, $^{199}Hg$, $^{200}Hg$, $^{201}Hg$, $^{202}Hg$, $^{204}Hg$). These isotopes can be subjected to two types of isotopic fractionation: mass-dependent fractionation (MDF) and mass-independent fractionation (MIF). The MDF values are reported as δ, while the MIF values are described with Δ. The unit of measure for both are units per mil (‰).

#### 1.3.2.1 Mass-Dependent fractionation (MDF)

The isotope fractionation is based on isotope zero-point energies which are derived from molecular vibrational frequencies. Differences in energy lead to isotope fractionation. The strength of the bond is inversely proportional to the number of zero-point energies. A molecule that has lower zero-point energies is more stable because the bond is less easily broken. Heavier isotopes have a stronger bond because they have the lowest zero-point energies.

The zero-point energies are also the protagonists for the different activation energies in the light and heavy isotopes. Thus, the activation energy of a molecule containing the heavy isotope is greater, due to lower zero-point energies and the opposite occurs for molecules having the light isotope.

In chemical reactions, lighter isotopes react faster than heavier isotopes. This kinetic phenomenon has a mass-dependent nature, leading to the formation of MDF. Some processes that act on MDF are metabolic transformations and diffusion, among others.

The MDF was defined as the following equation (Blum and Bergquist, 2007):

$$\delta^{x}Hg\ (‰) = [(^{x}Hg / ^{198}Hg)_{sample} / (^{x}Hg / ^{198}Hg)_{NIST3133} - 1] \times 1000 \quad \text{(Equation 1.1)}$$

where x = 199, 200, 201, and 202, depending on the mass of the isotope under analysis, NIST3133 is the reference material commonly used.

In the literature, the $\delta^{202}Hg$ is the mostly discussed MDF form.

### 1.3.2.2 Mass-Independent Fractionation (MIF)

Sometimes there is no relationship between the fractionation range and the mass difference between the isotopes. So, some isotopes do not follow the MDF kinetics. This phenomenon also happens for mercury, which in addition to MDF, also has MIF.

Mercury MIF is the deviation of the measured isotope ratio from the theoretical ratio predicted in MDFs and they are following the equation:

$$\Delta^{x}Hg\ (‰) = \delta^{x}Hg - (\beta \times \delta^{202}Hg) \qquad \text{(Equation 1.2)}$$

where β is a theoretical mass-dependent scaling factor of 0.252, 0.5024 and 0.752 for $^{199}Hg$, $^{200}Hg$ and $^{201}Hg$, respectively (Blum & Bergquist, 2007) and x is the mass of the isotope.

Two mechanisms are responsible for MIF, the magnetic isotope effect (MIE) and the nuclear volume effect (NVE). Most naturally occurring MIFs are due to MIE. This is an effect owing to light exposure and it is caused by direct or secondary photolysis (Bergquist & Blum, 2009). It usually concerns odd isotopes and is related to the reaction rate during spin reactions. The NVE is affected by the nuclear volume and the nuclear charge radius. This radius is not related to the number of neutrons and usually the nuclides with odd neutrons show a smaller size than expected. The resulting different density and shape of the electron cloud between the isotopes implies a different participation in the reactions.

In the literature, MIF has been predominantly described for the odd isotopes $^{199}Hg$ and $^{201}Hg$, but the composition of the $^{200}Hg$ isotope has recently been considered a good tracer for precipitation (Lepak et al., 2018).

### 1.3.3 Mercury stable isotope application

The uses of stable isotopes of mercury in the literature have been manifold. Both MDFs and MIFs played a critical role for various purposes.

A first MDF use was made to evaluate the reaction that involves the reduction of $Hg^{2+}$ to $Hg^{0}$ (Kritee et al., 2007; Wiatrowski & Barkay, 2005). Some studies have looked at MIF resulting from photoreactions in different environments. Photoreduction and photodemethylation enrich odd isotopes due to MIE (Bergquist & Blum, 2009; Bergquist & Blum, 2007).

Others have used the MDF isotopic composition to determine the methylation/demethylation process at the microbial level (Perrot et al., 2013; Rodriguez-Gonzalez et al., 2009). Another work has associated the value of an isotope, $\Delta^{200}$, with a specific source, precipitation (Lepak et al., 2018). Mercury isotopes can also elucidate information about the origin of methylmercury (Janssen et al., 2016; Li et al., 2016).

The isotope signature provides valuable information in biological samples such as fish. Some information has a purely ecological value, such as stress factors and eating habits (Kwon et al., 2013; Senn et al., 2010). The trend in MIF composition has provided information on which fish are benthic, since going down in depth the MIF signatures are lower than on the surface (Blum et al., 2013).

Stable isotopes of mercury have their broadest use in ascertaining sources and evaluating their contribution to samples and environments (Reinfelder & Janssen, 2019; Cransveld et al., 2017; Gehrke et al., 2011). A source has unique MDF and MIF compositions. Hence, mercury stable isotope signatures can reveal the contribution of sources with high specificity.

### 1.3.4 Mercury stable isotopes signatures in different samples

The composition of the stable isotopes of mercury has been studied in different media. To obtain as much information as possible from the analysed samples, both Hg MDF and MIF values have been well-described in the literature. All the samples can be divided into two big macro-sections: samples related to the environment and samples related to anthropogenic activity.

#### 1.3.4.1 Mercury stable isotopes: environmental samples

The isotopic signatures of major geochemical reservoirs that are present in the Earth system belong to four categories: Earth's crust, surface, hydrosphere, and atmosphere (Yin et al., 2014; Figure 1.9).

The registered MDF and MIF signatures contribute to the understanding and quantification of the important source processes in the global mercury cycle. A great variability of MIF is observed in environmental compartments (about 10%), while a lower variation is reported for MDF (about 6%).

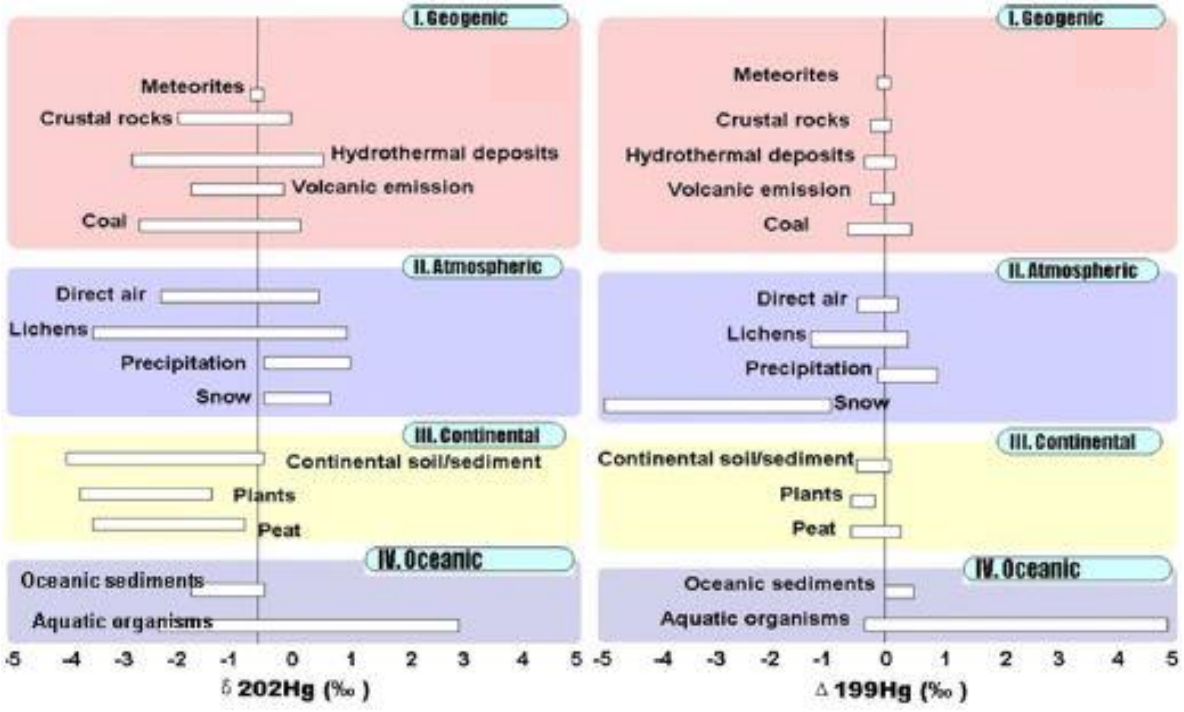


**Figure 1.9.** Overall summary $\delta^{202}$Hg and $\Delta^{199}$Hg values of environmental samples. Image adapted from Yin et al., 2014.

### 1.3.4.2 Mercury stable isotopes: anthropogenic samples

Since the industrial revolution, anthropogenic activities have released important amounts of mercury into the atmosphere (Figure 1.10A).

The simulated total mercury emissions from all sectors between the 1850s to 2010s show an increasing trend for MDF and a stable trend for MIF. The MDF trend reflects a shift of historically dominant Hg emissions encompassing from Hg mining 19th century and liquid Hg uses for Au/Ag refining, coal combustion and non-ferrous metal production in the 20$^{th}$ century (Sun et al., 2016; Figure 1.10B).

These production differences have involved changes in the composition of the mercury isotopes over the years which have been reflected in different MDF and MIF composition (Figure 1.11).

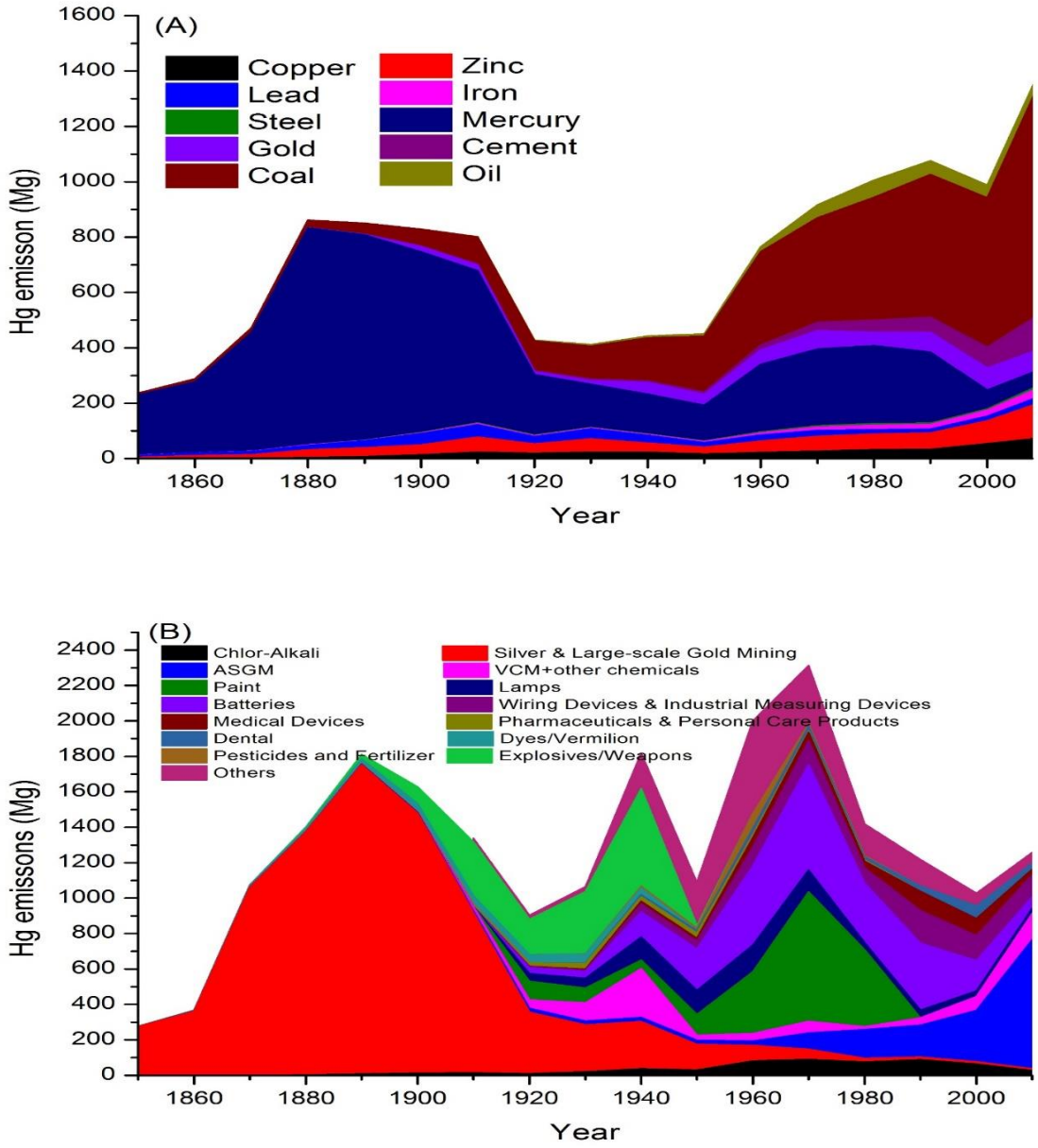


**Figure 1.10.** Historical inventories of metal emissions (A). Total mercury emissions through years by industrial activities (B). Image reprinted from Sun et al., 2016.

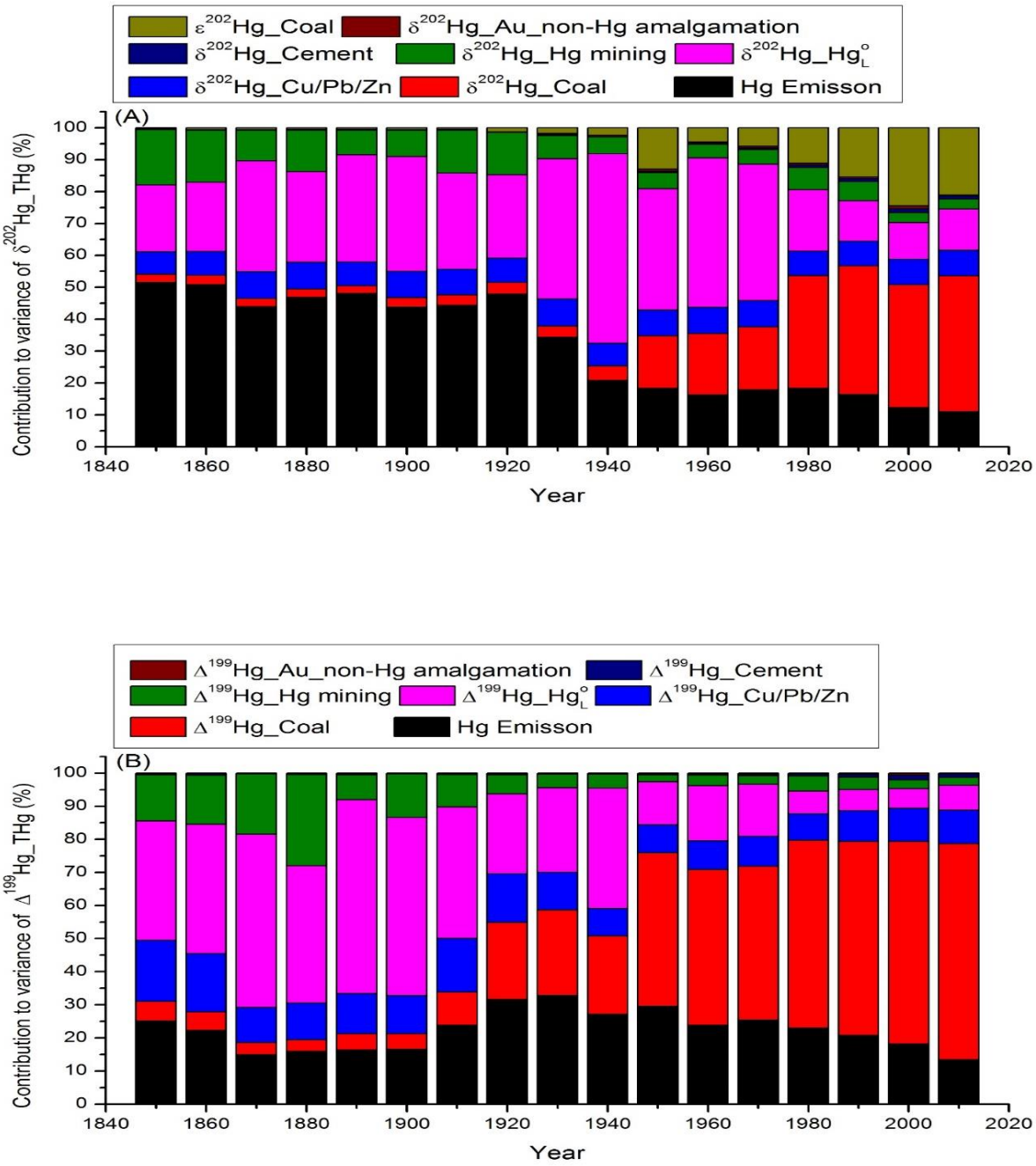


**Figure 1.11.** Historical Hg emission inventory and speciated Hg isotope shifts to the variance of estimated $\delta^{202}$Hg (A) and $\Delta^{199}$Hg (B) of total mercury emitted from all sectors. Image reprinted from Sun et al., 2016.

To date, the source that creates the most emissions is the combustion of coal, followed by the Chlor-alkali plant spills (Sun et al., 2016).

## 1.4 Study framework

The current study is devoted to address several objectives, considering the gaps in the literature that emerge during the work.

Despite the large number of studies on mercury and its isotopic composition in fish, no one has found a single answer for the provenance of mercury.

This is particularly relevant for the Mediterranean Sea where the consumption of fish is significant. In data obtained in Menorca, mercury levels in fish and shellfish caught in nearby areas and consumed on the island are higher than those found in other seas and oceans, with a percentage of around 65% exceeding the maximum limit for human consumption set by the European Union (OJEU, 2006): 0.5 mg/kg or 1 mg/kg for wet weight depending on the species. This means that the intake of this neurotoxic metal by children on the island is more than double the maximum recommendation of the European Food Safety Authority (Junqué et al., 2017; EFSA, 2004).

The higher concentrations of mercury compared to other nearby seas and oceans are characteristic of the Western Mediterranean and also affect the fish consumption of the European population, included Catalonia. In fact, several studies carried out in populations in the Iberian Peninsula and the rest of Europe show that the people in Spain and Portugal (mothers and children) have the highest concentrations of this metal in their hair within the whole European Union (Smolders et al., 2015; Garí et al., 2013; Ramon et al., 2011; Freire et al., 2010).

The current study is carried out in the Mediterranean Sea, a context that can be considered as a reference environment for this type of works, given its characteristics and considering its high percentage of mercury in waters, sediments, and biota (Bonsignore et al., 2020; Cinnirella et al., 2019; Ogrinc et al., 2007; Horvat et al., 2003). Thus, although the Mediterranean is a large area that supplies fish to nearly 500 million people, no risk assessment for those populations that are used to consuming local fish in their diet has been completed. Furthermore, in the plethora of fish species from the Mediterranean that can be eaten, no one has ever assessed which species can be considered completely safe for human consumption. Finally, no one has examined the relative mercury isotopic composition of fish samples from a highly polluted area influenced by an industrial complex to see how much anthropogenic sources affect mercury levels.

Here, both the stable isotope composition of mercury and the total mercury concentration in a number of edible fish samples from the Western Mediterranean are evaluated. The results obtained from the isotopic signatures make it possible to trace the

contribution of the sources acting on the samples, while the values of the mercury concentrations expose the criticality of the mercury levels present in the Mediterranean area.

An overview of the data collected allows to delineate several edible fish species that have critical levels of mercury, but others that are safe for human consumption.

These results are then compared with samples coming from an area highly affected by emissions of mercury-rich waste, showing the variability of results given the fish capture distance from an industrial plant.

### 1.4.1 Sampling scheme

The fish were collected during several sampling campaigns, starting from October 2018 to May 2021. Samples of fresh fish were purchased from local markets in ten different locations throughout the Western part of the Mediterranean Sea. All these fish were intended for human consumption. Only those certified fish that came from the Mediterranean Sea were included in the study. The fish provenance was ascertained by the label imposed by European legislation for the traceability of the catch (European Community regulation n. 1224 / 2009 and European Union regulation n. 404 / 2011). In addition, the fish were caught the day/night before purchase and within a maximum radius of 15-20 km from the coast.

The samples were collected in three countries bordering the Western Mediterranean Sea: Spain, France, and Italy. In Spain, fish samples were purchased in the Balearic Islands, Alacant, L'Ampolla, and Ametlla de Mar. In France, sampling campaigns were organized in Marseille. In Italy, the fish were bought in Genoa, Civitavecchia, and Alghero.

In the Balearic Islands, samples have been recovered on four different occasions in the three largest islands of the archipelago Mallorca, Menorca, and Eivissa. A greater number of samples have been collected in the Balearic Islands thanks to the help of the General Direction of Public Health and Consumption of the Government of the Balearic Islands, which provided around 250 samples. The remainder was recovered in Mallorca, Menorca, and Eivissa markets (Figure 1.12). These islands, in the middle of the Western Mediterranean Sea are far from any industrial inputs, but several cases of people with a high concentration of mercury are reported (Junquè et al., 2017; Garì et al., 2013). A total number of 572 specimens of 41 different species were collected between October 2018 and January 2021.


**Figure 1.12.** Balearic Islands markets.

In Alacant, in the Valencian community, fish were collected in a single campaign (Figure 1.13) from the main fish market, made up of a series of individual fish shops. Alacant, located south of the Iberian Peninsula does not receive any influence from nearby chlor-alkali plants. Here, in March 2019, 197 specimens belonging to 31 different species were collected.

**Figure 1.13.** Alacant fish market.

In L'Ampolla and Ametlla de Mar (Figure 1.14), in Catalonia, there is not a single large fish market, but the fish are sold in local fishmongers. The samples were collected in June 2020. These towns overlook the Mediterranean Sea but are located near the delta of the Ebro river, known for the high quantity of pollutants and mercury produced from chlor-alkali spills and transported downriver by currents (Palanques et al., 2020). In those towns, 99 specimens from 14 species were collected.

**Figure 1.14.** L'Ampolla (on the left) and Ametlla de Mar (on the right).

In France, three sampling campaigns were organized in Marseille, one in October 2018, one in July 2019, and one in December 2019. Fish were collected directly from local fishermen (Figure 1.15). The samples were recovered in the Eastern part of the Lion Gulf, near the mouth of the Rhone River, which receives inputs from chlor-alkali plants. In Marseille, 192 specimens of 23 different species were taken.

**Figures 1.15.** Marseille fish market.

In Civitavecchia, three different sampling campaigns were organized between June 2019 and May 2021. In this city, there is a large fish market, and all the samples were purchased there (Figure 1.16). Civitavecchia is located in the centre of Italy about 200 km from Rosignano Solvay and Volterra, places where there are or have been chlor-alkali plants, and about 130 km from Monte Amiata, site of a mercury mine that was abandoned about 30 years ago, but which continues to release today mercury into the

Tiber River, which then flows into the Tyrrhenian Sea. In total 126 species of 11 different species were collected in Civitavecchia.

**Figure 1.16.** Civitavecchia fish market.

In Genoa, the samples were collected during May and June 2019 in the local fish market, one of the best supplied in North Italy (Figure 1.17). Genoa is located 400 km from Marseille but is not directly affected by any anthropogenic input. In Genoa, 78 specimens belonging to 9 species were obtained.

**Figure 1.17.** Genoa fish market.

In Alghero, the samples were purchased in the two small fish markets in the area (Figure 1.18). Alghero, located on the northwest side of Sardinia, has a clean environment that is not contaminated by industrial sources. In Alghero, 81 specimens of 8 species were purchased from June 2019 to April 2021.

**Figure 1.18.** Alghero fish markets.

In total 1345 specimens belonging to 58 different species throughout the Western Mediterranean were purchased.

Some places have fewer fish species than others. The main causes of this phenomenon are multiple. The seasonality of some fish species is the principal one. Some fish are found in a certain area only for a determinate period of the year. Furthermore, at certain times of the year, it is strictly forbidden to fish using a specific type of fishing, and/or it is not possible to catch certain species of fish due to prohibitions expressed by local authorities. Another cause of the fish species absence in some places is related to the will of the local population who prefer to eat other types of fish. So, some species are not fished and not available in markets. Other species of fish are not caught at certain times because they are at risk of extinction or awaiting restocking. For this reason, the different sampling campaigns were organized in different months all over the year to try to minimize these issues.

### 1.4.2 Study-specific goals

The specific goals are reported as follow:

- *Chapter 2:* Distribution of mercury and metals in sediment and biota of the Catalan Ebro river stretch and nearby marine area.
- *Chapter 3:* Predominant mercury sources in fish from the Mediterranean Sea.
- *Chapter 4:* Overall burden of mercury in fish from the Western Mediterranean. Evaluation of risky and safe fish species for human health concerning mercury concentrations.

# CHAPTER 2

# DISTRIBUTION OF MERCURY AND METALS IN SEDIMENT AND BIOTA OF THE CATALAN EBRO RIVER STRETCH AND NEARBY MARINE AREA

CHAPTER 2

# DISTRIBUTION OF MERCURY AND METALS IN SEDIMENT AND BIOTA OF THE CATALAN EBRO RIVER STRETCH AND NEARBY MARINE AREA

## 2.1. Introduction

Mercury is a persistent pollutant. As mentioned in Chapter 1, it is found everywhere. The sources of mercury emission are numerous, and we divide them into two major sub-categories: natural and anthropogenic. Both can release mercury nearby or into the atmosphere, where it can be transported for miles.

Most of the sources emitting mercury to the atmosphere are natural, involving about 1000–3000 t Hg per year (Lamborg et al., 2002; Seigneur et al., 2001; Mason et al., 1994). Volcanoes and geothermal springs tend to expel mercury (Witt et al., 2008). However, there is a disagreement in their relevance. Some studies indicate that emission fluxes exceed 500 t Hg/yr (Varekamp & Buseck, 1986), while others estimate them to have fluxes below 1.3 t Hg/yr (Pirrone et al., 2001; Ferrara et al., 2000). Pyle and Mather (2003) estimated that the time-averaged volcanic Hg emission is ∼700 t/yr (20–40% of total natural emissions), based on data from active volcanoes. Among the total volcanic activities, 75% of volcanic Hg is released during small eruptions (<10–102 t/event), 15% in explosive eruptions (>103 t/event), and 10% from continuous degassing events (Pyle & Mather, 2003). High concentrations of mercury have been found in ancient populations who lived in remote areas in the vicinity of volcanoes and geothermal springs (Walser et al., 2019). The geothermal activities are associated with extinct volcanoes. Depending on the distinct geothermal system, the emission fluxes are different. In any case, these fluxes are highly uncertain and hard to measure (Gustin, 2003). Some studies have estimated the Hg emission flows of geothermal sources to be less than 60 t/yr (Nriagu & Becker, 2003; Varekamp & Buseck, 1986).

Erosion of rocks that contain mercury involves the release of this metal. When the rocks are eroded, all the metals are scattered. The atmospheric agents transport these components enriching soils and sediments in the vicinity, but a part is moved to greater distances going into the atmosphere (Garrett, 2000). In New Zealand, for example, there are several mineralized rocks composed mostly of marcasite, pyrite ($FeS_2$), and cinnabar

(HgS). The erosion of these rocks leads to the release of cinnabar mercury in the near environment and into the atmosphere (Craw, 2005).

A small constant percentage of mercury released into the atmosphere is provided by water and soil surfaces that are exchanged owing to the concentration gradient of mercury between the top-water and top-soil layer and air above the surface (Hedgecock et al., 2006; Wang et al., 2006). Mercury evasion estimates from great water basins, such as ocean and lakes register a total flow of 2778 t/yr to the atmosphere (Pirrone et al., 2009). The estimated contribution of the Mediterranean Sea for mercury release was about 70 t/yr (Hedgecock et al., 2006). The estimates of mercury released fluxes from soils are diverse (Nacht et al., 2004).

Almost all Hg present in the vegetation is due to exchange with the atmosphere (Ericksen & Gustin, 2004) and atmospheric deposition to foliage and/or root uptake (Rea et al., 2002). Hg is accumulated in roots, rhizomes, leaves, sap flow (Schwesig & Krebs, 2003). The vegetation evasion fluxes are about 1664 t Hg/yr (Pirrone et al., 2009).

Fires release substantial amounts of mercury from the burned material to the atmosphere. Biomass burning is releasing nearly 675 t every year (Friedli et al., 2009).

Another natural source is glacial ice. During melt episodes, there is a direct Hg release into the water or on soils. The mercury is stored in the glacier ice through the years (Ferrario et al., 2017; Beal et al., 2015; Sharma et al., 2015). Also, during the glaciers ice retreats huge amounts of pollutants, including Hg, are released into the atmosphere (Nagorski et al., 2021).

The world mercury contributions from anthropogenic sources into the atmosphere ranged between 1660 and 2320 t/year (Pirrone et al., 2010; Pacyna et al., 2006; Pirrone et al., 1996).

The primary source of mercury released into the atmosphere is the combustion of fossil fuels, but cement and steel production, non-ferrous metal smelting, waste disposal, and incineration, chemical manufacturing also play a great role. Furthermore, we must not forget the direct release of mercury in wastewater, carried out by chlor-alkali plant activities, artisanal gold mining, and mercury mines.

Fossil fuel burning releases into the atmosphere an amount of about 810 t Hg/yr (Pirrone et al., 2010). The most used fuel is coal. The US - Energy Information

Administration estimates coal consumption for 2019 to be 8639 t because mercury concentration in coal was estimated to vary between 0.01 and 1.5 g per t (Pirrone et al., 2009; Mukherjee et al., 2008; Toole-O'Neil et al., 1999). Fuel oil is a minor Hg source, since it contains this metal in a very small amount (normally 3.5 g/t; Wilhelm, 2001), representing 0.015% of the total anthropogenic emission. Natural gas may contain small amounts of mercury that is usually removed by the preparation process but there were cases in which Hg had been found in traces. In any case, it is not a relevant mercury source (Pirrone et al., 1996). In total, the total flow from fuel combustion is estimated to be about 810 t Hg/yr (Pirrone et al., 2010).

Cement production also releases mercury depending on kiln technology. About 0.1 g of Hg is liberated per tonne of cement, involving the release of 236 t/yr into the atmosphere (Pacyna et al., 2006).

Steel manufacturing production is a secondary Hg emission source. However, around 43 t of mercury per year are released to the environment from selected countries. Asia and Europe-North America, which are the principal steel producers, are releasing 14.4 t Hg/yr and 25 t Hg/yr, respectively.

The manufacture of copper, zinc, lead, nickel, and gold ores involves the release of large quantities of mercury into the atmosphere, especially in developing countries (Pirrone et al., 2009). This is due to the important quantities of mercury present in these minerals. So, in the smelting processes, mercury is vaporized and flies to the atmosphere. The global annual contribution of non-ferrous metal production is approximately 294 t/yr (Pirrone et al., 2010; Hylander & Herbert, 2008).

Waste also involves mercury release. Many mercury-containing products have been used in homes. Items that contain mercury are batteries, light bulbs, electrical equipment, pigments, drugs, plastic catalysts, dental amalgams. After use, these tools must be disposed of in a suitable way, or they release mercury. Fluorescent bulbs have about 10 mg of mercury. The old batteries contained a high mercury concentration. The most recent zinc-air button batteries also contain about 25 mg of mercury. In many specialized medical applications, such as blood pressure cuffs, specialized batteries, cantor tubes, oesophageal dilators, lung Scholander devices and in some vaccines, there is an amount of mercury (Pirrone et al., 2009). Once their use is complete, these tools must be disposed of adequately. Incinerators emit a small percentage of mercury into the

atmosphere. In any case, although the quantities of mercury coming from these installations has been lowered as they are regulated by local and international regulations. On the other hand, mercury, buried in landfill waste, is transformed into methylmercury by microorganisms. The gases that evaporate have values between 3.45 and 2953 ng/$m^3$ (Kim & Kim, 2002). Waste disposal involves the emission of approximately 187 t Hg/yr (Pirrone et al., 2010).

The production of mercury directly from the mines is in decline because of the bans from different countries. However, mercury is still used for gold artisanal extraction in some countries, involving massive releases of this metal into the environment. Mercury is used as it forms an amalgam that binds gold. Once mercury and gold are mixed, they are heated to high temperatures. Hg is thus volatilized or charged with waste products, called "tailings". These processes are often not regulated. People who carry out the extraction do not have the necessary equipment and often waste products are released into the environment, leading to health problems for workers and environmental disasters. The activity linked to the artisanal extraction of gold ores records about 1000 t Hg/yr. About 350 t is released directly into the atmosphere, while the remainder is discharged into water basins (Pirrone et al., 2009).

The chlor-alkali plants usually produce chlorine, hydrogen, and caustic soda. The cathode of the process may be mercury cell implants, diaphragms, or mercury-free membrane technology. In 2004, there were around 150 chlor-alkali plants worldwide. In November 2017, the European Union banned the use of mercury in these installations. The waste products of the mercury cells can be released into the air, but especially into water and wastewater, heavily contaminating the aquatic environment. Chlor-alkali plants are the industrial complexes that produce the most mercury-containing waste. In 2010, mercury cell-based production accounted for 21% of total world chlor-alkali capacity (Pirrone et al., 2010).

In the mercury-cell process, also called Castner–Kellner process, a saturated brine solution (sodium chloride and water) floats on top of a thin layer of mercury. In the cathode, when sodium is dissociated with chloride, it binds mercury and forms a sodium-mercury amalgam - Na(Hg). Na(Hg) is continuously drawn out of the cell and reacted with water which decomposes the amalgam into sodium hydroxide (NaOH; caustic soda), hydrogen and mercury. The mercury is recycled into the electrolytic cell. In the anode, chlorine gas ($Cl_2$) is produced and bubbles out of the cell (Figure 2.1). In this process a

quantity of mercury is expelled, and it is precisely this what contaminates the wastewater or the surrounding area.

The Ebro River is the largest river in Spain with a watershed of 84000 km$^2$. It is discharging into the Mediterranean Sea with a delta mouth of more than 30000 ha. The Ebro River receives water from several tributaries and influences about 3 million people (Carrasco et al., 2011). Its course has been used for agricultural and industrial purposes over the years. The intensive use of its flow has led to continuous and extensive contamination of its waters, leading to the spillage of urban wastewater, agricultural and industrial waste (Terrado et al., 2006). Here, 3 out of 8 Spanish chlor-alkali plants are located nearby Ebro riverbanks (Carrasco et al., 2010).

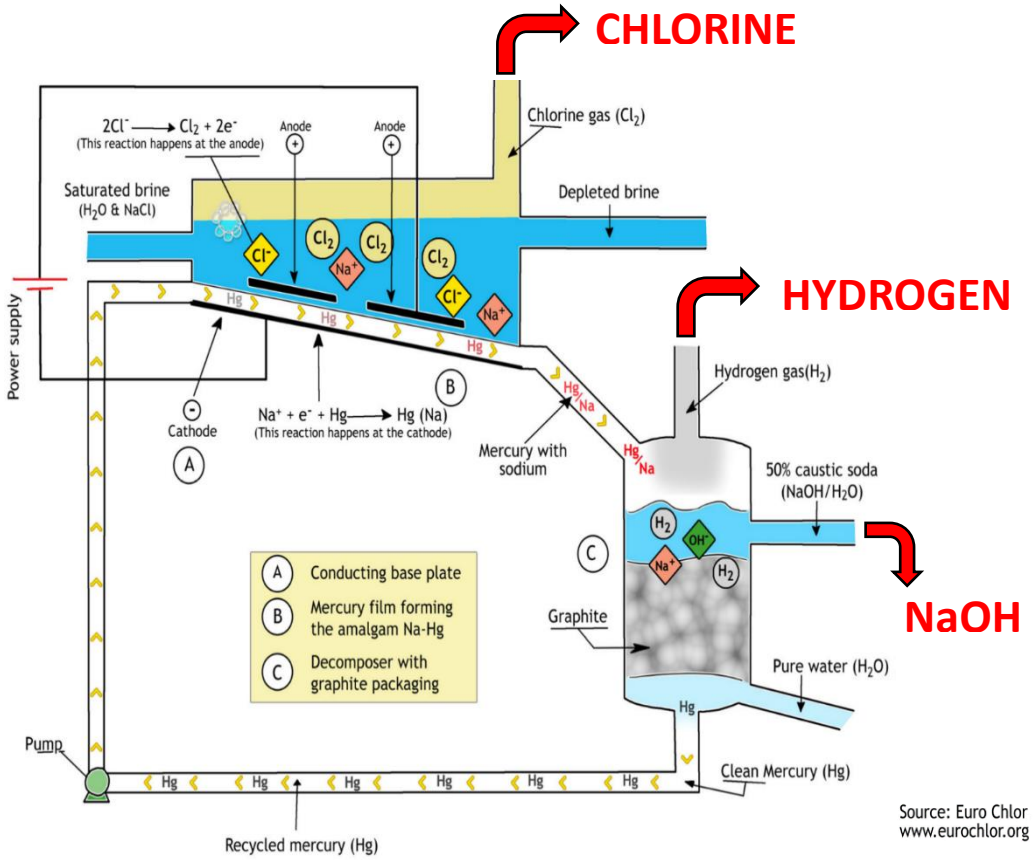


**Figure 2.1.** Reactions involved in a mercury-cell of a chlor-alkali plant. Figure adapted from www.eurochlor.org

The lower part of the Ebro River is located in Catalonia. Within the Ebro River entire course, there are almost 190 dams (Vericat & Batalla, 2006). The last downstream dam is in Flix, at the height of the Sebes natural riverine reservoir, located 110 km upstream from the Ebro River mouth. The dam was built in 1948 a few hundred metres downstream from a chlor-alkali complex. From 1949, the Flix chlor-alkali plant used the mercury-cell technique. So, the lower Ebro River stretch have been affected by the leakage wastes dumped from the industrial complex for many decades. This continuous spill of industrial waste along the river course resulted in a sludge deposit of over $3\ 10^5$ t (Palanques et al., 2020).

The Sebes natural riverine reservoir (41°14′ N, 0°32′ E) is a relatively small area (nearly 320 ha) located at the other Ebro side of the Flix chor-alkali plant. It preserves part of the river properties, having a very short water residence time (0.15 days; Carrasco et al., 2008). From 1945, the Sebes natural riverine reservoir takes the name of "Espai Natural Protegit de la Ribera d'Ebre" (Protected Natural Area of the region Ribera del Ebro; Figure 2.2). Beyond this area, the Ebro River forms a pronounced meander. The reservoir preserves a great variety of aquatic and river vegetation. It includes an area of wetlands, river islands, and a belt of river protection that surrounds it (Figure 2.2). It has a unique environment that includes many animal species that are characteristic of the area, such as kingfishers, barn owls, storks, wild horses, and otters (Figure 2.3).

However, since this reservoir is located in front of the chlor-alkali complex, the dumped Hg wastes affect not only sediments and water, but also the biota (Carrasco et al., 2011; Suarez-Serrano et al., 2010; Navarro et al., 2009; Carrasco et al., 2008; Ramos et al., 1999). The negative effect on biota is so extensive that a link is found between the amount of mercury and some species even many kilometers downriver (Carrasco et al., 2011; Navarro et al., 2009). Anyway, the high amount of mercury is not the only pollutant released into the sea by the industrial complex. In fact, a large number of pollutants in the river have been found, such as metals, organochlorine compounds (OCs), and radionuclides (Grimalt et al., 2003). Significant amounts of pollutants from Flix are transported downstream, finishing their course first in the Ebro Delta and later in the Mediterranean Sea. The final portion of the Ebro River and its delta are of enormous importance from an ecological and economical point of view. There are thriving agricultural businesses in this area that draw water and nutrients from the river. Furthermore, in the final part of the delta and nearby in the Mediterranean zone, there are

numerous aquaculture activities, and it is one of the richest fishing areas (Alcaraz et al., 2011).

Considering the current situation in the Flix reservoir and the extensive contribution of the chlor-alkali plant to the environment, a goal of this chapter was to understand how far the effects of pollutants in the Ebro River could be monitored. Accordingly, fish samples were taken from the cities near the mouth of the Ebro River, which directly overlook the Mediterranean Sea. Thus, a sampling campaign was carried out in L'Ampolla (40°48′51″N, 0°42′36″E) and in Ametlla de Mar (40°53′02.03″N, 0°48′08.87″E). Furthermore, fish and sediment samples were taken in the meander of the river, about 1 km downriver of the chlor-alkali plant. This site has not been restored by the reparation works developed in the Flix water reservoir. Furthermore, since the pollution of the chlor-alkali plant had been released for a long time, sediment samples collected in 1995-1996 in the Flix water reservoir and downriver have also been included in the overall evaluation of the environmental impact of the chlor-alkali plant, as this chapter is devoted to characterizing the spills of these installations as a guideline for evaluation of the origin of mercury found in the fish collected from the diverse Mediterranean Sea sites.

**Figure 2.2.** Flix meander and Sebes riverine natural reservoir.

**Figure 2.3.** Native animal species from the Sebes riverine reservoir.

## 2.2. Materials and methods

### 2.2.1. Sample collection and preparation

#### 2.2.1.1. Fish

The fish samples were collected between July 2019 and September 2020. The fish samples from the Ebro River were caught by authorized personnel of the Flix reservoir using the electric capture technique. The small and medium sized fish were thrown back into the river, while the medium to large sized fish were placed in an airtight plastic bag. These samples were placed in a portable freezer (2-4º C) and taken to the laboratory. A total of 17 specimens belonging to 7 different species: largemouth bass (*Micropterus salmoides*), pumpkinseeds (*Lepomis gibbosus*), welsh catfish (*Silurus glanis*), common bleak (*Alburnus alburnus*), common roach (*Rutilus rutilus*), common carp (C*yprinus carp*) and prussian carp (*Carassius gibbosus*) were cataloged.

Fish from L'Ampolla and Ametlla de Mar was collected in local markets intended for human consumption. All fish from these locations came from the Mediterranean Sea and the information relating to these samples (weight, length, trophic level) was recorded. Among these fish samples, two specimens of the angler (*Lophius piscatorius*), one from L'Ampolla, and one from Ametlla de Mar were selected for Hg isotope analysis because they represent the entire pool.

Big pieces of epaxial muscle (about 25 g) of welsh catfish and Prussian carp from the Flix reservoir and the angler from Ametlla de Mar and L'Ampolla, were taken and cut into three parts: one (10-12 g) was placed in a plastic bag and intended for the analysis of mercury isotopes, another (8-10 g) was stored in a plastic container for the analysis of metals and the last part (3-5 g) was crushed into small pieces (about 10-50 mg) and placed in a sterilized 20-mL glass vial for the analysis of the total mercury concentration. The three parts were stored in the freezer at -20º C. Prior to the analysis, the samples were completely thawed. Only wet samples were tested. All preparation tools used were made of plastic to avoid metal contamination. The instruments were cleaned with 96% ethanol (Honeywell, France) to avoid cross-contamination between samples.

#### 2.2.1.2. Otter

A sample of otter was provided by the Ebro reservoir staff in February 2020. The animal was found dead on the edge of the main road inside the reservoir, probably hit by a car. The sample was collected and taken to the laboratory.

The body of the otter was dissected. The liver and hind leg muscles were removed. All other tissues (skin, bones, joints, furs) were removed. The liver and muscle were then placed in two separate plastic vials. The samples were maintained in a freezer at -20º C. Before analysis, the samples were defrosted. About 20 grams of muscle and liver were collected, divided into two equal pieces (10 g), placed in a sterilized plastic bag and prepared for mercury isotope and metal analysis. A small part (2-3 g) of liver and muscle were instead chopped into very little chunks (10-50 mg) and placed in 20-ml glass vials. These comminute samples were then used for the analysis of the total mercury concentration. All the tools used were made of plastic and cleaned with 96% ethanol (Honeywell, France) to avoid any possible contamination.

#### 2.2.1.3. Sediment

##### 2.2.1.3.1. Sediment from meander

Surface sediment samples from the Ebro River meander in Flix were collected in May 2020. Dry sediments and wet sediments were sampled. Dry sediments were picked in the immediate vicinity (about half a meter from the riverbank) of the Ebro River and were reported in tables as "side sediment". Wet sediments were recovered in the river about one meter away from the shore and were called "center sediment" in the tables.

Aliquots of about 500 grams of sediment were taken and stored in sterilized plastic bags. All aliquots were recovered utilizing plastic utensils. The samples were hermetically sealed and kept in a portable freezer at 4º C. In the laboratory, these samples were frozen at -22º C, until the day of analysis, when they were thawed. As for the fish and otter samples, a portion was used for the analysis of the stable isotopes of mercury (about 10 g) and metals (about 10 g), while another part (about 2-3 g) was used for the analysis of the total mercury concentration. Again, the utensils used were made of plastic and cleaned with 96% ethanol (Honeywell, France).

#### 2.2.1.3.2. Sediment from the reservoir and downriver

The sediment sampling campaign was carried out in the Ebro reservoir. A flat-bottomed boat was used to operate in a shallow context and a sediment penetration system suitable for the Flix basin going deep. The boat was transported from Barcelona to Ribarroja, a few kilometers upstream from Flix, where it was put into water and where all the equipment was prepared (Figure 2.4). The sampling took place on the right side of the river, as the depth of the river is less on that side.

The sampling involved the removal of five cores of sediment within the Flix reservoir, near the chlor-alkali plant.

**Figure 2.4.** Sediment sampling campaign.

Two sampling campaigns were carried out for the recovery of sediment cores between the Catalan stretch of the Ebro River and the Mediterranean Sea. The samples were taken with a dredge.

Fifteen samples were obtained in eleven different places distributed along the stretch of the river studied. Sediment samples were placed in plastic bags and stored in a portable freezer (at 4° C) and transported to the laboratory.

### 2.2.2. Metals analysis

#### 2.2.2.1. Mercury

##### 2.2.2.1.1. Total mercury concentration

The total mercury concentration analysis was performed using AMA-254, Advanced Mercury Analysis (Altec LTD, Czech Republic), an automated atomic absorption spectrometer. A small amount of sample was placed in a sterilized container, composed of an alloy resistant to heat. Then, the sample matrix was introduced into a combustion tube, which was heated up to ~750º C for some minutes. The sample was vaporized and transported to catalytic compounds that completely removed interfering impurities. Oxygen moved the sample vapour to an amalgamator, consisting of a glass tube containing gold-plated ceramics, which is able to bind mercury. Thus, mercury-bound-amalgamator was heated to ~900º C and the released mercury vapour was carried to a cuvette, positioned in the path length of an atomic absorption spectrometer that used an element-specific lamp that emitted light at a wavelength of 253.7 nm, and a silicon UV diode detector for mercury quantitation.

The detection limit for the instrument was 0.0009 $\mu g\ g^{-1}$ in dry weight. Empty samples (blanks) were analysed to confirm that Hg was not being transferred between samples. Blanks are performed between each sample and the following one.

Instrumental accuracy was checked using the European Reference Material (ERM-BB422) from the Institute for Reference Materials and Measurements of the European Commission's Joint Research Centre (Geel, Belgium). ERM-BB422 consisted of powdered dry fish muscle and was chosen according to sample Hg concentration levels. The values obtained had to be always within the ERM-BB422 confidence interval ($0.601 \pm 0.03$ mg $kg^{-1}$). The reference material was used for every 10 samples analysed.

If the mercury levels detected in the samples were too high, more blanks were performed to bring the instrument mercury levels below the detection limit. At the end of each analysis, the sample matrix container was cleaned with distilled water to remove residual impurities caused by the previous sample analysis.

Depending on the nature of the sample analysed, the instrument needed a different quantity of samples. However, less than 100 mg was used for single sample measurement.

#### 2.2.2.1.2. Mercury stable isotope composition

Mercury isotope analyses were performed by a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) Neptune Plus (Thermo-Scientific, Waltham, Massachusetts, USA) at ALS Scandinavia laboratory, using external calibration with bracketing isotope standard reference materials (SRMs). The ALS Scandinavia choice for this type of analysis was accurate as this laboratory has great expertise in performing it. The protocol used in the measurements of the mercury isotope signatures, total digestion, and leaching procedures were executed within ALS Scandinavia Lab, while the preliminary sample preparation was performed in our laboratory, as described previously. Hg stable isotope signatures were evaluated as per-mil deviation from SRM, named NIST (National Institute of Standards and Technology) 3133. This SRM is intended for use as a primary calibration standard for the quantitative determination of mercury. A unit of SRM 3133 consisted of five 10-mL sealed borosilicate glass ampoules of an acidified aqueous solution prepared gravimetrically to contain a known mass fraction of mercury (confidence interval: 10.004 mg/g ± 0.040 mg/g).

The standard deviation of samples was calculated after two independent and consequent measurements. As explained in detail in chapter 3, mercury has mass-dependent and mass-independent isotope fractionation.

The mass-dependent fractionation (MDF) was described as δ, reported as unit per-mil and defined by the Equation 1.1 (Blum and Bergquist, 2007).

The mass-independent fractionation (MIF) was reported as Δ in unit per-mil and delineated as the deviation of a measured delta value from the theoretically predicted value due to the kinetic MDF according to the Equation 1.2 (Blum and Bergquist, 2007).

#### 2.2.2.2. Other metals

##### 2.2.2.2.1. Sediment

The samples were digested using a full attack technique. The sediment samples were previously dried and ground with an agate grinder. This technique has two parts: 1) digestion of volatile elements and 2) Digestion of non-volatile elements.

*Digestion of volatile elements*. Take 100 mg (+/- 0.01 mg) of a sample, and place in a Teflon pump. 2.5 mL of 65% suprapure nitric acid are added, the pump is closed, and heated to 90º C for a minimum of 4 hours. The pumps are then brought to room temperature. The sample dissolved in nitric acid is collected with double distilled water and centrifuged at 2000-3000 rev./min for 20 min. The supernatant is collected using a plastic pipette and placed in a 100 mL Pyrex flask. The centrifugation process is repeated 2 times, each time washing the pumps with double-distilled water. Finally, a solid residue is obtained on the one hand, where the non-volatile elements are present, and a solution in the flasks on the other, where the most volatile elements are present.

*Digestion of non-volatile elements.* The solid residue at the bottom of the centrifuge tube is collected with 2.5 mL of 65% suprapure nitric acid and 7.5 mL of 40% suprapure hydrofluoric acid and fed back into the pump. It is closed and heated at 90º C for a minimum of 4 hours. Then 2.5 mL of 60% superpure perchloric acid were added, and the solution was heated to 245º C on a plate until completely evaporation. At the end, a residue was obtained where all the non-volatile metals are present, and they were collected with 2.5 mL of 65% suprapure nitric acid and introduced into the 100 mL flask, where the volatile elements had been deposited previously.

Finally, both fractions are flushed to 100 mL with double-distilled water and stored in a refrigerator at 4º C, until further analysis.

#### 2.2.2.2.2. Biota

Teflon vessels were cleaned by rinsing with 7% $HNO_3$. Then, they were filled with 7% $HNO_3$, left in the oven overnight at 90 ºC and finally rinsed with abundant MilliQ water. All polypropylene material was cleaned by soaking into 7% $HNO_3$ for 48 hours, followed by rinsing with abundant MilliQ water.

A total of 100 mg of each fish and otter sample were introduced in Teflon vessels, together with 1 mL of 60 % $HNO_3$ (Merck; Darmstadt, Germany) and 0.5 mL of $H_20_2$ (Merck). They were then left in an oven at 90º C overnight. After cooling, the vessels were opened, and the samples were dissolved in 16.5 mL of 1% $HNO_3$ dilution. Finally, samples were placed in plastic tubes and stored in a refrigerator until instrumental analysis. Before analysis, an internal standard of indium (10 ppb) was added to the samples. Depending on density, samples were diluted with MilliQ water to 30 mL or 60 mL to avoid spectral interferences.

#### 2.2.2.2.3. Metal instrumental analysis and quality control

The analysis technique was high frequency induced plasma atomic emission spectroscopy (ICP-AES) for Al, Ca, Fe, and Zn; and high frequency induced plasma mass spectroscopy (ICP-MS) for the other metals (As, Cd, Cr, Cu, Ni, Pb, Se, Tl, U).

In each batch of analysis (typically 20 samples in total), a target, a certified material (MESS-3 and PACS-2 from the National Research Council of Canada), and a replicated sample were analyzed as quality control.

### 2.2.3. Data analysis

The statistical software R (R Development Core Team, 2019) and Office packages (Microsoft Corporation, 2020) were used for data analysis and figures and tables realization. The images were adapted to Office packages software. Principal component analysis (PCA) of sample metals were performed with the Statistical Package for Social Science – SPSS Statistics - software (International Business Machine Corporation, 2019).

## 2.3. Results

### 2.3.1. Mercury

The mercury concentration was measured in sediments and biota. Table 2.1 shows the main parameters of the sediment samples, such as the grain sizes and the coordinates of the points where they were taken. The sediment mercury concentration values are also shown in this Table. They ranged between 16 and 120 μg/g in the Flix reservoir, between 7.2 and 19 μg/g in the meander, and between 0.07 and 1.9 μg/g downriver (Figure 2.5). The granulometric parameters showed muddy bottoms in the part of the river near the reservoir, while the same values mainly describe sandy bottoms for the remaining stretch of the river. The granulometry values oscillated between 93.5% and 97.9% of granules that have a diameter lower than 63 μm in the Flix reservoir (with an average that stands at 95.2%), while they were between 10% and 94% in the remaining part of the river (with an average of 60%).

The percentage of total organic matter (TOC) in these sediments also varied greatly from one sample to another. For instance, in the Flix reservoir, the percentage of TOC varied from 0.6% to 11%, with an average value of 4.6%, while it was between 0.12% and 1.5% downriver, with an average of 0.76%.

The sediments of the Ebro River are highly variable because they contained variable amounts of pebbles, sands, and fine material (<60 μm). These characteristics were determinants of their mercury retention capacity. For this purpose, the ratios between Hg and TOC were also considered in an attempt to normalize Hg values. The mercury levels of the sediments in relation to the organic matter in the Flix reserve ranged from 2.8 to 45, while in the final part of the Ebro River they ranged from 0.23 to 4.3.

**Table 2.1.** Sediment samples, coordinates, granulometry, and Hg concentration.

| Sediment sample | | Coordinates | | Granulometry (%) | | | Hg concentration | |
|---|---|---|---|---|---|---|---|---|
| Sites | Sample ID | Latitude (°N) | Longitude (°E) | <63 μm | >63 μm | TOC (%) | Hg (μg/g) | Hg/TOC |
| Flix reservoir | R-a | 41º14’1.6’’ | 0º32’5.4’’ | 97.9 | 2.1 | 9.1 | 120 | 13 |
| | R-b | 41º14’0.57’’ | 0º32’12’’ | 94.3 | 5.7 | 11 | 31 | 2.8 |
| | R-c | 41º14’1.2’’ | 0º32’16.5’’ | 97 | 3 | 1.3 | 22 | 17 |
| | R-d | 41º13’59.8’’ | 0º32’27’’ | 93.5 | 6.5 | 1.2 | 54 | 45 |
| | R-e | 41º13’58.1’’ | 0º32’36.8’’ | 93.5 | 6.5 | 0.6 | 16 | 26 |
| Flix meander | Side | 41°14'00.6" | 0°32'55" | | | | 7.2 | |
| | Center | 41°14'00.6" | 0°32'55" | | | | 19 | |
| Ebro downriver | D-a | 40º50’56’’ | 0º31’35’’ | 27 | 73 | 0.31 | 1.1 | 3.6 |
| | D-b | 41º01’48’’ | 0º35’21’’ | 73 | 27 | 1.3 | 1.9 | 1.5 |
| | D-c | 40º46’51’’ | 0º31’14’’ | 10 | 90 | 0.25 | 0.83 | 3.3 |
| | D-d | 40º43’42’’ | 0º34’27’’ | 94 | 6 | 0.73 | 0.21 | 0.29 |
| | D-e | 40º42’33’’ | 0º37’29’’ | 76 | 24 | 0.31 | 0.07 | 0.23 |
| | D-f | 40º43’03’’ | 0º39’20’’ | 31 | 69 | 0.12 | 0.52 | 4.3 |
| | D-g | 40º42’56’’ | 0º42’44’’ | 77 | 23 | 0.98 | 1.2 | 1.2 |
| | D-h | 40º42’40’’ | 0º45’24’’ | 73.5 | 26.5 | 0.95 | 1.6 | 1.7 |
| | D-i | 40º42’34" | 0º49’18’’ | 65 | 35 | 1 | 1.4 | 1.4 |
| | D-j | 40º42’27’’ | 0º49’38’’ | 43.5 | 56.5 | 1.5 | 0.79 | 0.52 |
| | D-k | 40º43’02’’ | 0º50’38’’ | 86 | 14 | 0.92 | 1. 5 | 1.7 |

Biota also showed high mercury levels. The total mercury (T-Hg) concentrations in fish collected from the Sebes natural reservoir are shown in Table 2.2. A total of 6 specimens out of the 17 samples recorded T-Hg values above the EU limit recommended for human consumption (0.5 mg $kg^{-1}$ wet weight, ww). The fish samples had a T-Hg mean value of 0.39 mg $kg^{-1}$ ww, and a median of 0.28 mg $kg^{-1}$ ww, ranging from 0.15 to 0.83 mg $kg^{-1}$ ww.

The samples from L'Ampolla and Ametlla de Mar showed high levels of mercury. In the angler (*Lophius piscatorius*), the EU recommended threshold for human consumption is 1 mg $kg^{-1}$ ww. In both samples, the T-Hg concentration is higher than this value.

The otter sample showed even higher total mercury levels. The liver sample had mercury concentrations almost ten times higher than the muscle, which was still very high. The liver and muscle showed mercury levels of 10 mg/kg ww and 1.5 mg/kg ww, respectively.

The stable isotope composition of mercury was reported in Table 2.3. Mass-dependent fractionation (MDF) and mass-independent fractionation (MIF) have been described. The MDFs were highly variable. The maximum variability was reached in the δ202Hg isotope, which ranged from strongly negative values (-2.105 ‰) to positive values (0.483 ‰).

The MIF values were much more homogeneous than the respective MDFs. In the odd-numbered MIFs, sediment samples had negative values, while fish and otter samples showed positive values. Some variability was observed in the even-MIF values, which, despite being very low, they were fluctuating from positive to negative values, both in sediments and biota.

**Table 2.2.** Biota samples and total mercury concentrations.

| Sample information | | | Total-Hg |
|---|---|---|---|
| Sample site | Sample species | | mg/kg ww |
| Flix meander | Largemouth bass | *Micropterus salmoides* | 0.23 |
| | Largemouth bass | *Micropterus salmoides* | 0.30 |
| | Largemouth bass | *Micropterus salmoides* | 0.25 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.69 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.63 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.52 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.42 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.22 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.24 |
| | Pumpkinseed | *Lepomis gibbosus* | 0.20 |
| | Welsh catifish | *Silurus glanis* | 0.83 |
| | Welsh catifish | *Silurus glanis* | 0.83 |
| | Common bleak | *Alburnus alburnus* | 0.21 |
| | Common roach | *Rutilus rutilus* | 0.34 |
| | Common roach | *Rutilus rutilus* | 0.16 |
| | Common carp | *Cyprinus carp* | 0.18 |
| | Prussian carp | *Carassius gibbosus* | 0.80 |
| L'Ampolla | Angler | *Lophius piscatorius* | 1.4 |
| Ametlla de Mar | Angler | *Lophius piscatorius* | 1.1 |
| Flix reservoir | Otter (muscle) | *Lutra lutra* | 1.5 |
| | Otter (liver) | *Lutra lutra* | 10 |

### 2.3.2. Metals

The metals examined were aluminium (Al), arsenic (As), calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), nickel (Ni), lead (Pb), selenium (Se), thallium (Tl), uranium (U), zinc (Zn) (Table 2.4).

Arsenic had concentration ranges between 12-27 μg/g in the sediments of the Flix reservoir (average = 17 μg/g), between 8.6-20 μg/g downriver (average = 12 μg/g) and 0.026-3.1 μg/g in the meander (Figure 2.6). Great variability was observed in fish, between 0.027-0.13 μg/g in the Ebro River, and between 8.9-16 μg/g in the Mediterranean Sea. The otter showed a concentration of 5.2 μg/g.

The sedimentary cadmium concentrations ranged between 0.76 and 7.6 μg/g in the Flix reservoir, 0.60 and 1.6 μg/g, downriver, and 0.026 μg/g and 0.19 μg/g in the meander (Figure 2.7). In fish, the values were close to 0, never exceeding 0.0004 μg/g. In the otter, the cadmium concentrations were 0.22 μg/g.

The concentrations of copper in the Flix reservoir sediments ranged between 29 μg/g and 52.5 μg/g, they were between 10 μg/g and 34 μg/g downriver and between 10 μg/g and 12 μg/g in the Flix meander (Figure 2.8). In fish, they ranged between 0.070-0.23 μg/g, with an average of 0.12 μg/g. The otter showed high levels, 13 μg/g.

The sedimentary concentrations of nickel observed in the Flix reservoir sediments ranged between 44-120 μg/g, with an average of 71 μg/g. Downriver, they were between 14-36 μg/g (average 25.5 μg/g), and in the meander they showed high variability, with values close to zero near the side and around 6.6 μg/g in the center (Figure 2.9). In fish, the Ni levels ranged from 0.0037 to 0.0081 μg/g, while in the otter they were 12 μg/g.

The sedimentary lead concentrations ranged between 14 μg/g and 47 μg/g, with an average of 25 μg/g, in the Flix reservoir, while they varied between 11 μg/g and 140 μg/g, with an average of 47 μg/g, in the final stretch of the Ebro River (Figure 2.10). In the sediments of the Flix meander, they ranged from 0.0023 μg/g to 26 μg/g (Figure 2.10). The biota had variable values of Pb, between 0.0004 μg/g and 0.0051 μg/g in fish and 28 μg/g in the otter. Selenium had sediment concentration ranges of 11-24 μg/g in the Flix reservoir, 5.2-19.5 μg/g downriver, and 0.31-8-4.5 μg/g in the meander (Figure 2.11).

**Table 2.3.** Mercury stable isotope composition in sediment, fish, and otter from Flix meander, Flix reservoir, and the Mediterranean Sea.

| Sample ID | | Site | MDF signature - unit per mil (‰) | | | | MIF signature - unit per mil (‰) | | |
|---|---|---|---|---|---|---|---|---|---|
| Sample | | | $\delta^{199}Hg$ | $\delta^{200}Hg$ | $\delta^{201}Hg$ | $\delta^{202}Hg$ | $\Delta^{199}Hg$ | $\Delta^{200}Hg$ | $\Delta^{201}Hg$ |
| Sediment | Side | Flix meander | -0.140 | -0.039 | -0.144 | -0.008 | -0.138 | -0.035 | -0.138 |
| Sediment | Center | Flix meander | -0.049 | 0.138 | 0.134 | 0.298 | -0.124 | -0.011 | -0.090 |
| Welsh catfish | *Silurus glanis* | Flix meander | 0.484 | 0.192 | 0.634 | 0.447 | 0.371 | -0.033 | 0.298 |
| Welsh catfish | *Silurus glanis* | Flix meander | 0.387 | 0.156 | 0.552 | 0.376 | 0.292 | -0.033 | 0.269 |
| Prussian carp | *Carassius gibbosus* | Flix meander | 0.462 | 0.267 | 0.666 | 0.483 | 0.341 | 0.025 | 0.303 |
| Angler | *Lophius piscatorius* | L'Ampolla | 0.494 | 0.118 | 0.526 | 0.224 | 0.437 | 0.006 | 0.358 |
| Angler | *Lophius piscatorius* | AmetlladeMar | 0.331 | -0.098 | 0.178 | -0.152 | 0.370 | -0.022 | 0.292 |
| Otter | *Lutra lutra* | Flix reservoir | -0.051 | -1.069 | -1.189 | -2.105 | 0.479 | -0.012 | 0.394 |

Fish and the otter showed similar values, in the range of 0.26 μg/g and 0.615 μg/g, the last value being in the otter.

Sedimentary zinc ranged in an interval of 58-280 μg/g in the Flix reservoir sediments, between 43-150 μg/g in the Ebro downriver, and between 25-39 μg/g in the Flix meander (Figure 2.12). Zn levels in biota exceeded 2.9 μg/g, with maxima of 5.7 μg/g and 31 μg/g in fish and the otter, respectively.

In the Flix reservoir sediments, the chromium concentration ranged from 71 μg/g to 580 μg/g with an average value of 230 μg/g. In the last section of the Ebro River, Cr concentrations in sediments were much lower, having a range of 25-65.5 μg/g and an average of 44 μg/g. In the Flix meander, the sediment Cr levels were very low. The concentrations of this metal in the Flix meander and the Mediterranean fish were very similar, with an average value of about 0.0028 μg/g. In the otter, the values were very high, exceeding 9.6 μg/g.

As expected, calcium and iron were very high in the biota and in the sediments. The concentrations of aluminium in fish were low, 0.18 μg/g, while in the otter they were high, 4100 μg/g.

Thallium was not found in either biota or sediments. Uranium was low in fish, between 0 and 0.0003 μg/g, but not negligible in the otter, 1.5 μg/g.

**Table 2.4.** Metals in sediment, fish, and mammal (otter). The samples were collected in the Flix meander, Flix reservoir, downriver, and Mediterranean sites near the Ebro River mouth.

| Site | Sample ID | Metals (µg/g) | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Al | As | Ca | Cd | Cr | Cu | Fe | Ni | Pb | Se | Tl | U | Zn |
| Flix reservoir | R-a | | 20 | | 1.5 | 160 | 52.5 | | 52 | 35.5 | 24 | | | 96 |
| | R-b | | 13 | | 1 | 170 | 33 | | 63 | 16 | 23 | | | 72 |
| | R-c | | 12 | | 1.8 | 180 | 42.5 | | 72 | 14 | 11 | | | 110 |
| | R-d | | 12 | | 0.76 | 71 | 29 | | 44 | 47 | 15 | | | 58 |
| | R-e | | 27 | | 7.6 | 580 | 48 | | 122 | 14 | 17 | | | 280 |
| Flix meander | Side | 78 | 0.026 | 74 | 0.026 | 0.0008 | 12 | 160 | 0.0023 | 0.011 | 4.5 | 0.003 | 0.0001 | 25 |
| | Center | 2500000 | 3.1 | 8300 | 0.19 | 5.7 | 10 | 5500 | 6.6 | 26 | 0.31 | 0.025 | 1.8 | 39 |
| Ebro downriver | D-a | | 10 | | 0.95 | 31 | 11 | | 16 | 26 | 5.8 | | | 75 |
| | D-b | | 13 | | 1.6 | 46 | 26 | | 23 | 91 | 5.4 | | | 120 |
| | D-c | | 8.6 | | 0.96 | 25 | 10 | | 14 | 18 | 5.6 | | | 50 |
| | D-d | | 20 | | 1.3 | 50 | 34 | | 28 | 140 | 5.2 | | | 150 |
| | D-e | | 12 | | 0.64 | 29 | 12 | | 18 | 11 | 9.2 | | | 43 |
| | D-f | | n.d.* | | n.d.* | n.d.* | n.d.* | | n.d.* | n.d.* | n.d.* | | | n.d.* |
| | D-g | | 10 | | 1.2 | 47 | 20 | | 33 | 30 | 10 | | | 69 |
| | D-h | | 11 | | 0..835 | 51.5 | 27 | | 31.5 | 33.5 | 16.5 | | | 93 |

*not determined.

**Table 2.4. (Cont.)** Metals in sediment, fish, and mammal (otter). The samples were collected in the Flix meander, Flix reservoir, downriver, and Mediterranean sites near the Ebro River mouth.

| Site | Sample ID | Metals (µg/g) | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Al | As | Ca | Cd | Cr | Cu | Fe | Ni | Pb | Se | Tl | U | Zn |
| Ebro downriver | D-i | | 10 | | 1.1 | 55 | 26 | | 32 | 31 | 12 | | | 100 |
| | D-j | | 10.5 | | 0.595 | 35.5 | 17.5 | | 23.5 | 23.5 | 19.5 | | | 70 |
| | D-k | | 14 | | 0.645 | 65.5 | 29.5 | | 36 | 37 | 17 | | | 110 |
| Flix meander | Welsh catfish | 38 | 0.052 | 83 | 0.0001 | 0.0018 | 0.1 | 1 | 0.0047 | 0.0005 | 0.32 | 0.0008 | 0 | 3 |
| Flix meander | Welsh catfish | 34 | 0.028 | 81 | 0.0001 | 0.0021 | 0.1 | 0.98 | 0.0037 | 0.0004 | 0.26 | 0.0009 | 0 | 3 |
| Flix meander | Prussian carp | 72 | 0.13 | 980 | 0.0004 | 0.005 | 0.23 | 3.6 | 0.0078 | 0.0051 | 0.5 | 0.0005 | 0.0003 | 5.7 |
| L'Ampolla | Angler | 130 | 16 | 130 | 0.0001 | 0.0041 | 0.12 | 0.97 | 0.0065 | 0.0023 | 0.5 | 0.0001 | 0.0001 | 3.2 |
| Ametlla de Mar | Angler | 91 | 8.9 | 110 | 0.0002 | 0.001 | 0.07 | 1.1 | 0.0081 | 0.0021 | 0.45 | 0.0001 | 0.0001 | 2.9 |
| Flix reservoir | Otter | 4100000 | 5.2 | 100000 | 0.22 | 9.6 | 13 | 9800 | 12 | 28 | 0.61 | 0.034 | 1.5 | 31 |

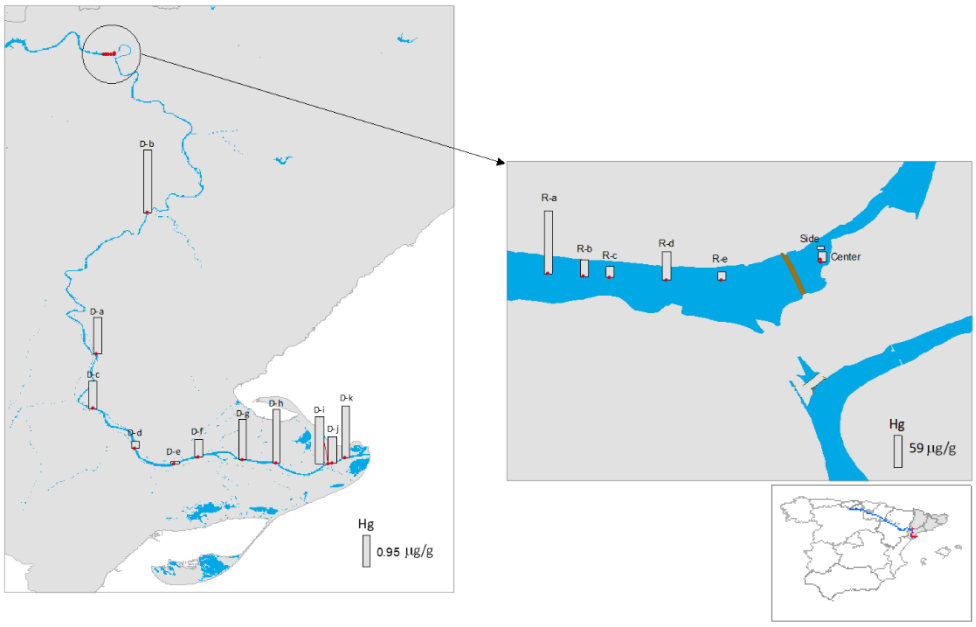


**Figure 2.5.** Concentrations of mercury in the sediments of the Flix reservoir and downriver.

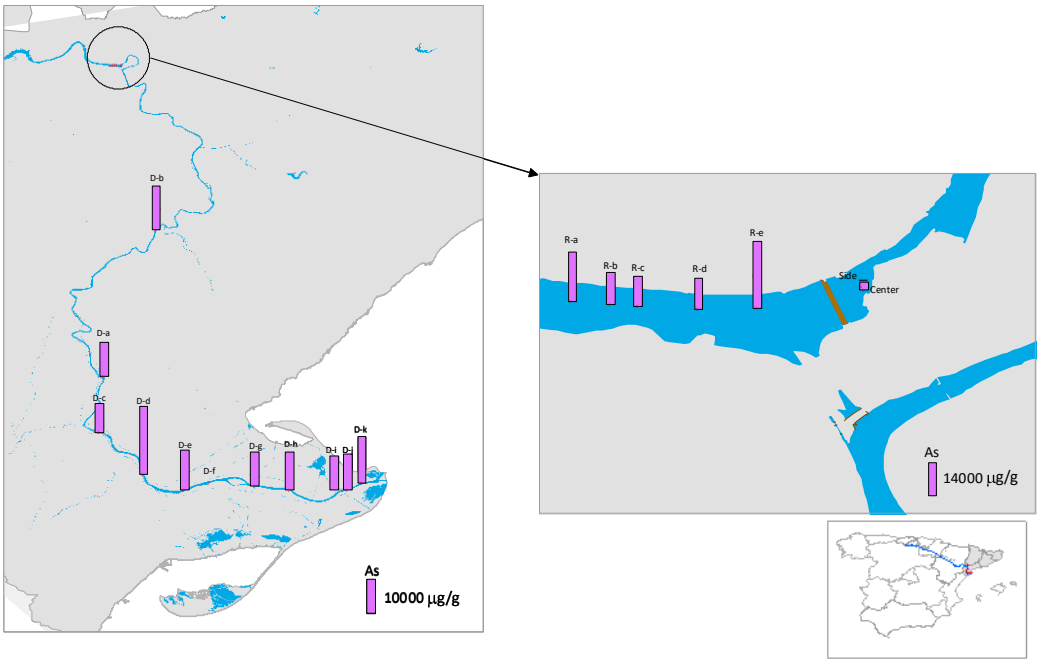


**Figure 2.6.** Concentrations of arsenic in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

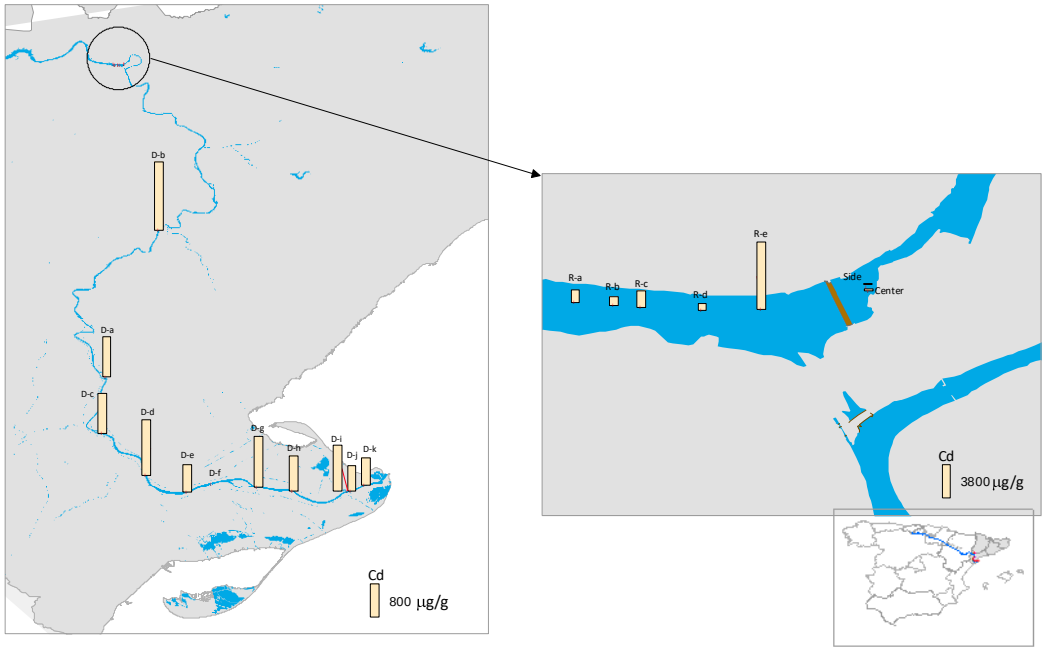


**Figure 2.7.** Concentrations of cadmium in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

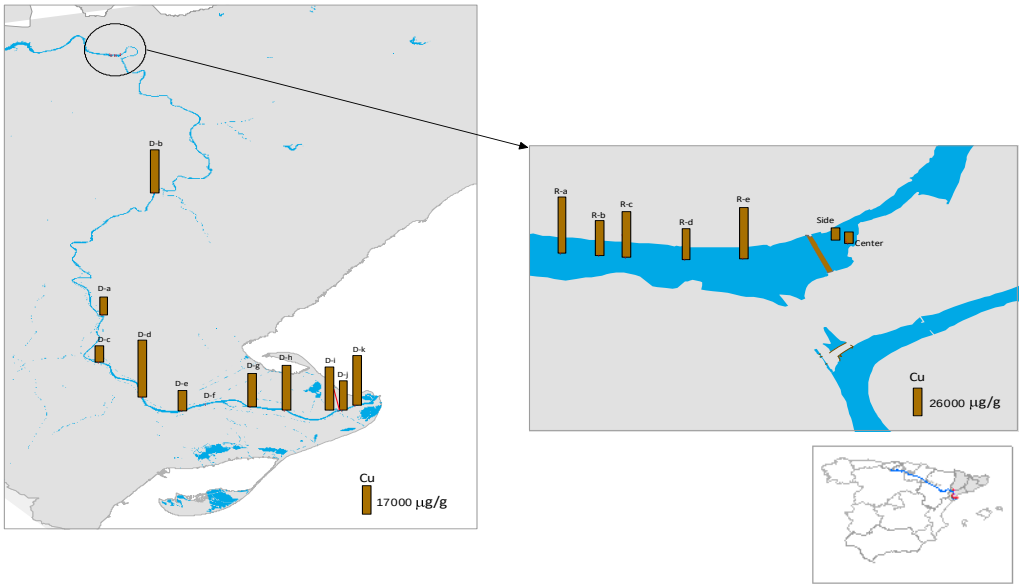


**Figure 2.8.** Concentrations of copper in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

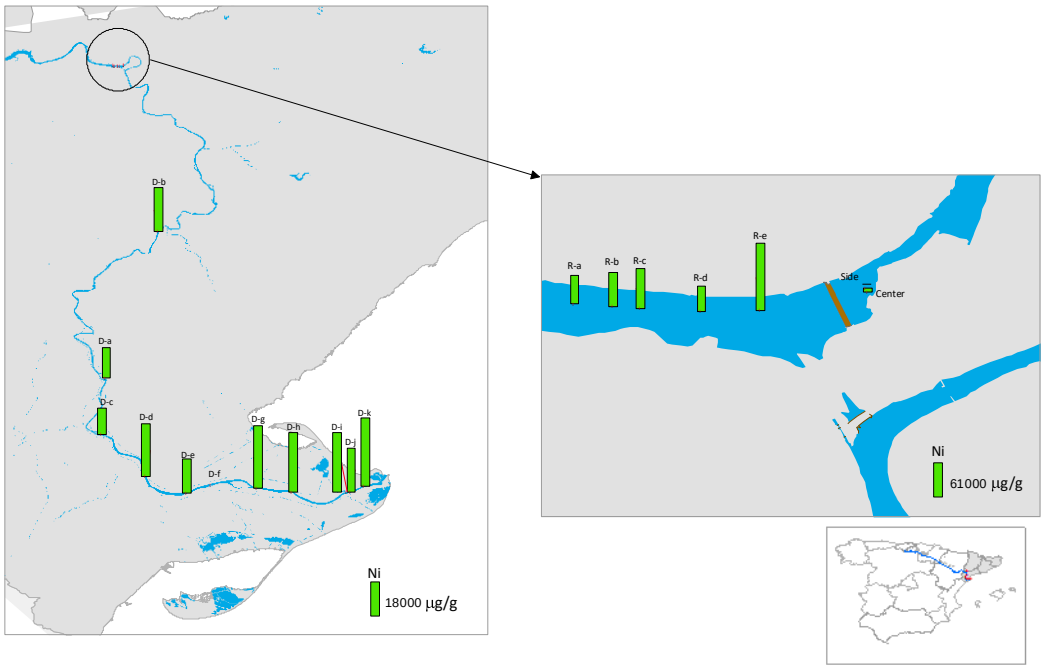


**Figure 2.9.** Concentrations of nickel in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

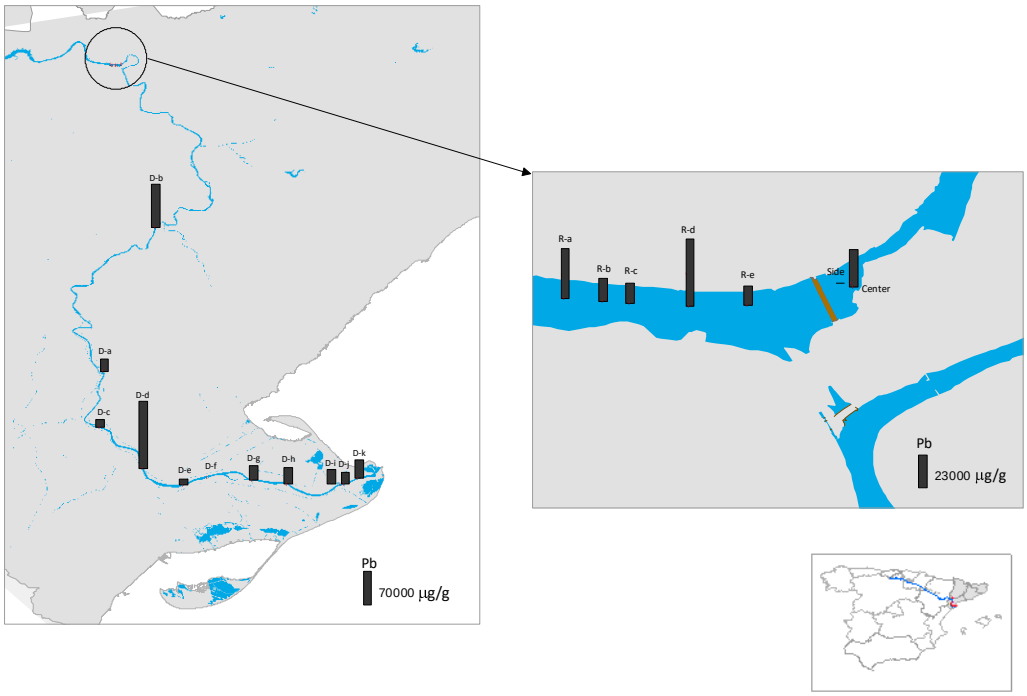


**Figure 2.10.** Concentrations of lead in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

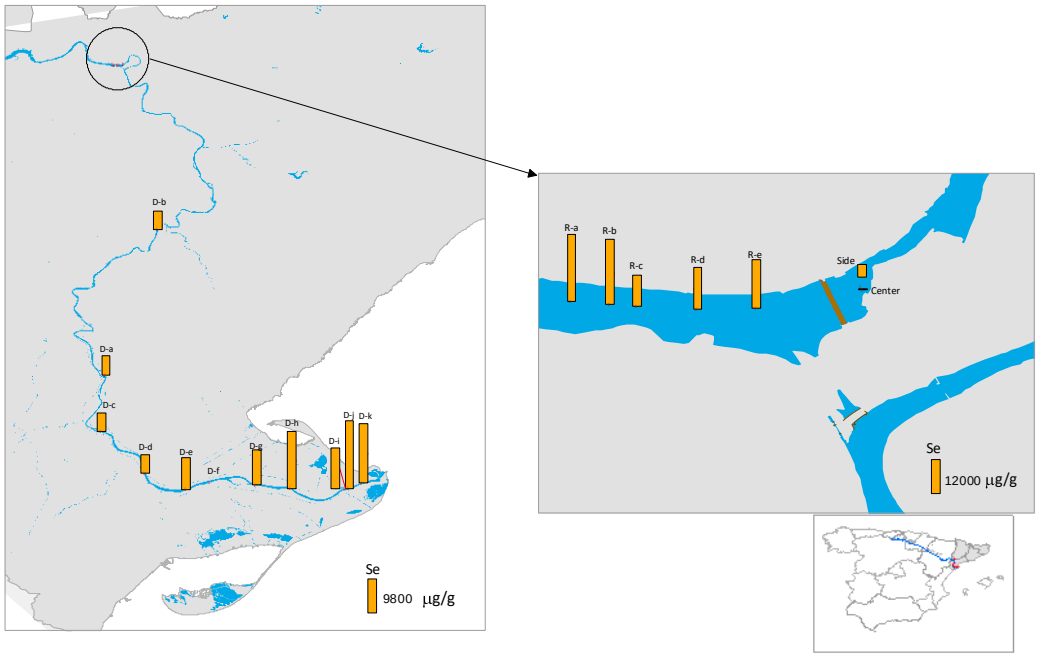


**Figure 2.11.** Concentrations of selenium in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

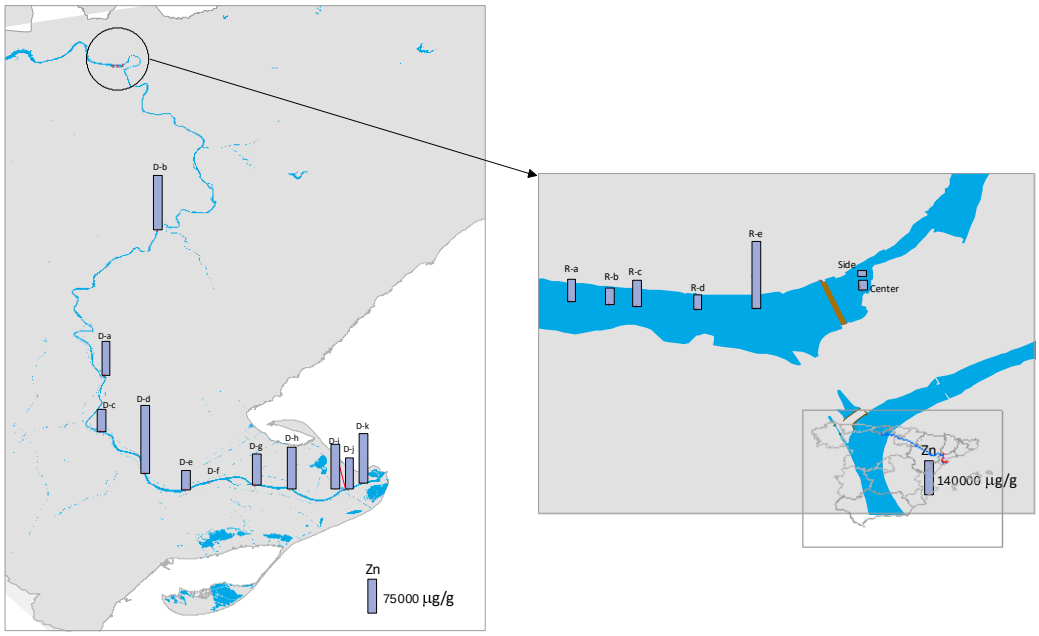


**Figure 2.12.** Concentrations of zinc in the sediments of the Flix reservoir and downriver. No data was available for sample D-f.

### 2.3.2.1. Principal Component Analysis (PCA)

Principal component analysis (PCA) was performed for the assessment of the main distribution patterns of the metals in the sediments. The Kaiser-Meyer-Olkin's (KMO) parameter was calculated for evaluation of the usefulness of this multivariate method in this specific case. KMO should reach a value of 0.5 or more to show that variables are interdependent among them (Tabachnick & Fidell, 2001). In the present database, the KMO value reached 0.646, which justifies the choice of PCA for pattern assessment.

The metals from which no concentrations were available in all samples were excluded. Five principal components were chosen, which explained 98.3% of the variance of the database. The loadings are summarized in Figure 2.13.

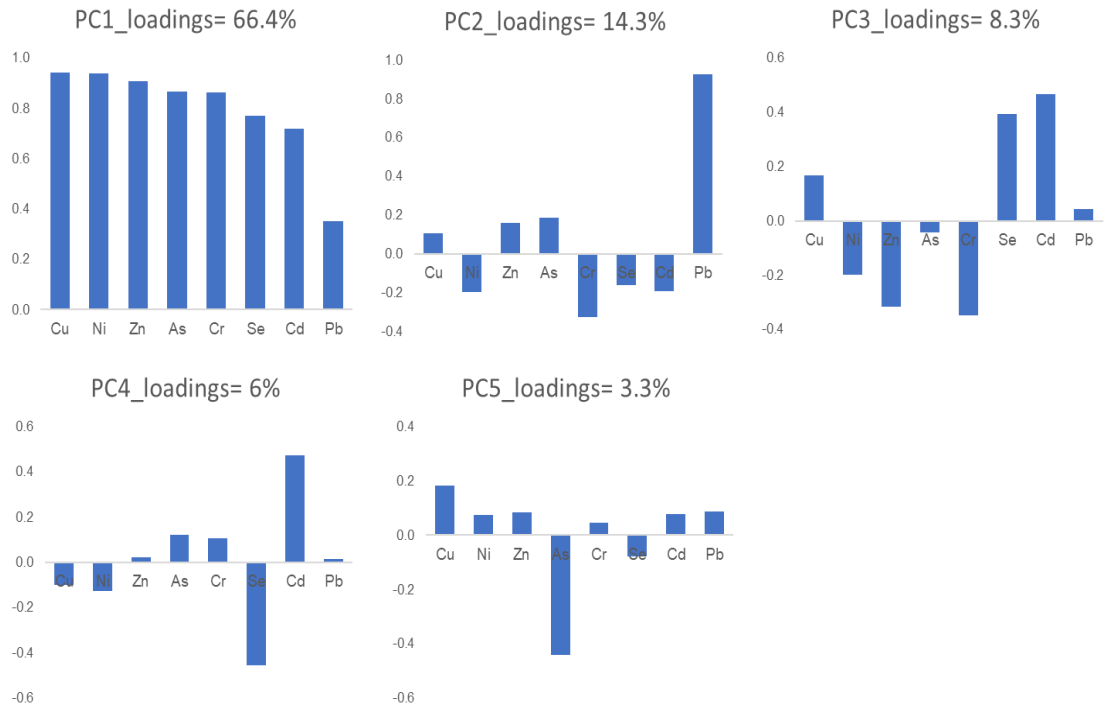


**Figure 2.13.** Principal component loadings.

The first PC has a dominant gradient of most metals. The second PC is mainly recording Pb variance because this metal showed additional sources than those of the other metals. PC3-5 were not indicating consistent differences and were mainly related to the database variability.

Examination of the correlation matrix (Table 2.5) provided additional insight into these PCA results. All metals were positively correlated among them, except Pb, that was not correlated with most of the other metals.

**Table 2.5.** Correlation matrix among metals in PCA.

| | As | Cd | Cr | Cu | Ni | Pb | Se | Zn |
|---|---|---|---|---|---|---|---|---|
| As | 1.000 | 0.586 | 0.689 | 0.746 | 0.741 | 0.436 | 0.594 | 0.788 |
| Cd | 0.586 | 1.000 | 0.576 | 0.691 | 0.561 | 0.108 | 0.551 | 0.497 |
| Cr | 0.689 | 0.576 | 1.000 | 0.705 | 0.932 | -0.001 | 0.533 | 0.842 |
| Cu | 0.746 | 0.691 | 0.705 | 1.000 | 0.866 | 0.444 | 0.785 | 0.813 |
| Ni | 0.741 | 0.561 | 0.932 | 0.866 | 1.000 | 0.145 | 0.715 | 0.859 |
| Pb | 0.436 | 0.108 | -0.001 | 0.444 | 0.145 | 1.000 | 0.129 | 0.456 |
| Se | 0.594 | 0.551 | 0.533 | 0.785 | 0.715 | 0.129 | 1.000 | 0.548 |
| Zn | 0.788 | 0.497 | 0.842 | 0.813 | 0.859 | 0.456 | 0.548 | 1.000 |

In order to examine the sample distribution according to the two main PCs, a scatterplot was generated (Figure 2.14). The scores of the PC1 distributed the samples uniformly, except for two sediment samples of the reservoir, which have high concentrations for almost all metals (Table 2.4). Those of PC2 highlighted the two samples with higher concentrations of Pb than the others (Figure 2.14) which were located downstream.

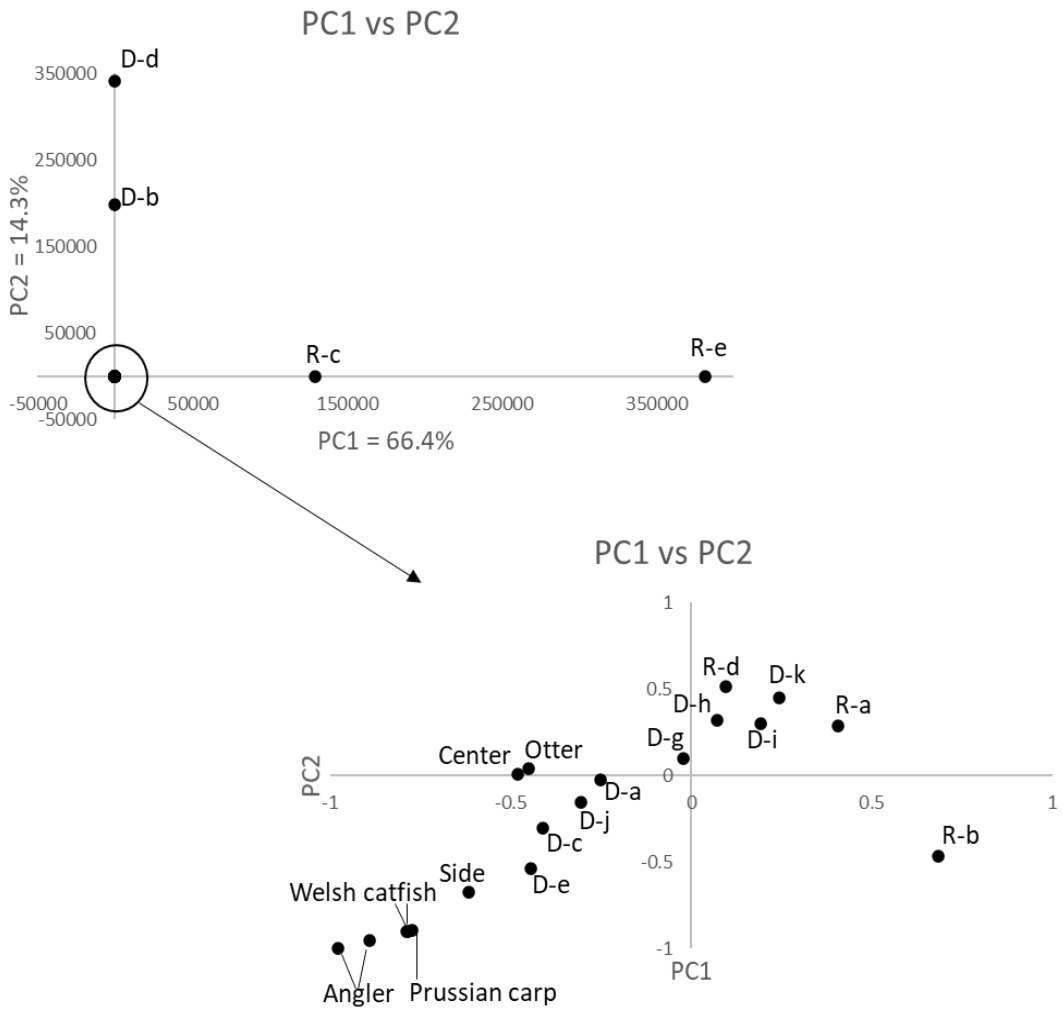


**Figure 2.14.** Scores of the Principal components 1 and 2.

## 2.4. Discussion

Total organic matter content was much higher in the reservoir than downstream. The highest mercury concentrations were logically recorded in the vicinity of the chlor-alkali plant, which is consistent with the use of this metal in the cathode of this installation. However, high levels of mercury were also recorded downstream which evidences the impact of the spills of this factory along the river course. The observed mercury concentrations were very high in some sites, reaching a maximum value of 120 μg/g. Similar concentrations have been observed in other water basins contaminated by mercury-cell chlor-alkali spills (Bolanos-Alvarez et al., 2016; Ullrich et al., 2007). The Hg spills contaminated not only the watercourse and its sediments but also the entire local ecosystem since a great part of the biota analysed have high mercury concentrations.

Thirty-five percent of the fish collected in the Flix meander had T-Hg concentration above the EU threshold values for human consumption (0.5 mg/kg ww; OJEU, 2006). The observed mean, median, and range of total mercury concentrations are in accordance or lower than values found in previous studies (Carrasco et al., 2011; Navarro et al., 2009; Carrasco et al., 2008). The observed variability of the present database reflects the characteristics of the different species analysed, e.g. dietary habits, specimen sizes, and ages (Liu et al., 2014; Ruelas-Inzunza et al., 2008; Cizdziel et al., 2002).

The high mercury levels of the otter reflect that this organism is one step higher than fish in the food web. Otters are omnivorous, they live near rivers where they spend a lot of time, hunt, and prepare a den. They eat everything but are very fond of fish (Adrian & Delibes, 1986). So, this behaviour justifies the high amount of mercury in the muscles and liver. The different results of the total mercury concentration in these body compartments are consistent with the organ functions (Havelkova et al., 2008). The liver is the organ where methylmercury is mainly stored and redistributed. Methylmercury de-methylation also occurs in the liver (Barst et al., 2013; Young, 1992) that has the function of eliminating toxic substances that have been introduced into the body. The muscle is supplied continuously with blood and accumulates nutrients and pollutants throughout the animal's entire existence. Thus, muscle accumulates mercury over the life of the animal, while the liver provides a record of the mercury levels at the moment in which the animal perished (Rua-Ibarz et al., 2019).

The concentration of mercury in Mediterranean fish is equally high. As already reported in chapter 1 of this dissertation, the Mediterranean is a hot spot for mercury (Rajar et al., 2007). The mercury isotope analysis allows to discriminate and trace the different sources. In the comparison of the MDF values of the analysed samples, a great variability is observed. Each type of sample undergoes a series of chemical-physical reactions that involve a different contribution of the MDF isotopic signature.

A different picture is reflected on the values of MIFs. Excluding the even-MIF isotope, which have a very low value in comparison with the others, the odd-MIF show differences only when we compare inert samples (sediments) and biota (fish and otter). Odd-MIFs are isotopic signatures influenced by the magnetic isotope effect (MIE) and the nuclear volume effect (NVE; chapter 1), which have a very similar contribution in the study context. Therefore, odd-MIF values can explain more clearly and precisely if there

is a relationship between the levels of mercury in the biota and the presumed source (Yin et al., 2014). In Table 2.3, the values of Hg odd-MIFs in all biotas are very similar. Although they belong to different species and categories of animals and even though they have been analysed in different periods and live in different ecological niches (river and sea), the fish and otter of the Flix reservoir and the Mediterranean fish have a very similar isotope signature for odd-MIFs. The odd-MIF isotopic signature in the Flix reservoir otter and fish from the meander was certainly influenced by the input of chlor-alkali plant spills. Since the Mediterranean fish also have the same odd-MIF isotopic signature, it can be stated that there is an extensive effect of the mercury contamination due to the chlor-alkali plant contribution. This mercury contamination starts from the Flix reservoir, crosses the Ebro River final stretch, and reaches the Mediterranean Sea, far more than 100 kilometers from the chlor-alkali plant. The mercury isotopic composition in other rivers is very different from the fish samples in this study (Tsui et al., 2020; Li et al., 2016; Kwon et al., 2014). Samples from other environments (oceans, sea, and lakes) also have a different isotopic composition than the samples from the Ebro River (Blum et al., 2020; Lee et al., 2020; Motta et al., 2020; Rua-Ibarz et al., 2019; Madigan et al., 2018; Blum et al., 2013; Gantner et al., 2009) which confirms the assignment to the chlor-alkali spills.

Concerning the general metal analysis, literature data on otters are very limited. So, the data of the present study are highly valuable because they concern a scarcely considered species in a highly contaminated context. Furthermore, given its importance in its habitat as top predators, the otter could be an excellent sentinel organism for mammals.

Chromium has been classified as a molecule that possesses mutagenic and teratogenic properties. In an aquatic environment, it is very persistent and has a high accumulation potential, especially in sediments. A large difference in concentration for Cr is reported between the Flix reserve and the Ebro downriver sediments. These values are indicative of significant contamination by this metal. In uncontaminated river sediments, there are concentrations of the order of 35-50 μg/g (Palanques et al., 1999). Therefore, the Cr concentrations in Flix reservoir sediments are ten times more Cr-enriched than in normal sediments. The concentrations of Cr found in the sediments of the Flix reservoir are very similar to those from other contaminated basins (Jordao et al., 1997). Ebro downriver sediments indeed fit with uncontaminated fluvial sediments. The

Cr in the Flix fish samples from the meander is significantly lower than in Cr-contaminated fish samples (Jordao et al., 1997). This is probably due to the different sensitivity of the fish species to Cr (Velma et al., 2009). In fact, some fish species appear to show greater sensitivity to Cr toxicity than others (Aslam & Yousafzai, 2017; Velma et al., 2009). The Cr levels in the otter are much higher than in other studies on the same animal (Brand et al., 2020; Walker et al., 2011; Mason & Stephenson, 2001).

Zinc is an essential element (Clearwater et al., 2002). However, even essential elements, if present in high quantities, can lead to toxicity (Yilmaz, 2003). The zinc concentrations in the fish analysed are much lower than in other studies (Yilmaz, 2003; Unlu & Gumgum, 1993). The otter analysed in the present study also has significantly lower Zn levels than other literature reports (Brand et al., 2020; Walker et al., 2011; Mason & Stephenson, 2001). So as far as biota is concerned, there is no problem of Zn contamination. Natural zinc levels in river sediments are between 50 and 90 μg/g. In some samples of the Flix reservoir has Zn concentrations 3 times higher than these values. A high level of contamination from this metal is also observed in the final part of the Ebro River.

Nickel is present in the aquatic environment due to natural weathering and geochemical processes (Schaumloffel, 2005). Anthropogenic activities can also produce Ni and reverse it into the waters through runoff or direct spills near industrial and urban areas or wastewater treatment facilities (Schaumloffel, 2005). Ni is an essential nutrient for plants and terrestrial animals. It has been stated that it could also be an essential nutrient for fish (Pyle & Couture, 2011), but there is no evidence. The toxicity related to Ni may lead to adverse effect in fish, such as branchial lesions, kidney deregulation, and genotoxic effects, among others (Pyle & Couture, 2011). Muyssen et al. (2004) stated a negative relationship between Ni exposure and Ni concentration in fish samples which could be explained by fish Ni regulation in the uptake and elimination processes (Muyssen et al., 2004). This effect probably occurred in the fish samples analysed in this study that show low Ni levels when compared to otter samples. In comparison with the Ni levels of other fish, the fish of the present study have lower Ni levels (Andreji et al., 2005; Yilmaz, 2003). The otter that does not have the Ni purification system of fish, shows high Ni concentrations when compared with other otters (Brand et al., 2020; Walker et al., 2011; Mason & Stephenson, 2001). Ni levels in the sediments of an uncontaminated river are in the order of 20 μg/g (Palanques et al., 1999). The average

level recorded in the Flix reservoir is also more than three times higher than those in these uncontaminated river courses. It shows strong variability between different river sediments that are consistent with inputs originating from the Flix reservoir, as the concentrations are 36 μg/g upstream and 14 μg/g downstream.

The 10% of total cadmium concentration is derived from natural sources (vegetation decaying, forest fire, and airborne soil particles), whereas 90% is derived from anthropogenic activity (phosphate fertilizers, pigments, industrial activities; Kumar & Singh, 2010). Cd is present in the aquatic environment, mostly because of industrial spills (Okada et al., 1997), and gets biomagnified in the food chain. High levels of Cd in fish are a potential risk concern for human consumers' health (Kumar & Singh, 2010). Cadmium is a highly toxic element leading to nephrotoxic, cytotoxic, genotoxic, immunotoxic, and carcinogenic effects (Varma & Jain, 2016; Okocha & Adedeji, 2011). The Cd levels in the fish examined in the current study are lower than in others (Ganjavi et al., 2010; Andreji et al., 2005; Yilmaz, 2003). As for the otter, the Cd levels are in line with others in the literature (Brand et al., 2020; Walker et al., 2011). The concentration levels of Cd in uncontaminated river sediments are in the order of 0.1 μg/g (Palanques et al., 1999). Thus, the concentrations found in the present study are high both in the Flix reservoir and downriver. Their distribution is consistent with the effect of spills from the reservoir downriver.

Lead is sourced from natural and man-made sources. Earth's crust contains lead, which is found in all environments (Cheng & Hu, 2010), including the aquatic one. The anthropogenic activities that produce this element are Pb smelting and mining and the production of paints, cement, and batteries (Kim & Kang, 2016). Lead has a multiplicity of adverse effects on human health (Ishaque et al., 2020). Some problems caused by lead toxicity affect the nervous (Campara et al., 1984; Hogstedt et al., 1983), immune (Gidlow, 2015), cardiac (Lai et al., 1991), reproductive (Telisman et al., 2000), digestive (Sakai, 2000), skeletal (O'Flaherty, 1995), nephrological (Ehrlich et al., 1998) systems, carcinogenicity, and genotoxicity (IARC, 2006). Pb bioaccumulation can be fatal for some aquatic animals (Kim & Kang, 2015). The Pb levels in the fish from the Flix reservoir and the Mediterranean coast examined in this chapter are very low in relation to other locations contaminated with Pb (Has-Schon et al., 2008; Yilmaz, 2003; Rashed et al., 2001). The Pb levels in otters are slightly lower than others reported in the literature (Brand et al., 2020). The Pb concentrations of uncontaminated river sediments are in the

range of 17–30 μg/g (Palanques et al., 1999). The Pb contamination observed in the current study is not as high either in the Flix reservoir or the Ebro sediments, apart from three sediment samples, one from the reservoir and two downstream from downriver. With regard to the latter two sediment samples, Pb concentrations are significantly higher than the others. This probably reflects a local problem for lead contamination, which is also highlighted in the PCA. This local problem may be related to old traffic inputs in Tortosa and Miravet, the two areas nearby the location of the samples. The Flix meander sediment does not show Pb contamination, staying in accordance or below the uncontaminated values (Palanques et al., 1999).

Selenium is an essential element, needed for the prevention of certain diseases and for the production of selenocysteine, a peculiar amino acid. Se can be naturally extracted in many minerals or released as by-product of some industrial processes (Lemly, 1993). If introduced in high quantities, Se leads to toxicity and might play a role in cancer, cardiovascular disease, cognitive decline, and thyroid disease (National Institutes of Health, NIH, 2021). High levels of Se can also lead to adverse effects in fish (Sato et al., 1980). The Se concentrations in fish samples from the Flix reservoir and the Mediterranean are slightly higher than in other studies (Burger et al., 2001), while those of the otter are significantly lower than those evaluated in the literature (Brand et al., 2020; Walker et al., 2011). The Catalan part of the Ebro River does not have a problem related to Se contamination comparing to data reported in other riverine sediment systems (Palanques et al., 1999).

Another essential element analysed is copper. This metal is required as a cofactor of several physiological processes. At high levels, it becomes toxic. Cu is found in natural elements, such as minerals and rocks, but can also be released by industrial activities, such as the production of fertilizers with algaecide, fungicide, molluscicide action and in foundry, plating, steelworks, refinery, mining, and domestic waste emissions (Flemming & Trevors, 1989). The action of Cu is essential for many enzymes. Its toxic absorption can lead to interfere with many cellular processes, some of which are essential for the metabolism and proper neuronal functioning (Gaetke & Chow, 2003). The Cu concentration ranges in the fish samples examined are in line with other fish species and divergent from others (Keskin et al., 2007). This shows a wide variability regarding this metal, which is absorbed differently according to the species. However, compared to other fish in contaminated places, the Cu levels are lower (Yilmaz, 2003), while they agree with those in non-heavily contaminated locations (Andreji et al., 2005). The Cu levels in the

otter of the present study are slightly higher than those observed in some cases (Mason & Stephenson, 2001) and lower than in others (Brand et al., 2020; Walker et al., 2011). Cu levels in the sediments of unpolluted rivers are in the order of 25-40 μg/g (Palanques et al., 1999). Accordingly, the Ebro downriver and Flix meander sediments have normal values, despite some samples of the Flix reservoir exceeding these values but not much.

Arsenic is a widespread metal in terrestrial and aquatic environments because it is released from natural and anthropogenic sources. It is mainly attributable to industrial mining activities, but also to other activities such as agriculture and forestry which have contaminated soil and water on a local scale (Smith et al. 2003). Among the As natural sources, aquifers may contain it in concentrations deleterious for humans (Smedley & Kinniburgh 2002). As is highly toxic to humans (Chowdhury et al. 1999). It has deleterious effects on the nervous, cardiovascular, respiratory, and gastrointestinal systems. It also led to adverse effects on blood, skin, liver, and kidneys. Probably, As has an effect on diabetes-related problems and leads to cancer (Mazumder, 2008). The concentration levels of arsenic in uncontaminated river sediments (Palanques et al., 1999) are between 10-15 μg/g. Therefore, there are some sediment samples analysed that exceed this value, even if the average values approach or are within the ranges of As levels in uncontaminated rivers. Therefore, it can be stated a small problem of As contamination in the Flix reservoir, while it is not in the Ebro downriver. Fish species appear to have evolved defensive mechanisms for the biotransformation of As into less toxic forms, which are then promptly excreted (Kumari et al., 2017; Bears et al. 2006). The fish of the Mediterranean, in comparison with those of the Flix reservoir, show much higher levels of As, from 70 to 129 times greater which reflects a difference between freshwater and marine fish. The latter contains high amounts of arsenobetaine which eliminates the toxicity of this metalloid (Larsen & Francesconi, 2003). The As concentrations found in the marine fish of the present study have very high levels. Also, a very high As concentration is present in the otter when compared with As levels in other studies (Brand et al., 2020; Walker et al., 2011).

Thallium is a very toxic element. It is mainly produced in industrial processes, such as heavy metal refining, manufacturing of electronic components, pharmaceuticals, insecticides, and glass, among others. Its presence in nature is found in several minerals. Thallium poisoning involves first hair loss, damage to peripheral nerves and then death, so much so that it is considered a powerful poison. Thallium appears to be toxic not only to humans and animals in general, but also causes adverse reactions in plants and

microorganisms. Its toxicity derives from the ability to replace the cations in the body. It is also considered a suspected carcinogenic molecule (Zitko, 1975). Fortunately, thallium levels in reservoir and Mediterranean fish are far from the lethality limit (Zitko, 1975) and elevated Tl concentrations in other locations (Fard et al., 2017). Sediments have much lower Tl values than others in contaminated places (Belzile & Chen, 2017; Lis et al., 2003). The otter also has low Tl values compared to other studies (Sanders et al., 2020).

Uranium is an extremely toxic and radioactive element. Radioactive activity linked to the industrial activity of the Flix chlor-alkali complex has been previously reported (Grimalt et al., 2003). Years later, radioactivity has remained virtually unchanged. The uranium levels in the sediment samples and the otter sample are visible. In fish, the uranium levels are nearly 0, showing very low absorption.

The levels of thallium and uranium in the sediments and in the otter sample in the Flix reservoir are very similar which suggests a possible common origin related with the residues of the phosphate that is used for the manufacture of bicalcium phosphate.

The results of the PCA show that all metals except Pb were positively correlated, showing a common origin related with the by-products of the phosphorite use in the chlor-alkali plant and that the mixtures generated are the main metal pollution source downriver. In the PC2 loadings, there is a clear predominance of Pb over all other metals. Looking at the sample loadings and scores of this PC, local inputs are outlined in sites D-b and D-d. These two samples were taken in the vicinity of Miravet and Tortosa, two towns which may have generated Pb contamination due to past traffic activities.

## 2.5. Conclusions

The observed levels of mercury and other metals in the Flix reservoir and downriver show a predominant pollution source from the historical spills of the chlor-alkali plant related with the use of mercury as cathode in the electrolytic process and the phosphate residues in the manufacture of this mineral. The mercury isotope composition confirms the mercury assignment since the MIFs of the odd atomic weight isotopes have very similar values in the sediments of the meander, fish from this area and from the Mediterranean coast nearby the Ebro River mouth, 110 km downriver.

Comparing to literature data, the concentrations of metals in the sediments are very high, namely in the case of chromium, zinc, cadmium and arsenic. The fish from the meander have metal concentrations roughly in line with those of other fish in contaminated locations. The composition of metals in fish from the Mediterranean and

from the Flix meander is similar. Arsenic is the only element showing a strong difference which reflects the different accumulation pattern of this element in freshwater and marine fish.

The otter Hg isotopic signature is very similar to that of fish, suggesting mercury contamination due to the spillage of waste by the chlor-alkali plant in this animal as well.

# CHAPTER 3

## PREDOMINANT MERCURY SOURCES IN FISH FROM THE MEDITERRANEAN SEA

CHAPTER 3

# PREDOMINANT MERCURY SOURCES IN FISH FROM THE MEDITERRANEAN SEA

## 3.1. Introduction

Mercury (Hg) is a ubiquitous and very persistent pollutant. It occupies all environment compartments being present even in the most remote parts of the globe (Wangberg et al., 2016; Gabriel et al., 2005; Poissant et al., 2005; Bargagli et al., 1993). Hg emitted by natural or anthropogenic sources is transported through the atmosphere and deposited everywhere, arriving far away from its source (Johansson et al., 2001).

Hg is deposited in all terrestrial and aquatic environments. It bioaccumulates over the course of time. Thus, mercury is found in consistent quantities in soils (Wang et al., 2020; Zhu et al., 2018), sediments (Ruiz-Fernandez et al., 2019; Ting & Hsi, 2019), air (Liu et al., 2019; Streets et al., 2018), snow (Cairns et al., 2021; Spolaor et al., 2019), water (Kuss et al., 2018) and biota (Blum et al., 2020; Rudd et al., 2018). Due to biomagnification, this pollutant moves from one organism to another. Hence, mercury moves through the food web. Organisms that prey on others absorb a higher amount of Hg, uptaking it from the prey's body. Humans, being at the top of the food chain, have high mercury levels. The most prevalent mercury exposure for humans is due to fish consumption (Junqué et al., 2017; Garí et al., 2013). The high level of mercury in aquatic environments and its consequent passage from one organism to another through the food chain to humans is still a serious problem (Amoatey & Baawain, 2019; Rodrigues et al., 2019; Beldowska et al., 2018).

Mercury like other elements has isotopes with mass-dependent fractionation and mass-independent isotopes, mentioned as MDF and MIF, respectively. These two categories of isotopes could be fractionated by radical-pair spin-selectivity and nuclear volume differences and / or by reaction selectivity based on nuclear mass (Buchachenko et al., 2008; Schauble, 2007).

MDFs are reported with the lowercase Greek letter delta ($\delta$). In the analysis of MDFs, there are mainly 4 forms ($\delta^{199}$Hg, $\delta^{200}$Hg, $\delta^{201}$Hg, $\delta^{202}$Hg), but sometimes there is a fifth one ($\delta^{204}$Hg). The MIF values are indicated with the Greek letter delta in

uppercase (Δ). MIFs mainly come in 3 forms ($\Delta^{199}$Hg, $\Delta^{200}$Hg, $\Delta^{201}$Hg), although sometimes there is a fourth ($\Delta^{204}$Hg).

The ratios of Hg isotopes vary according to the environmental samples (Rutter et al., 2011; Sherman et al., 2010; Carignan et al., 2009; Laffont et al., 2009; Biswas et al., 2008; Smith et al., 2008).

These stable isotopes have been used to evaluate mercury reduction reactions (Kritee et al., 2007), to estimate photoreactions (Bergquist & Blum, 2009), to determine the methylation / demethylation process (Perrot et al., 2013), to obtain information of an ecological nature (Kwon et al., 2013) and to assess the origin of methylmercury (Li et al., 2016). The broadest use is undoubtedly assigned to the ability of the isotopic signature to bind an element to its source. The isotopes are able to trace the sources that contributed to their compositions. Different studies used both MDF and MIF signatures to trace Hg sources (Cransveld et al., 2017; Jiskra et al., 2017; Xu et al., 2016; Foucher et al., 2009). If an organism has a high level of a pollutant, the isotopic composition provides information on the link between the organism in question and the source. So, isotope signatures can give valuable information even in living organisms, such as fish (Gehrke et al., 2011; Gantner et al., 2009).

The Mediterranean Sea laps a large number of nations. It is located between Europe, North Africa, and Asia Minor. It has unique characteristics of its kind because, despite being very large, is semi-closed. It is connected to the Atlantic Ocean to the west, through a natural outlet in the Strait of Gibraltar, while the Suez Passage to the south-east connects the Mediterranean to the Red Sea and then to the Indian Ocean. To the east, it is connected to the Black Sea, via the Dardanelles Strait. Due to the little exchange with the ocean, the tides are very limited. The temperature of Mediterranean water has extremes between 11 and 32° C. Generally, it ranges from 12 ~ 18° C in the winter months, up to 23 ~ 30° C in the summer months, depending on the area. The Mediterranean Sea is divided into two main basins: Western and Eastern. These two basins are ideally separated by the Strait of Sicily which lies between Tunisia and Sicily. The western part is made up of large abyssal plains, while the eastern part is much more rugged and dominated by the Mediterranean ridge. For the purpose of this dissertation, it is only the western part that is described. However, not many differences are found between the western and eastern parts of the Mediterranean in terms of temperatures and biota.

The Western Mediterranean encompasses several seas, such as the Alborán Sea, the Balearic Sea, the Sardinian Sea, the Corsican Sea, the Tyrrhenian Sea and the Ligurian Sea. It has a maximum depth of about 3800 m. In some sections it has a high depth, as in the abyssal plain of the Balearics (2600-2800 m) or in the Trench of the Tyrrhenian (3800 m). However, it also shows long strokes with shallow depths. The seabed is characterized by numerous volcanic reliefs and ridges. It also has active (Marsili) and inactive (Vavilov) volcanoes in its seabed.

Each sea features a variety of distinctive fish and seafood. The Mediterranean Sea due to its geochemical characteristics is able to host more than 700 species of which about 200 are edible. This number is destined to rise for the so-called "alien" species that over the years have settled in it due to global warming, ship transport or following the opening of the Suez Passage. In any case, the Mediterranean Sea supplies fish to over 480 million people (European Environment Agency, EEA, 2016).

Over the decades, the Mediterranean Sea has accumulated a large amount of mercury. This sea has received a massive input of mercury from the industrial revolution to the present day. The sources that influence the Mediterranean are several, divided into natural, industries and bound to atmospheric deposition. Today, the Mediterranean Sea is a hot spot for mercury, and significant levels of this metal have been found in its waters (Horvat et al., 2003), sediments (Ogrinc et al., 2007) and biota (Cinnirella et al., 2019). The very little water exchange of the Mediterranean with the ocean does not help to dispose of the enormous amount of mercury poured into it. The biota and fish species belonging to Mediterranean Sea showed higher Hg concentrations than same fish species in the Atlantic Ocean (Junque et al., 2018; FAO, 1986). The cause of this high mercury level in fish has been debated (Ogrinc et al., 2019; Monperrus et al., 2007; Rajar et al., 2007), but never came to conclusion.

In the present study, an evaluation of total mercury concentration is performed in fish samples collected throughout the western Mediterranean to assess the Hg situation in this sea.

Fish have been used multiple times as sentinel organisms for Hg abundance and uptake (Santos et al., 2021; Adams et al., 2018; Schaefer et al., 2014). Fish are also sentinel organisms for isotope analysis. Nitrogen and carbon are the two types of isotopes most used in these organisms, but there are many studies regarding others. In recent years,

the number of studies using Hg stable isotopes have increased dramatically, especially for investigation of mercury sources (Kwon et al., 2020; Cransveld et al., 2017; Li et al., 2016; Kwon et al., 2012; Perrot et al., 2010). Atmospheric deposition, the background activities and industrial spills are the three main contributors to Hg uptake in fish (Panagos et al., 2021; Perrot et al., 2019; Rudd et al., 2018; Turner et al., 2018; Hammerschmidt & Fitzegerald, 2006; Wiener et al., 2006; Joiris et al., 1995).

However, no one has clearly established to date which sources contributed to the isotope signatures of the marine fish. This information could be very relevant for implementation of measures for reduction of exposure to this metal in human consumers. Identification of these sources could guide the administrations towards implementation of adequate strategies to limit the amount of mercury released and subsequently lower the risk of mercury poisoning. Obviously, this improvement would involve significant socio-economic advantages, such as the lowering of costs related to the treatment of mercury poisoning episodes, greater safety in the consumption of local fish, safer aquatic environments, drop in episodes related to mercury toxicity in humans, animals, fish and seafood.

The main objective of the study presented in this chapter is to propose a method that sheds light on which sources contribute to the Hg present in the fish from the Mediterranean Sea. For this purpose, the isotopic composition, namely MIFs is used. The work is devoted to assess what are the human risks of mercury intake due to fish consumption. Accordingly, fish specimens were obtained from main markets of major cities located in the Mediterranean coast. The proposed methodology is able to distinguish contributions from anthropogenic activities on one hand and contributions from background and atmospheric deposition.

Furthermore, a quantitative analysis of the contributing sources has been developed allowing to estimate the specific percentages of source contribution.

Full evaluation of the information contained in all Hg isotopes was performed with principal component analysis (PCA).

Besides collecting fish from markets, specimens were sampled in the Ebro river meander that belongs to the Sebes Nature reservoir. These samples were collected in an area located 1 km downriver of the waste discharge sites of Flix chlor-alkali plant where mercury-rich waste was dumped for years. These fish were considered as reference

material as they were born and raised in a context of strong influence from the chlor-alkali plant. Comparing the isotopic composition of these fish with those of the Mediterranean Sea may provide relevant information on the qualitative characteristics of the contributions from chlor-alkali plants.

## 3.2 Materials and methods

### 3.2.1 Sample collection and preparation

Fish samples were collected in different sites throughout the Western Mediterranean area. The specific places from which we brought fish were Alacant, L'Ampolla, Ametlla de Mar, Menorca, Mallorca, Eivissa, Flix in Spain, Marseille in France, Genoa, Civitavecchia, and Alghero in Italy. A map reporting the sample locations and the chlor-alkali plants is shown in Figure 3.1.

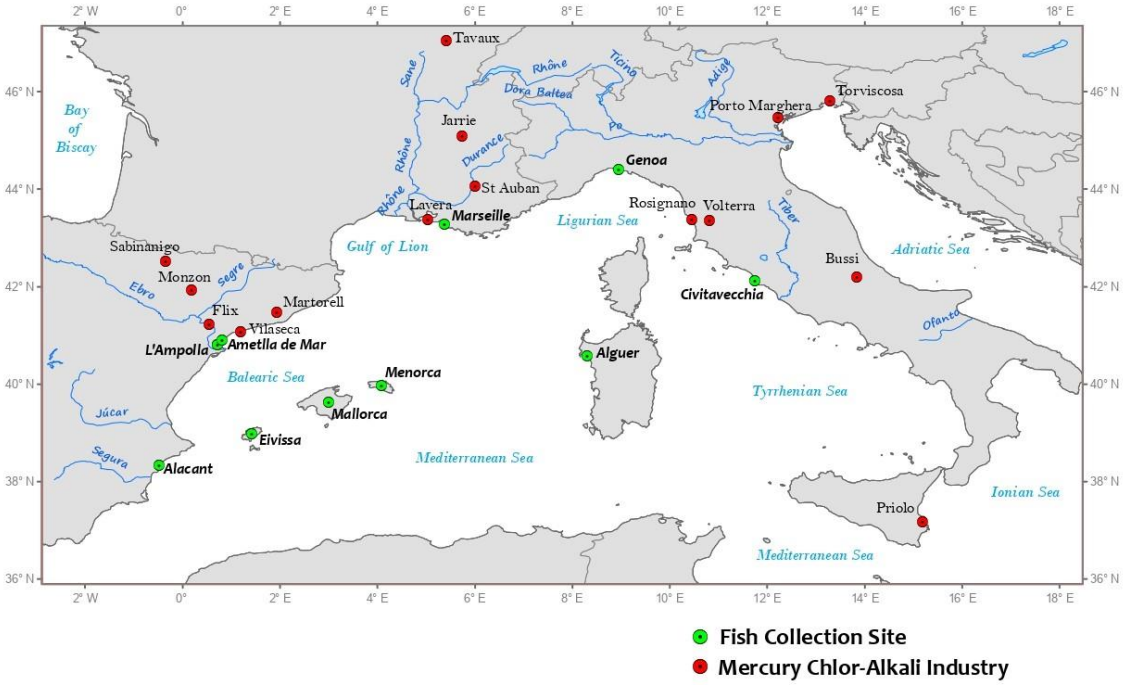


**Figure 3.1.** Map of the Western Mediterranean Sea indicating the locations where fish samples were collected (in green) and the sites having chlor-alkali plants (in red).

The sampling campaigns took place in different months and seasons during the whole year, starting from October 2018 until May 2021. The fresh fish from Mediterranean were collected in local markets and their provenance was certified from

the European Union label. This also allowed to locate the precise area in which these samples were captured. Instead, the Flix samples instead were provided by the staff of the Ebro nature reserve in Flix. They were captured by electro-fishing and recovered with the use of a net. All fish were placed into a portable freezer and taken to the laboratory. A portion of the epaxial muscle, the upper part of the dorsal muscle (approximately 10-15 g), was cut and separated from the bones and skin. These operations were carried out with plastic tweezers and knife. After that, the piece of muscle was placed in an airtight plastic bag and stored in a freezer at -22° C. Between each sample, the tools were thoroughly cleaned with distilled water, soap and rubbed with 96% ethanol (Honeywell, France). Some samples were packed in a box containing dry ice and shipped to the ALS Scandinavia laboratory in Luleå (Sweden) for isotope analysis. To quantify the total concentration of mercury (T-Hg), an additional small part of epaxial muscle (approximately 1 g) was removed, chopped, and placed in a sealed 20-mL glass vial. The vial was then labelled and placed in a freezer at -22° C. Prior the analysis, the samples were thawed. To avoid possible contamination, only ceramic and plastic utensils were used in sample preparation operations. The muscles of the fish in the different preparations were sampled in wet weight.

The fish samples included in this study were angler (*Lophius piscatorius*, n = 14), common seabream (*Pagrus pagrus*, n = 8), Welsh catfish (*Silurus glanis*, n = 2) and Prussian carp (*Carassius gibelio*, n = 1). The species, provenance, latitude, and longitude coordinates of each single fish specimen are described in Table 3.1. Also, the estimated irradiance rate ($W/m^2$) of the sampling sites is included in this Table.

### 3.2.2. Mercury measurements

The tools were thoroughly cleaned with distilled water, soap and wiped with 96% ethanol (Honeywell, France) after each measurement. Portions of 1 g wet weight –ww- were removed, chopped and stowed in sealed 20-mL glass vials for quantification of total mercury. From the vial were taken and measured about 10-20 mg ww aliquots of the sample. It was used an automated atomic absorption spectrometer (Model AMA-254, Altec LTD, Czech Republic) for the total mercury concentration analysis. The samples were introduced inside a combustion tube that was subsequently heated to ~750º C. The generated gases were transported by an oxygen carrier to a catalyst that removed all interfering impurities and then to a glass tube containing gold-plated ceramics that

amalgamated the mercury vapours. This amalgamator was then heated to ~900° C, which released the metal into a spectroscopy cuvette placed in the path length of an atomic absorption spectrometer equipped with an element-specific lamp that emits light at a wavelength of 253.7 nm and a silicon UV diode detector for mercury quantitation. The instrument's detection limit was 0.0009 μg $g^{-1}$ dry weight. Blanks were analysed after each sample to check for possible Hg cross-contamination. The accuracy was checked every 10 samples, using the European Reference Material (ERM-BB422) from the Institute for Reference Materials and Measurements of the European Commission's Joint Research Centre (Geel, Belgium). ERM-BB422 consisted of powdered dry fish muscle and was chosen according to sample Hg concentration levels. The values obtained were always within the ERM-BB422 confidence interval (0.601±0.03 mg $kg^{-1}$).

For isotope measurements, aliquots (18-30 g ww) were packed with dry ice and shipped to ALS Scandinavia laboratory (Luleå; Sweden). The mercury isotope ratios were measured by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS; Neptune Plus; Thermo-Scientific, Waltham, Massachusetts, USA), using external calibration with bracketing isotope standard reference materials (SRMs). Standard deviation of the values was calculated from two independent consequent measurements. The delta values for Hg isotope signatures were computed against NIST SRM 3133.

### 3.2.3. Isotope measurements

The isotope ratios were reported as per-mil deviation from this standard. The mass dependent fractionation (MDF) was defined by the Equation 1.1 (Blum & Bergquist, 2007).

The mass independent fractionation (MIF) was delineated as the deviation of a measured delta value from the theoretically predicted value due to the kinetic MDF according to the Equation 1.2 (Blum & Bergquist, 2007).

### 3.2.4. Similarity Index

Mercury MIFs were used to estimate the similarity of the Hg isotopic composition of the studied specimens with the mean MIFs composition of fish samples collected 1 km downstream from a chlor-alkali plant that was taken as a reference. The overall

differences were calculated from the summed squared differences of each $\Delta^{x}$Hg (‰) as described in Equation 3.1 and were reported as Similarity Index (SI):

$$SI=\sqrt{[(\Delta^{199}Hg_{sample}-\Delta^{199}Hg_{ChlA\ mean})^2+(\Delta^{200}Hg_{sample}-\Delta^{200}Hg_{ChlA\ mean})^2+(\Delta^{201}Hg_{sample}-\Delta^{201}Hg_{ChlA\ mean})^2]}$$

(Equation 3.1)

Low SI values corresponded to compositions close to those generated by chlor-alkali inputs.

### 3.2.5. Data and statistical analysis

Data analysis, graphics, and correlations between MIF values and irradiance and latitude were performed with the statistical software R (R Development Core Team, 2019). Principal component analysis (PCA) of the MIF values were calculated with the Statistical Package for Social Science – SPSS Statistics - software (International Business Machine Corporation, 2019).

**Table 3.1.** Fish species collected and sampling sites indicating the coordinates and irradiance at these locations (in W/m²).

| **Species** | | **Site (Country)** | **Latitude (°N)** | **Longitude (°E)** | **Irradiance (W/m²)** |
|---|---|---|---|---|---|
| Angler /monkfish | *Lophius piscatorius* | Alacant (Spain) | 38.345 | -0.481 | 333.291 |
| Common Seabream | *Pagrus pagrus* | Alacant (Spain) | 38.345 | -0.481 | 333.291 |
| Common Seabream | *Pagrus pagrus* | Alghero (Italy) | 40.565 | 8.318 | 325.188 |
| Angler /monkfish | *Lophius piscatorius* | Alghero (Italy) | 40.565 | 8.318 | 325.188 |
| Angler /monkfish | *Lophius piscatorius* | Ametlla de Mar (Spain) | 40.885 | 0.803 | 324.284 |
| Common Seabream | *Pagrus pagrus* | Balearic Islands (Spain) | 39.999 | 3.958 | 327.254 |
| Angler /monkfish | *Lophius piscatorius* | Balearic Islands (Spain) | 39.999 | 2.646 | 327.254 |
| Angler /monkfish | *Lophius piscatorius* | Civitavecchia (Italy) | 42.092 | 11.797 | 319.614 |
| Common Seabream | *Pagrus pagrus* | Civitavecchia (Italy) | 42.092 | 11.797 | 319.614 |
| Welsh catfish | *Silurus glanis* | Flix meander (Spain) | 41.231 | 0.544 | 322.757 |
| Prussian carp | *Carassius gibelio* | Flix meander (Spain) | 41.231 | 0.544 | 322.757 |
| Angler /monkfish | *Lophius piscatorius* | Genoa (Italy) | 44.414 | 8.915 | 311.139 |
| Common Seabream | *Pagrus pagrus* | Genoa (Italy) | 44.414 | 8.915 | 311.139 |
| Angler /monkfish | *Lophius piscatorius* | L'Ampolla (Spain) | 40.813 | 0.709 | 324.282 |
| Common Seabream | *Pagrus pagrus* | Marseille (France) | 43.296 | 5.376 | 315.220 |
| Angler /monkfish | *Lophius piscatorius* | Marseille (France) | 43.296 | 5.376 | 315.220 |

[a]See Figure 3.1 for locations. [b]Similarity indices between the mass independent fractionations of the Hg isotopes of the studied specimens and fish located 1 km downriver of a reference chlor-alkali plant (Equation 3.1). The specimens are ordered according to this index from the highest to the lowest.

## 3.3 Results

### 3.3.1 Total mercury concentrations

The total mercury concentrations in the samples analysed are summarized in Table 3.2. Generally, the values are high. Among the 14 samples of angler, 10 exceeded the threshold of 1 mg/Kg ww set by the European Commission as suitable for human consumption. The locations of the fish that revealed mercury concentrations above this threshold were distributed throughout the Western Mediterranean. Concerning the common seabream, two of them exceeded the above mentioned threshold. In summary, 12 of the 22 specimens analysed, 55%, exceeded the mercury threshold for human consumption.

### 3.3.2 Isotopic analysis

The sample mass-dependent and mass-independent fractionation ratios are described in Table 3.3. The trend of the $\Delta^{199}$Hg and $\Delta^{201}$Hg levels can be correlated (with $R^2 = 0.98$), while that of the $\Delta^{200}$Hg isotope has a high variability and much lower values. The values of $\Delta^{199}$Hg and $\Delta^{201}$Hg isotopes in the samples from the Mediterranean range respectively from 0.203 to 1.089 per-mil (‰) and from 0.231 to 1.023‰. The $\Delta^{200}$Hg has values between -0.055 and 0.072‰.

The $\delta^{199}$Hg shows values ranging from 0.287 and 1.261‰, while the $\delta^{201}$Hg from 0.178 to 1.574‰. The values of the isotope $\delta^{200}$Hg vary from -0.108 to 0.554‰ and the ones of the isotope $\delta^{202}$Hg from -0.181 to 0.971‰. A significant correlation trend was found between the MDF of the odd numbered isotopes ($R^2 = 0.87$) and the MDF of the even numbered isotopes ($R^2 = 0.97$).

The higher correlation for the MDFs of the even numbered isotopes reflects the larger span of values in these compounds, encompassing from negative values to almost 1‰, that is a range of 1.152‰, whereas in the odd numbered isotopes the variation range encompasses 0.974‰.

**Table 3.3.** Mercury isotope composition in the fish specimens examined.

| Sample ID | | Hg stable isotope signatures (‰) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Sample site** | **Sample specimen** | **$\delta^{199}$Hg** | **$\delta^{200}$Hg** | **$\delta^{201}$Hg** | **$\delta^{202}$Hg** | **$\Delta^{199}$Hg** | **$\Delta^{200}$Hg** | **$\Delta^{201}$Hg** |
| Alacant | *Lophius piscatorius* | 0.870 | 0.243 | 0.899 | 0.341 | 0.784 | 0.072 | 0.642 |
| Alacant | *Pagrus pagrus* | 0.710 | 0.134 | 0.873 | 0.277 | 0.640 | -0.005 | 0.665 |
| Alacant | *Lophius piscatorius* | 0.481 | 0.061 | 0.461 | 0.068 | 0.464 | 0.027 | 0.410 |
| Alghero | *Pagrus pagrus* | 1.060 | 0.554 | 1.547 | 0.971 | 0.815 | 0.066 | 0.817 |
| Alghero | *Lophius piscatorius* | 0.896 | 0.102 | 0.932 | 0.230 | 0.838 | -0.013 | 0.759 |
| Ametlla de Mar | *Lophius piscatorius* | 0.331 | -0.098 | 0.178 | -0.152 | 0.370 | -0.022 | 0.292 |
| Balearic Islands | *Pagrus pagrus* | 1.261 | 0.354 | 1.537 | 0.683 | 1.089 | 0.011 | 1.023 |
| Balearic Islands | *Pagrus pagrus* | 1.210 | 0.428 | 1.574 | 0.770 | 1.016 | 0.041 | 0.995 |
| Balearic Islands | *Lophius piscatorius* | 1.161 | 0.169 | 1.128 | 0.387 | 1.063 | -0.026 | 0.838 |
| Balearic Islands | *Lophius piscatorius* | 1.099 | 0.164 | 1.131 | 0.378 | 1.004 | -0.026 | 0.847 |
| Balearic Islands | *Lophius piscatorius* | 0.972 | 0.150 | 1.027 | 0.408 | 0.869 | -0.055 | 0.720 |
| Civitavecchia | *Lophius piscatorius* | 0.580 | 0.210 | 0.634 | 0.283 | 0.509 | 0.067 | 0.422 |
| Civitavecchia | *Lophius piscatorius* | 0.585 | 0.242 | 0.713 | 0.398 | 0.485 | 0.042 | 0.414 |
| Civitavecchia | *Pagrus pagrus* | 0.365 | 0.362 | 0.715 | 0.643 | 0.203 | 0.039 | 0.231 |
| Genoa | *Lophius piscatorius* | 0.824 | 0.159 | 0.827 | 0.269 | 0.756 | 0.024 | 0.625 |
| Genoa | *Pagrus pagrus* | 0.795 | 0.358 | 1.130 | 0.687 | 0.622 | 0.013 | 0.613 |
| Genoa | *Lophius piscatorius* | 0.592 | 0.192 | 0.635 | 0.274 | 0.523 | 0.054 | 0.429 |
| L'Ampolla | *Lophius piscatorius* | 0.494 | 0.118 | 0.526 | 0.224 | 0.437 | 0.006 | 0.358 |
| Marseille | *Pagrus pagrus* | 0.419 | 0.108 | 0.554 | 0.221 | 0.363 | -0.003 | 0.388 |
| Marseille | *Lophius piscatorius* | 0.377 | -0.010 | 0.323 | -0.011 | 0.380 | -0.004 | 0.331 |
| Marseille | *Lophius piscatorius* | 0.322 | -0.108 | 0.182 | -0.181 | 0.367 | -0.017 | 0.318 |
| Marseille | *Pagrus pagrus* | 0.287 | -0.068 | 0.184 | -0.120 | 0.317 | -0.008 | 0.274 |

### 3.3.3. Similarity Index

The Similarity Index (SI) values were calculated for assessment of the qualitative differences between the isotopic composition of the samples analysed and the isotopic composition of the fish specimens collected downstream of the waste discharge site of a chlor-alkali plant. These indices were calculated using Equation 3.1 and are reported in Table 3.2.

The index recorded very low values for the fish specimens collected in Ametlla de Mar, 0.036, and l'Ampolla, 0.124, which *a priori* could be expected having in mind that these specimens belonged to an area under the influence of the Ebro River where the reference chlor-alkali plant is located, 100 km upstream (Flix reservoir). These low values reflect that the chlor-alkali Hg inputs have effectively an influence in the open sea area around the river being accumulated in the fish species studied. This result is consistent with previous findings of mercury present in the suspended particles in the water of the river (Palanques et al., 2020) or in freshwater fish river species, such as catfish (*Silurus glanis*) and common carp (*Cyprinus carpio*), 1.3±0.9 µg/g and 0.35±0.31 µg/g, respectively (Carrasco et al., 2011).

Furthermore, these indices also show very low values for fish collected in Marseille, 0.024-0.102, indicating a strong affinity with the MIF inputs of chlor-alkali plants. These fish were collected in the area of influence of the Rhône river which also has received mercury inputs from these installations.

The sites located in the central western Mediterranean, Balearic Islands and Alghero, are those exhibiting highest values, 0.687-1.052 and 0.688-0.718, respectively, indicating highest difference from chlor-alkali plant inputs.

### 3.3.4. Principal Component Analysis

Further insight into the information contained in these MIF has been elucidated by principal component analysis (PCA) of the samples examined. The loadings resulting from this calculation are shown in Figure 3.2A together with the corresponding scores. PC1 shows a strong dominance of $\Delta^{199}$Hg and $\Delta^{201}$Hg, the two odd numbered isotopes, whereas PC2 is largely dominated by $\Delta^{200}$Hg. The variance explained by these two PCs,

67.6% and 31.5%, respectively, (99.1% in total) describes well the variability of these isotopes.

The scores corresponding to PC1 are roughly distributed in a mode paralleling the above mentioned SI differences (Figure 3.2B). Thus, the fish samples from the chlor-alkali plant, Ametlla de Mar, L'Ampolla and Marseille group in one area and those from the Balearic Islands and Alghero in the other. These fish specimens from Civitavecchia are also closer to those from the specimens from chlor-alkali plants, as well as one sample from Alacant and another from Genoa.

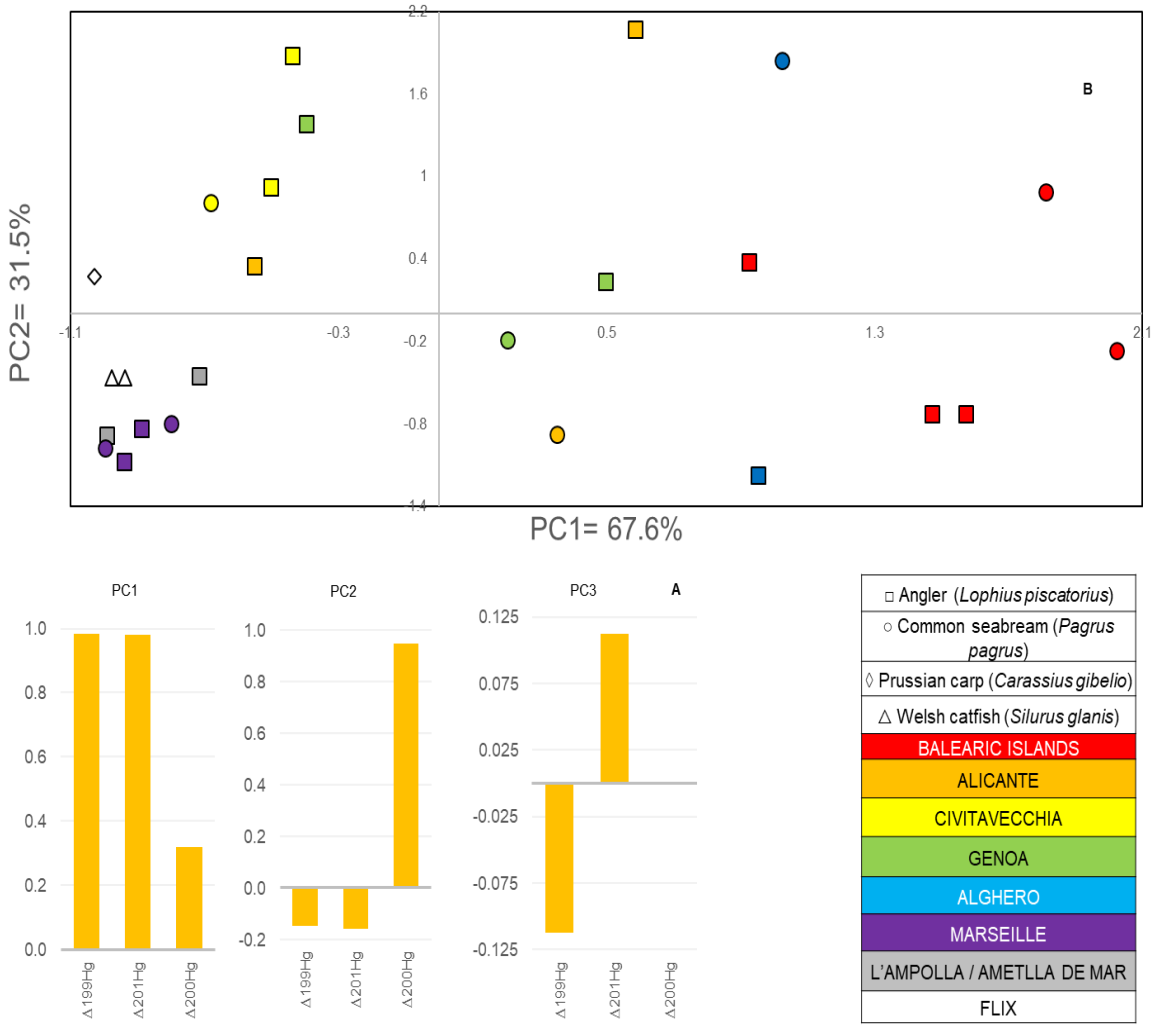


**Figure 3.2.** Loadings and scores of the Principal Component Analysis of the composition of $\Delta^{199}$Hg, $\Delta^{200}$Hg and $\Delta^{201}$Hg in the fish specimens analysed.

### 3.4 Discussion

In all papers that reported Hg MIF levels in fish, there is a correlation between the isotopes $\Delta^{199}$Hg and $\Delta^{201}$Hg that approaches $R^2 = 1$, as in the present study. The MIF values of the samples are higher (Bonsignore et al., 2020) or approach those of other works on fish in the Mediterranean Sea (Cransveld et al., 2017; Bonsignore et al., 2015). This variability of the results can be explained by the different species analysed and by the different place where the study was conducted. The position is in fact a discriminating feature in this type of analysis, since different locations can be influenced by a number and by a type of different factors (natural or anthropological contribution).

There are notable interesting results, analysing the Similarity Index (SI) value. Specimens that have a close direct inlet of mercury-rich waste have lower SI value (Table 3.2). Samples collected in Marseille, Ametlla de Mar and L'Ampolla are attesting with values closer to 0 and therefore more similar to those of fish that have been fully influenced by the spills of the chlor-alkali plants. These places have one thing in common: they are all in the vicinity of a river mouth that have received mercury-rich wastes from chlor-alkali plants downstream. Therefore, the MIFs of Marseille fish samples are consistent with chlor-alkali inputs from the coast of the Rhone River (Rajar et al., 2007; Cossa & Martin, 1991; Vernet & Thomas, 1972), which collects waste from these installations in the South of France (Figure 3.1). L'Ampolla and Ametlla de Mar fish were located a few kilometres from the delta of the Ebro River, which is notorious for the huge amount of mercury-rich waste emitted by a chlor-alkali plant (Palanques et al., 2020; Carrasco et al., 2011; Soto et al. al., 2011; Carrasco et al., 2008; Chapter 2). Therefore, it can be stated that the input of the spills of the chlor-alkali plants gives a similar MIF isotopic signature in the fish of the Western Mediterranean Sea. The proximity to a source which is spilling mercury-rich waste results in a well-defined isotopic signature with regard to isotopes $\Delta^{199}$Hg and $\Delta^{201}$Hg. Although they were taken at different times of the year and belong to two very different fish species, the odd MIF isotopic signatures in these samples are very similar. There are samples in Civitavecchia, which may have been indirectly affected by a nearby plants and by a disused mercury mine (Baldi & D'Amato, 1986; Baldi & Bargagli, 1984; Breder & Flucht, 1984). The isotopic signatures from Civitavecchia samples have two potential direct anthropogenic sources. The disused mercury mine of Monte Amiata still today pours a small quantity of mercury into the Tiber River, which subsequently flows into the sea, not very far from the city of

Civitavecchia. Further North, there are the industrial complexes of Volterra and Rosignano Solvay. The latter has been the subject of study for mercury release several times (Gibicar et al., 2009; Ferrara et al., 2001; Maserti & Ferrara, 1991). The contribution of the industrial complexes and the disused mine could explain the low SI value in Civitavecchia samples, which are very similar to those of L'Ampolla, Ametlla de Mar, and Marseille. The samples from Genoa, Alacant, Alghero, and the Balearic Islands are not influenced by direct sources of anthropogenic contributions. They are the samples that have a higher SI value. The highest SI values are recorded in fish samples collected in the Western Mediterranean islands, Sardinia and Balearic Islands. The samples from Genoa and Alacant, although far from direct anthropogenic sources, have lower SI values than those of the islands. It could be assessed that the remoteness from direct sources has led to higher SI values. Alghero and the Balearic Islands are located in an isolated geographical context. They also show similar MIF values between the samples analysed. However, it has to be ascertained whether or not these isotopic distributions could reflect chlor-alkali inputs despite their geographical location.

In PCA, the principal component-1 (PC-1) and the principal component 2 (PC-2) are the components with the greatest strength, explaining the variability of the Hg isotopic composition by 99.1%. Putting in a scatterplot and comparing these two PC shows the contribution of the individual samples into the phenomenon. Fish from the Ebro river was included with the specimens from the Western Mediterranean to see where they fit within the distribution of values. The fish distribution for these two PC is varied (Figure 3.2B) but can be grouped into some well-defined clusters. For example, it is possible to see a first cluster in the third quadrant in which all those samples that have been directly influenced by the input of the chlor-alkali plants are grouped. Here, not only two Ebro river specimens are found, but also the Marseille, L'Ampolla and Ametlla de Mar samples. Not far from the third quadrant, in the fourth quadrant there is the last sample from Flix, followed by the Civitavecchia samples and some specimens from Alacant and Genoa. On the right side of the scatterplot, it can be found a second cluster given by all those samples from Mediterranean islands. They have the highest values in the SI and in the percentage of inputs due to atmospheric deposition and background. It is really surprising how the specimens directly-influenced by a source are placed within a specific cluster and how those not directly-influenced deviate. These data further reinforce what has been described above and provide further importance to the Hg MIF

signatures, since they are fundamental for the reconstruction of the contributions coming from the different sources.

In Figure 3.3, a correlation between MIFs from fish of remote sites and the irradiance rate or latitude is represented taking the information reported in the literature (Motta et al., 2020; Madigan et al., 2018; Blum et al., 2013; Gantner et al., 2009). The represented irradiance is the one found in the upper atmosphere layer at the coordinates of the collected fish. The graphs show a strong correlation between irradiance or latitude and the MIF of the collected fish specimens. The correlations between latitude and irradiance are very significant ($p < 0.0005$) and those between odd-MIFs and irradiance or latitude as well ($p < 0.0005$ for both $\Delta^{199}Hg$ and $\Delta^{201}Hg$; $R^2 = 0.5901$ and 0.6123, respectively).

Accordingly, introducing the amount of irradiance in the place where the Mediterranean fish sample was captured to the equation obtained in the correlation among irradiance and isotopic signature, a numerical value was obtained. This value was then adjusted with the median of the Hg MIF signatures in fish captured in Ebro river, 1 kilometre downriver from the chlor-alkali implant. The value thus computed was the reference value used for the calculation of the source contribution percentage for atmospheric deposition plus background inputs, as follows:

$$Input_{atmospheric\ deposition+background}\ (\%) = (\Delta^{x}Hg_{sample} / \Delta^{x}Hg_{ref.value}) \times 100 \qquad \text{(Equation 3.2)}$$

where x is the isotope mass.

The percentage of the contribution provided by the chlor-alkali factories was obtained by subtracting from the total the amount of the atmospheric deposition plus the background contribution in percentage:

$$Input_{chlor\text{-}alkali\ plants}\ (\%) = 100 - Input_{atmospheric\ deposition+background}\ (\%) \qquad \text{(Equation 3.3)}$$

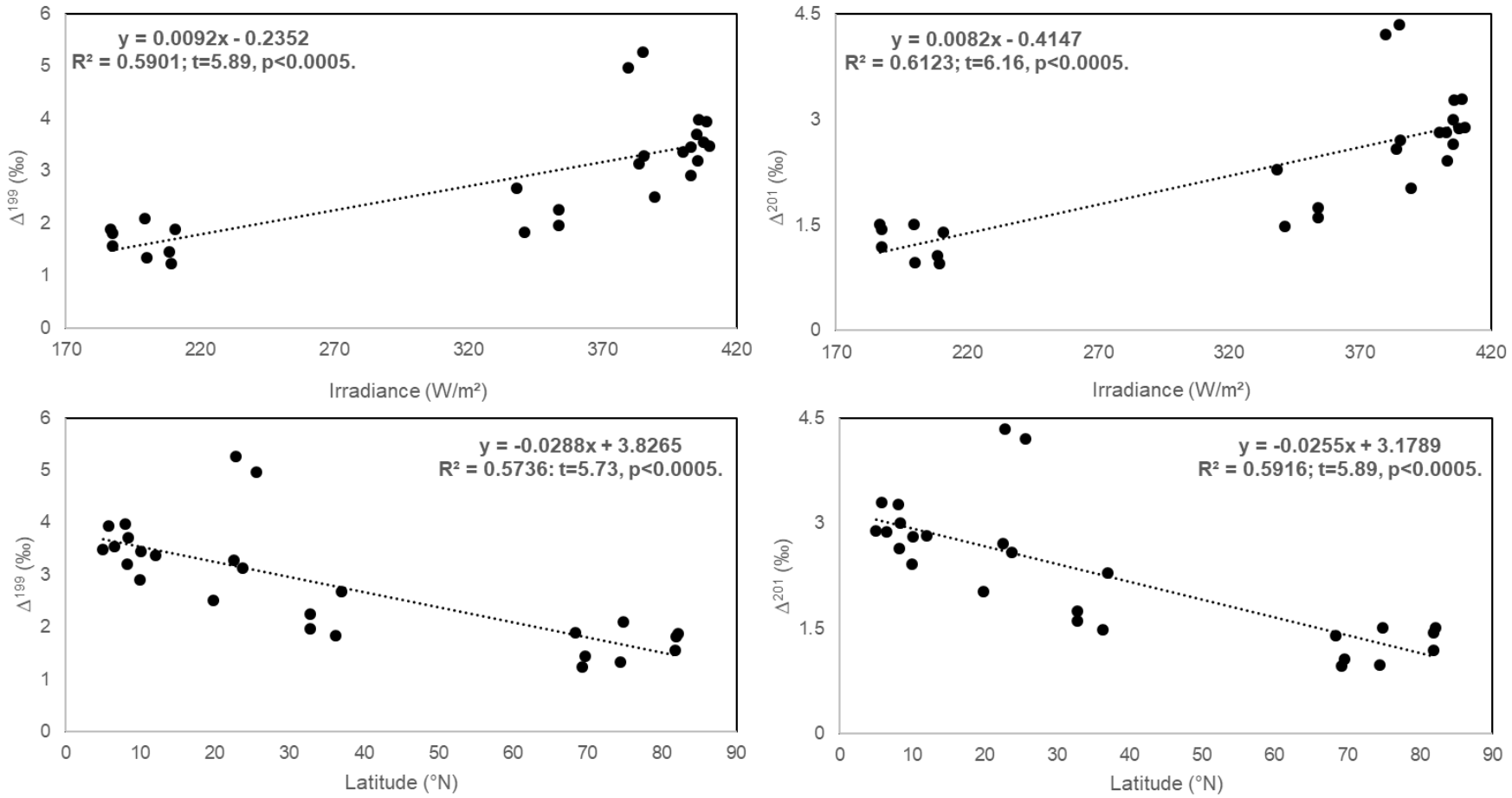


**Figure 3.3.** Correlation between Hg-odd MIF compositions and the irradiance rate (first row) and latitude (second row) in fish from a context far from any anthropogenic input. In each scatterplot is reported the equation of the distribution, the coefficient of determination, the t-value and p-value derived from the t-test.

The percentages of source contributions on individual samples were calculated, using Equations 3.2 and 3.3 and reported in Table 3.2. The inputs provided by atmospheric deposition and background reach 37% in the specimens from the Balearic Islands, which are those with the higher percentage in the Western Mediterranean and gradually decrease for all other specimens, up to 0% in three samples coming from Civitavecchia, Ametlla de Mar and Marseille respectively. The average contribution of atmospheric deposition and background on Western Mediterranean Sea samples is 12-14%, varying from 0% to 31-37%.

The percentages of the inputs due to the spills of the chlor-alkali plants are predominant with respect to the inputs of the atmospheric deposition and the background in Mediterranean fish. In some samples, 100% of the contribution is reached. Above 63% of chlor-alkali inputs are reported in all samples. The average value of the percentage of the contribution of chlor-alkali plants on Western Mediterranean fish is between 86 and 88%. The range is from 63-69% up to 100%.

Therefore, knowing the extent of the contribution coming from the atmospheric deposition and the background, it is drawn a clear and complete picture of the sources that act on the fish samples of the Western Mediterranean. Knowing exactly the value of the contribution of atmospheric deposition and background on individual samples reveals how much they affect total mercury concentrations. The hand of man is also present in atmospheric deposition as a very large number of industrial activities release a high quantity of mercury into the Earth's atmosphere. The rates of mercury emission from anthropogenic activities appear to decrease in Europe and North America through long-term observations of total Hg concentrations into the atmosphere (Slemr et al., 2003). However, the worldwide trend in anthropogenic mercury emission rates is increasing due to the increased amount of coal burning in developing countries (Pacyna et al., 2006; Jaffe et al., 2005). There is therefore a global trend in increasing mercury deposition, in which 68-86% is due to the hand of man, but only 14-32% due to natural resources (Sunderland et al., 2008).

## 3.5. Conclusions

The Mediterranean Sea can be considered a reference scenario for future studies on mercury. Due to the complex biogeochemistry, linking the mercury found in fish intended for human consumption to the sources that release it will be of great interest. Natural and anthropogenic sources simultaneously affect the composition of mercury in fish but knowing exactly to what extent they contribute to individual samples provides useful data for a more comprehensive assessment. It will be possible to evaluate the isotopic signature of a sample anywhere in the Western Mediterranean and understand which source acts on this sample and to what extent. Thus, it will be possible to act to minimize the release of mercury into the environment by industrial complexes and to discuss strategies to reduce the amount of mercury in a given environment.

The high contribution given by the direct spills of chlor-alkali plants in the Mediterranean Sea is certainly a problem that must be solved. First of all, the population could be made aware of this widely debated but still current issue. Secondly, a series of interventions aimed at lowering emissions by companies could be initiated. In fact, these emissions, even if they fall within the limits imposed by the agencies that regulate them, pose serious problems such as the one discussed in this chapter.

The contribution of atmospheric deposition is not to be underestimated. Many actions could be taken to reduce annual mercury emissions into the atmosphere. Surely an intervention that would bring great benefits is to try to regulate the activities that emit mercury into the atmosphere in developing countries. A regulation on a global scale should be required for industries that burn coal or produce cement and steel, melt non-ferrous metals, dispose and incinerate waste.

In an environment like the Mediterranean Sea, it is certainly difficult to act on the background levels of mercury, but it is not impossible. To date, no studies have proposed an effective method for lowering mercury levels in fish. However, recent studies have reported substantial successes in trapping and removing mercury present in the aquatic environment and in the water itself, thus lowering the Hg availability (Ackerman et al., 2019; Tunsu & Wickman, 2018; Jadàn-Piedra et al. al., 2017). One of these strategies could be used to decrease the current concentration of mercury present in the Mediterranean Sea.

Much progress has been made in the fight against mercury. Most European and North American countries have significantly lowered the release rates of this pollutant. Unfortunately, this trend is not global, and many countries have even increased the release of mercury into the atmosphere due to expanding industrial activities. Action should certainly be taken to find a global common line to lower the amount of mercury in the world. Equal regulation for all countries of the globe would be a big step forward. We all still have images of the Minamata disaster in our eyes. If we do not want that this happens again, we need to act in one direction only. Much can still be done to try to eradicate this problem, which is still current and dangerous.

# CHAPTER 4

# OVERALL BURDEN OF MERCURY IN FISH FROM THE WESTERN MEDITERRANEAN SEA. EVALUATION OF RISKY AND SAFE FISH SPECIES FOR HUMAN HEALTH CONCERNING MERCURY CONCENTRATIONS

# CHAPTER 4

# OVERALL BURDEN OF MERCURY IN FISH FROM THE WESTERN MEDITERRANEAN SEA. EVALUATION OF RISKY AND SAFE FISH SPECIES FOR HUMAN HEALTH CONCERNING MERCURY CONCENTRATIONS

## 4.1. Introduction

As described in the previous chapters, once mercury has been released into the environment, it can be transported through the atmosphere by diverse processes of evaporation and condensation which result in a final accumulation in water bodies, lakes, and mainly in the sea, because it is insoluble in water. It may also accumulate in very organic-rich soils such as those from the Scandinavian Peninsula.

Mercury is present in different forms: elemental or metallic ($Hg^0$), inorganic ($Hg^{2+}$), and organic (methylmercury, MeHg, and dimethylmercury). Each of these can be absorbed by the human body, but the most toxic form for humans and organisms is MeHg.

Hg has a harmful impact on the ecosystems and is able to cause several adverse effects in humans. It primarily induces neurotoxic effects, but it can also cause problems with kidneys, lungs, and cardiovascular system (Genchi et al., 2017; Ha et al., 2017; Bernhoft, 2012; Karagas et al., 2012; Park & Zheng, 2012; Grandjean & Herz, 2011; Bose-O'Reilly et al., 2010). Hg intake also involves problems at the immunological and endocrinal level (Gardner & Nyland, 2016; Hyman, 2004; Zhu et al., 2000).

In fact, mercury acts on the three major integrative and regulatory systems of the human body. That is the immune, endocrine, and nervous systems (Zhu et al., 2000).

*Immune system.* All forms of mercury have an immunosuppressive effect. Studies in vitro have shown that it leads to decreases in calcium signalling in lymphocyte membranes (Thompson et al. 1998), decreases in the production of T cells (Shenker et al., 1992), decreases in B cells and immunoglobulins-G and -M (Shenker et al. 1993). Studies in vivo with mice have shown that it is inductive of autoimmune diseases (Havarinasab et al., 2005). Mercury has the ability to alter gene expression by making epigenetic changes. In fact, mercury involves DNA methylation and demethylation, histone hypoacetylation and hyperacetylation, and the expression of certain noncoding

RNAs. All of these alterations can affect gene regulation and result in impaired responses of the immune system (Gardner & Nyland, 2016).

*Endocrine system.* Hormones are secretory products of the endocrine glands. They are transported through the blood to exert their effects on distant target tissues or organs by binding to specific receptors. They are responsible for maintaining homeostasis, reproduction, development, and behaviour. Hormonal destructive agents interfere with the synthesis, storage, release, transport, metabolism, binding, action, or elimination of hormones in the body. In animal studies, mercury has been found to be a potent hormonal destructive agent. In various animal models, it has been observed to have the potential to act on the hypothalamus (Lamperti & Niewenhuis, 1976), the pituitary gland (Nylander & Weiner, 1989; Moller-Madsen & Thorlacius-Ussing, 1986), the thyroid (Ghosh & Bhattachara, 1992; Sin & Teh, 1992), the adrenal gland (Rasmussen & Thorlacius-Ussing, 1987; Veltman & Maines, 1986), and the gonads (ovaries and testis; Mukherjee et al., 1994; Ng & Liu, 1990; Stadnicka, 1980). However, the physiological differences between species make it difficult to compare the results between animals and humans. Mercury studies on the human endocrine system are very limited.

*Nervous system.* Hg is mainly a strong neurotoxin. It has effects on neurodevelopment in children and it causes episodes of neurotoxicity in adults (Bjorklund et al., 2017; Llop et al., 2015; Aschner & Aschner, 1990; Chang, 1977). The effects of mercury at the molecular and cellular level in the nervous system have been extensively described. Mercury enters the body mainly in the form of MeHg. MeHg demethylation occurs in glial cells. Hg is then transferred to neurons, contributing to neurotoxicity (Syversen & Kaur, 2012). Furthermore, both MeHg and $Hg^{2+}$ have a strong affinity for thiol groups (-SH). These groups are required by several subcellular constituents in order to perform their function correctly (Risher & Tucker, 2017). Hg can then bind to them in proteins or membranes. Once bound to the thiol groups of proteins or membranes, the normal physiology of these molecules is disrupted, leading to an attenuation or complete block of their functions (Ynalvez et al., 2016). Furthermore, other alterations caused by MeHg are oxidative stress (Yin et al., 2007; Garg & Chang, 2006; Ou et al., 1999), damage to $Ca^{2+}$ homeostasis (Dreiem & Seegal, 2007) and glutamate dysfunction (Yin et al., 2007; Farina et al., 2003; Ou et al., 1999).

Attention must be paid to the intake of mercury in all its forms. Metallic mercury is used in dentistry. Alloys composed of mercury and other metals are applied to patients

to treat tooth decay. Mercury released from dental fillings may cross the epithelial barrier and dissolve in fluid tissues and blood. From here, it can then be transported to other areas of the body and can cross the blood-brain barrier (Maas et al., 1996). Metallic mercury is also found in a number of manufacturing processes, e.g. thermometers, manometers, and fluorescent light bulbs, although there is a trend to substitute these devices with other materials. Once they are broken, mercury can be released. Therefore, it is essential to dispose of them in the right way to avoid significant releases of $Hg^0$ into the environment (Aucott et al., 2003).

Another massive use of elemental mercury is in the artisanal gold mining. In this practice, the mineral ore is mixed with liquid mercury, creating a gold-mercury amalgam. After that, the amalgam is heated until the mercury evaporates and leaves the gold. These reactions often occur without any protection for both the personnel carrying out the entire process, and for the environment that is enriched with mercury, up to the point of contaminating waterways. In contaminated rivers, lakes, seas, and ocean coasts $Hg^0$ is converted into MeHg and becomes available for fish to be consumed by local people (Castilhos et al., 2017; Bose-O'Reilly et al., 2016). In the human body, $Hg^0$ is oxidized to $Hg^{2+}$ through the catalase–hydrogen peroxide pathway.

Mercury is also emitted to the atmosphere as consequence of the burning of coal and crude oil that contains this metal in small concentrations. Despite the low contents of this metal in these fuels, the massive amounts combusted involve the release of large amounts to the atmosphere and the spread of it throughout the planet.

Inorganic mercury is not absorbed significantly by humans. The use of skin lightening creams that contain mercuric chloride is the most likely way to absorb this form of mercury. $Hg^{2+}$ reacts with reduced glutathione (GSH) in the liver and is transported to the kidney, where it is secreted. However, a small part of $Hg^{2+}$ is not excreted (Wei et al., 1999). This process leads to the accumulation of mercury in the kidneys and damages these organs.

MeHg is undoubtedly the most abundant organic form of mercury. Microorganisms in the aquatic environment transform $Hg^0$ or $Hg^{2+}$ into MeHg (Harris et al., 2007; Mason et al., 2005). MeHg is accumulated along the lifetime of an organism and is transported through the aquatic food web by biomagnification. Accordingly, the main source of MeHg human exposure is the fish consumption (Junqué et al., 2017; Garí

et al., 2013). Fish taken from Hg-contaminated contexts have high amounts of MeHg. Predatory fish and mammals are the aquatic species that have this compound in largest abundance. The global average daily intake of MeHg for humans is 2.4 μg / person, although this quantity is an estimate that change considerably depending on the diet of the different populations (UNEP, 2002). About 95% of the MeHg in fish muscle consumed is absorbed in the human gastrointestinal tract (WHO, 1990). MeHg can quickly cross biological barriers, reaching any organ. MeHg is then demethylated into $Hg^{2+}$ in the liver, brain, and phagocytic cell populations. Once transformed into $Hg^{2+}$ in the brain and other tissues, it is extremely persistent (Suda & Hirayama 1992; Suda et al. 1992).

Almost the totality of the mercury present in fish is in the form of MeHg (Lescord et al., 2018; NIVA, 2017; Mieiro et al., 2009; Latif et al., 2001; Bloom, 1992).

Fish and seafood are essential foods for a healthy and balanced diet. They are consumed all over the world and for some populations they are the main protein source. FAO in 2020 stated that fish is the healthiest foods on the planet and the least impacting on the natural environment. Fish consumption in 2017 was around 20.3 kg per person worldwide and the first estimates for the following years show a small increase (FAO, 2020). Indeed, fish contains proteins with high nutritional power and high digestibility, and polyunsaturated fatty acids, such as omega-3 and omega-6, which help in the absorption of vitamins A and D, reduce blood cholesterol levels, regulate heart rhythm, help foetal development and have a neuro-protective effect. It also contains phosphorus, calcium and iodine that are needed for development and metabolism functionalities. However, fish is the food that provides humans with the greatest amount of Hg. Therefore, the consumption of fish must be monitored. Among the foods introduced with the diet, fish and seafood provide 93% to 98% of the total Hg absorbed (Abass et al., 2018; Junque et al., 2017; Jenssen et al., 2012). There is a significant positive correlation between absorbed mercury levels and dietary consumption of fish (Garí et al., 2013). The effects of mercury toxicity are primarily noticed on sensitive categories of individuals, such as children and pregnant women (Reuben et al., 2020; Santos-Lima et al., 2020; Stratakis et al., 2020; Gonzalez et al., 2019; Gump et al., Al., 2017). However, adults are not exempt from mercury neurotoxicity (Bernhoft, 2012; Grandjean & Herz, 2011; Bose-O'Reilly et al., 2010).

Several organizations have proposed mercury intake thresholds to avoid the negative effects of poisoning by this metal. The European Food Safety Authority (EFSA), the Food and Drug Administration (FDA), the World Health Organization (WHO), the Environmental Protection Agency (EPA) and the United Nations Organization for the food and agriculture (FAO) have issued communications, warnings, advice, suggestions on the consumption of fresh fish. Among all the threshold values reported by the various institutions and agencies, the European Commission has outlined a maximum safety limit for the consumption of mercury set at 1.0 mg $kg^{-1}$, wet weight (ww) for some fish species and a level of 0.5 mg $kg^{-1}$ ww for most fish species. The EPA has set a reference dose (concentration of mercury ingested in the daily diet with no adverse health effects for humans) at 0.1 μg of mercury per kilogram of body weight per day. However, the Environmental Working Group has shown that the negative effects of taking Hg can occur even at doses lower than these recommendations (EWG, 2016).

The Mediterranean Sea, as outlined in Chapter 3, is a well-known hotspot for mercury concentration (Rajar et al., 2007, Ogrinc et al., 2007). Its conformation and history have resulted in high concentrations of mercury throughout its biota (Cinnirella et al., 2019, Llull et al., 2017, Brambilla et al., 2013). Environmental pollution is high where industrial activities use Hg in production processes (Covelli et al., 2009). In Chapter 3, it has seen that chlor-alkali plants, using elemental mercury as a cathode for generating chlorine gas ($Cl_2$) and caustic soda (NaOH) by electrolysis of the NaCl brines, release large Hg quantities in the environment. Due to their environmental impact, from November 2017 onwards, the European Union banned the use of mercury cathodes in chlor-alkali plants.

The estimated consumption of fish and seafood in the countries of the European Union that are lapped by the Mediterranean Sea was about 12.5 million tons per year. Per capita consumption of captured (non-farmed) products was around 18 kg (EUMOFA, 2019), but this value is higher in those countries that have two / three dishes of fish and seafood in their weekly diet. Spain, France and Italy, the countries covered in the current study, are among the countries that consume the most fish per-capita in Europe. In 2018, they were the top three European countries in total spending for fish and seafood purchases in one year (Italy = 11679 euro; Spain = 10569 euro; France = 8901 euro; EUMOFA, 2019).

In the Mediterranean area, the populations living near the coast (about 150 million people) have eating habits with large consumption of local fish. In many coastal areas, the consumption of local fish reaches up to five courses a week. Cultural heritage (FAO / WHO, 2011) and the perception of some populations convinced that fresh and local fish provide the best quality fish (ISMEA, 2011) are the main reasons for these people having these culinary habits.

Studies evaluating mercury concentrations in marine biota were performed in all oceans and seas (UNEP - Environmental Global Mercury Assessment, 2018). The Mediterranean was also widely studied. The amounts of mercury are higher in the species of Mediterranean fish with the same size range as the Atlantic Ocean (Junque et al., 2018; Cossa et al., 2012; Lahaye et al., 2006). There are extensive differences, although mercury levels in the waters of the two water basins are similar (Bowman et al., 2015; Kotnik et al., 2007).

In the literature, studies have evaluated mercury levels of fish and seafood in specific areas (de Matos et al., 2021; Razavi et al., 2020; Yu et al., 2020; Burns & Riva-Murray, 2018; Cebalho et al., 2017), but no one has addressed the global problem in an area as large as the Western Mediterranean. Furthermore, previous studies have usually treated a very limited number of fish and seafood species. The Mediterranean Sea has a large plethora of fish species because of its vast biodiversity. It encompasses more than 700 different species between fish and seafood, more than 200 being edible. These numbers are changing because of global warming and the entrance of allochthonous fish species. These invasive species arrive through the Suez Passage and are transported on boats and yachts. The introduction of these new species puts the survival of many Mediterranean native species to serious risk.

The high species variability is also noted on the market desks of Western Mediterranean cities where large quantities of various fish are sold, belonging to different species, depending on the different season and climatic conditions. They also disclose the culinary habits regarding fish consumption in different countries. Some species of edible fish are highly appreciated in some countries and are not considered attractive in others. This is an aspect to consider for the monitoring studies if performed in several countries.

In the present study, sampling was planned to obtain as many as possible species at different times of the year to obtain the most varied fish database possible.

This chapter is initially devoted to report the levels of mercury present in the local edible fish consumed by the populations of the Western Europe. These data, combined with an appropriate study on the dietary habits, could be a focal point for assessment of the human health risk for Hg toxicity in this population. Assuming that almost the totality of T-Hg concentration is in the form of MeHg, the quantity of MeHg ingested by the individuals can be estimated from the fish concentrations. Thus, the Estimated Weekly Intakes (EWIs) were calculated for each country in which samples were collected and compared with the Provisional Tolerable Weekly Intake (PTWI) that is estimated at 1.6 μg/kilogram per body weight per week by FAO/ WHO Joint Expert Committee on Food Additives (EFSA, 2004).

A large number of fish species devoted to human consumption has been evaluated. However, the study is essentially focussed on non-oily species because these are those monitored to a lesser extent and those which *a priori* are considered having low Hg levels. Another goal of this chapter is the evaluation of the species that can be considered safe for human consumption, a goal that has not been addressed so far. As already reported previously, fish contain a high nutritional value in terms of proteins, polyunsaturated fatty acids, and minerals. Finding fish species that do not have harmful mercury values combines its high nutritional power with very low toxicity for mercury. So, these species should be eaten preferentially by children and pregnant women. In the current study, a fish or seafood species is considered entirely safe if no analysed specimen showed mercury levels above the threshold value indicated by the European Commission (0.5 mg $kg^{-1}$ ww for most species and 1 mg $kg^{-1}$ ww for some other species). In the case that only one specimen had a mercury concentration above the limit value, the entire species was excluded.

Crustaceans and molluscs are also considered to get a comprehensive complete description of the mercury concentration in edible marine foodstuffs of the Western Mediterranean.

## 4.2. Materials and methods

### 4.2.1. Sample collection and preparation

Fish specimens were collected in Eivissa, Mallorca, Menorca, Alacant, L'Ampolla, Ametlla de Mar (Spain), Marseille (France), Genoa, Civitavecchia, and Alghero (Italy). Fish samples were obtained in the local markets. All the purchased fish were fresh and intended for human consumption. All samples were captured a few hours before purchase with conventional methods. Each specimen was captured in the immediate vicinity of the coast of the place where it was sold, no more than 15 km away. The origin of each specimen was ascertained before purchase through the label imposed by European legislation for the traceability of the fish (European Community regulation n.1224 / 2009 and European Union regulation n. 404 / 2011). Only those fish whose origin was certain were considered for study.

Several sampling campaigns were organized, and more than 1300 specimens were collected from October 2018 to May 2021. The sampling campaigns were carried out at different times of the year. More than 78 fish samples per campaign were bought in each place.

The samples collected belonged to fifty-eight different species among fish, crustaceans, and molluscs. That is, squid (*Loligo vulgaris*), transparent goby (*Aphia minuta*), shrimp (*Aristeus antennatus*), conger (*Conger conger*), common dolphinfish (*Coryphaena hippurus*), common dentex (*Dentex dentex*), white seabream (*Diplodus sargus sargus*), anchovie (*Engraulis encrasicolus*), dusky grouper (*Epinephelus marginatus*), porbeagle (*Lamna nasus*), four-spot megrim (*Lepidorhombus boscii*), angler (*Lophius piscatorius*), european hake (*Merluccius merluccius*), red mullet (*Mullus barbatus*), surmullet (*Mullus surmuletus*), mediterranean moray (*Muraena helena*), mussel (*Mytilus galloprovincialis*), norway lobster (*Nephrops norvegicus*), blackspot seabream (*Pagellus bogaraveo*), common pandora (*Pagellus erythrinus*), axillary seabream (*Pagellus acarne*), greater forkbeard (*Phycis blennoides*), thornback ray (*Raja clavata*), sardine (*Sardina pilchardus*), round sardinella (*Sardinella aurita*), brown meagre (*Sciaena umbra*), black scorpionfish (*Scorpaena porcus*), red scorpionfish (*Scorpaena scrofa*), small-spotted catshark (*Scyliorhinus canicula*), greater amberjack (*Seriola dumerili*), comber (*Serranus cabrilla*), painted comber (*Serranus scriba*), common sole (*Solea solea*), european barracuda (*Sphyraena sphyraena*), picarel (*Spicara*

*smaris*), black seabream (*Spondyliosoma cantharus*), atlantic horse mackerel (*Trachurus trachurus*), pearly razorfish (*Xyrichtys novacula*), john dory (*Zeus faber*), common seabream (*Pagrus pagrus*), megrim sole (*Lepidorhombus whiffiagonis*), gurnard (*Chelidonichthys lucerna*), gilthead seabream (*Sparus aurata*), greater weever (*Trachinus draco*), poor cod (*Trisopterus minutus*), red bandfish (*Cepola macrophthalma*), white sardinella (*Sardinella albella*), offshore rockfish (*Pontinus kuhlii*), cuckoo wrasse (*Labrus bimaculatus*), blue whiting (*Micromesistius poutassou*), cuttlefish (*Sepia officinalis*), whiting (*Merlangius merlangus*), sand steenbras (*Lithognathus mormyrus*), salema (*Sarpa salpa*), mackerel (*Scomber scombrus*), flathead mullet (*Mugil cephalus*), european eel (*Anguilla anguilla*), and octopus (*Octopus vulgaris*).

Once purchased, these samples were placed in a portable freezer and kept at a constant temperature of 4° C. They were transported to the laboratory, where they were frozen at -22° C. The following day they were thawed. Each specimen was photographed, and all information of species, weight, and length was recorded. At this point, a portion of the epaxial muscle was removed from fish, e.g. 1-5 g, using a ceramic knife and plastic tweezers. In crustaceans a part of the pulp present inside the carapace was taken, while in molluscs an edible part of the main body was withdrawn, depending on the species treated. In the removed part, bones and skin were removed, so that only the muscle remained. The removed sample was further cut and shredded into small pieces of 10-50 mg. The resultant aliquots were then placed in a sterilized 20-mL glass vial and hermetically sealed with a suitable cap. After this step, the vials were stored in a freezer at -20 ° C, until the day of analysis. Each remaining specimen body was placed in a hermetically sealed plastic bag, labelled and deep-frozed at -22° C to get a reference biobank.

The tools used were made of ceramic and plastic to avoid metal contamination. Before preparation of each sample, the utensils were cleaned with soap, distilled water and 96% ethanol (Honeywell, France).

### 4.2.2. Total mercury concentration analysis

Shortly before analysis, the samples inside the vials were thawed. Before each use, the sample container was cleaned by placing it in the oven of the instrument at 750° C for 8 minutes.

An automated atomic absorption spectrometer (Model AMA-254, Altec LTD, Czech Republic) was used for total mercury (T-Hg) concentration analysis. The sample container with the sample matrix (around 10-100 mg depending on fish species) entered in a combustion tube, heated up to ~750º C. The sample was vaporized and transported to catalytic compounds that removed interfering impurities. Later, an oxygen carrier transported the gases to an amalgamator which consisted of a glass tube containing gold-plated ceramics, where mercury was retained. Finally, an oven heated the amalgamator (to ~900º C) and the released mercury vapour was conveyed to a cuvette, positioned in the path length of an atomic absorption spectrometer that used an element-specific lamp that emitted light at a wavelength of 253.7 nm, and a silicon UV diode detector for mercury quantitation.

The detection limit for the instrument was 0.0009 μg $g^{-1}$ in dry weight. Blanks were analysed between samples to prevent Hg transfer between them.

For every 10 samples the accuracy of the instrument was checked, using the European Reference Material (ERM-BB422) from the Institute for Reference Materials and Measurements of the European Commission's Joint Research Centre (Geel, Belgium). ERM-BB422 consisted of powdered dry fish muscle and was chosen according to sample Hg concentration levels. The values obtained were always within the ERM-BB422 confidence interval (0.601±0.03 mg $kg^{-1}$).

### 4.2.3. Methylmercury Estimation Weekly Intake (EWI)

Assuming that the totality of Hg is in the form of MeHg in fish, the Estimation Weekly Intake (EWI) of MeHg related to fish consumption for each country was calculated. EWI was found by multiplying the median of all fish samples by the weekly fish consumption per body weight in the country. Consumption data were obtained from international and local agencies or from literature articles (FAO, 2020; ISMEA, 2011; Bemrah et al., 2009; AESAN, 2006). The results of the EWI were compared with the Provisional Tolerable Weekly Intake (PTWI) for MeHg absorption calculated by the FAO / WHO Joint Expert Committee on Food Additives (EFSA, 2004). The percentage of the EWI with respect to the PTWI reference value was then calculated, according to the following equation:

$$EWI\ (\%) = 100 * (EWI / PTWI) \qquad \text{(Equation 4.1)}$$

### 4.2.4. Data analysis

Figures and tables were performed using the statistical software R (R Development Core Team, 2019) and Office packages (Microsoft Corporation, 2020).

## 4.3. Results

The mercury concentration results are reported in mg $kg^{-1}$ in wet weight. See the Appendix B to find all individual values.

In the three principal Balearic Islands (Mallorca, Menorca, Eivissa) fish specimens were collected from October 2018 to January 2021. Other samples were also provided by the General Direction of Public Health and Consumption of the Government of the Balearic Islands. Among those collected and those supplied by the institutions, 572 specimens were analysed. This large number of specimens involved 41 different species (Figure 4.1). The mean T-Hg level was 0.49 mg $kg^{-1}$ ww, while the median was 0.32 mg $kg^{-1}$ ww. The concentrations of T-Hg ranged between 0.01 and 3.8 mg $kg^{-1}$ ww. 172 samples (30% of the total) were above the T-Hg safety limits for human consumption of the European legislation. Twelve percent of the total (72 samples) exceeded these threshold values more than twice. All specimens of thornback ray, shrimp, norway lobster, and greater forkbeard, porbeagle and european barracuda were above the EU threshold for human consumption. More than 75% of the specimens of dusky grouper and common dentex were above this EU threshold.

In Alacant, 197 fish of 31 different species were collected (Figure 4.2) in March 2019. The median of Alacant's T-Hg in fish samples was 0.12 mg $kg^{-1}$ ww and the average 0.17 mg $kg^{-1}$ ww, ranging from 0.01 to 1.03 mg $kg^{-1}$ ww. Only 8 samples had T-Hg values that exceeded the limits while not a single specimen duplicated the cut-off values.

The samples collected in L'Ampolla and Ametlla de Mar are reported enclosed in a single cluster, called the Ebro Delta. Here, 99 specimens of 14 species (Figure 4.3) were collected in June, July, and August 2020. T-Hg concentration values started from 0.01 up to a maximum of 1.88 mg $kg^{-1}$ ww.

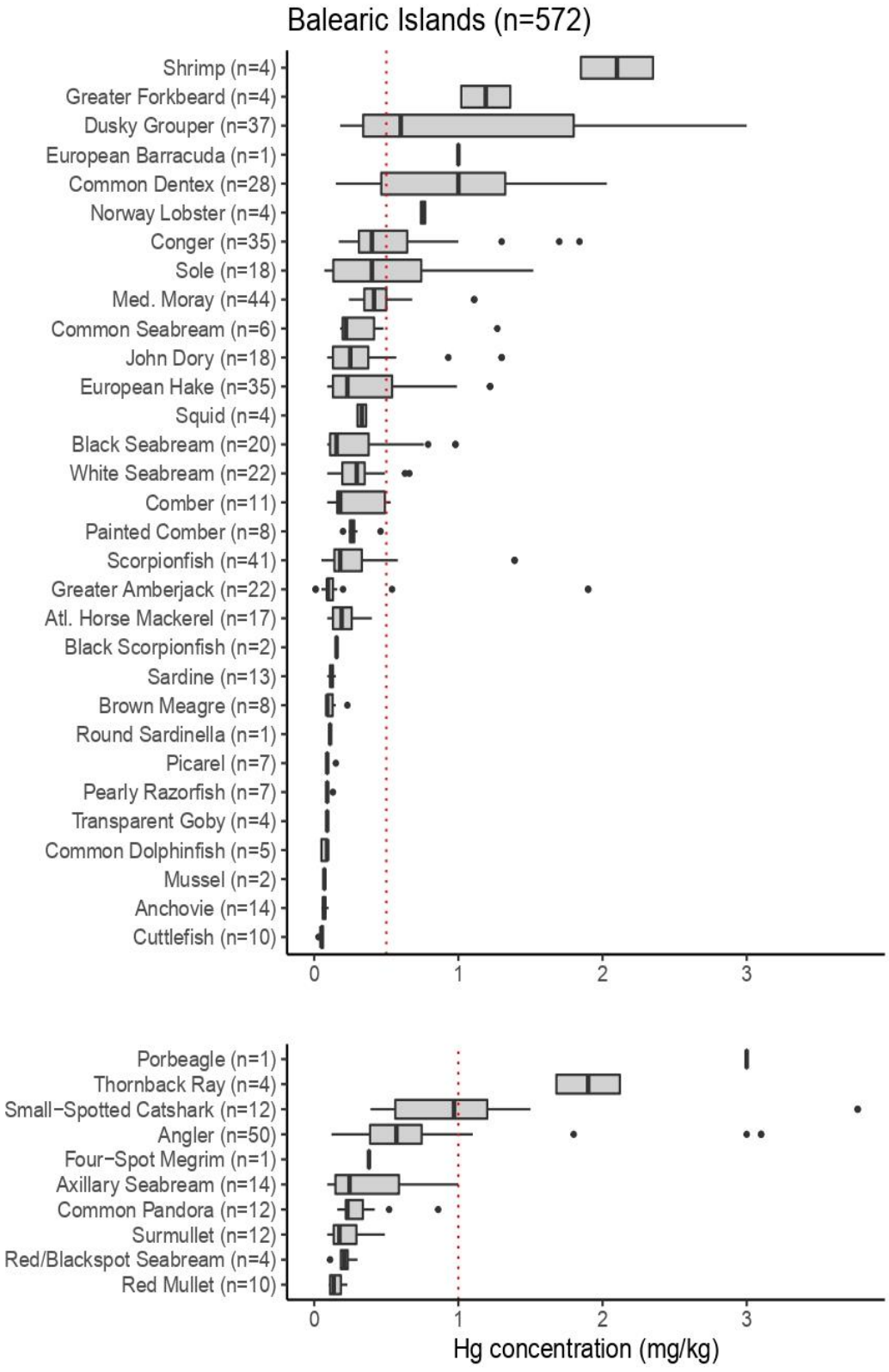


**Figure 4.1.** Total mercury concentrations in edible fish from the Balearic Islands. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

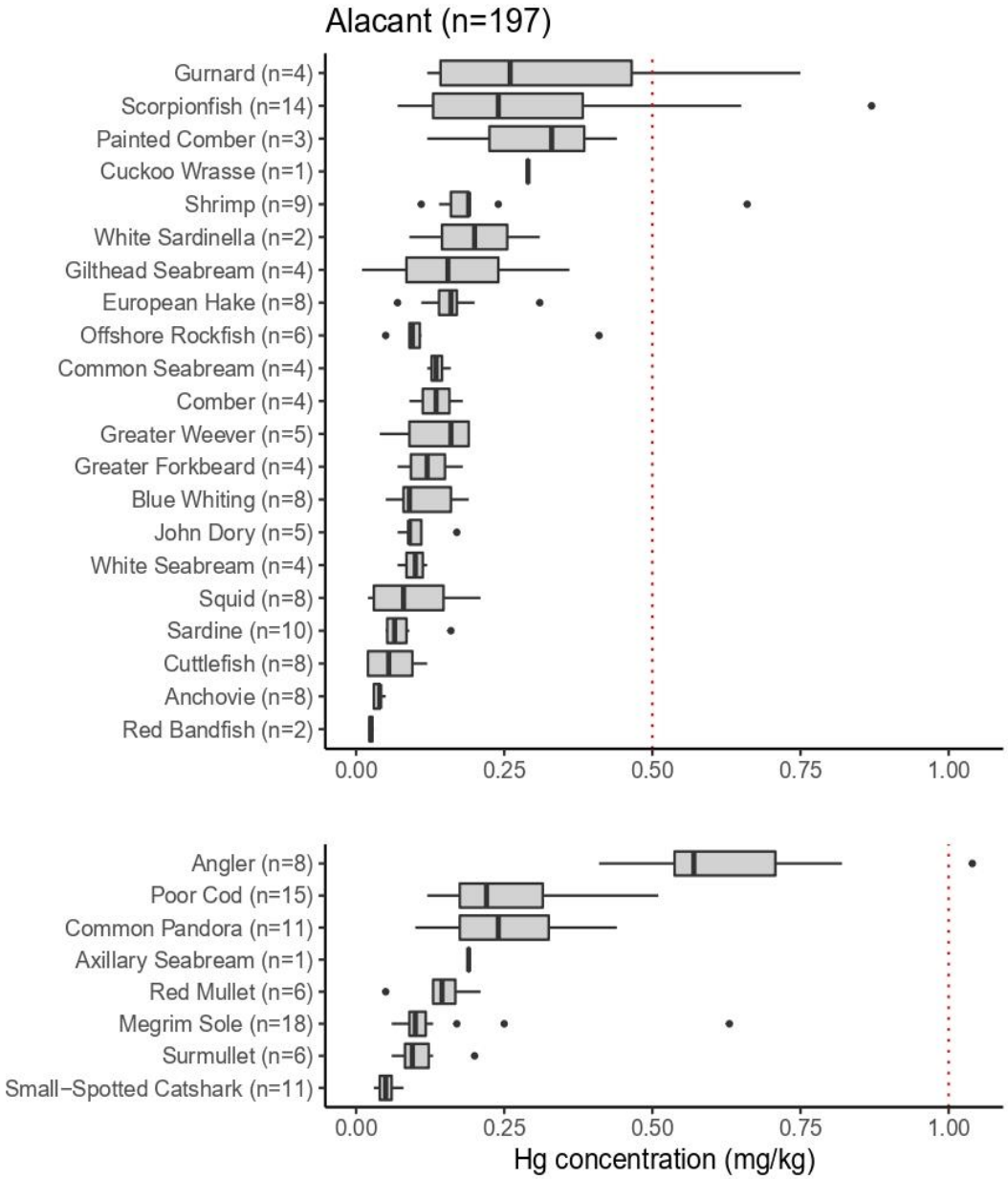


**Figure 4.2.** Total mercury concentrations in edible fish from Alacant. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

The T-Hg median was 0.21 mg $kg^{-1}$ ww and the mean of 0.37 mg $kg^{-1}$ ww. Nineteen samples exceeded the threshold limits (19%), but only four (4%) duplicated these values. More than 75% of the specimens of greater weever were above the EU threshold for human consumption.

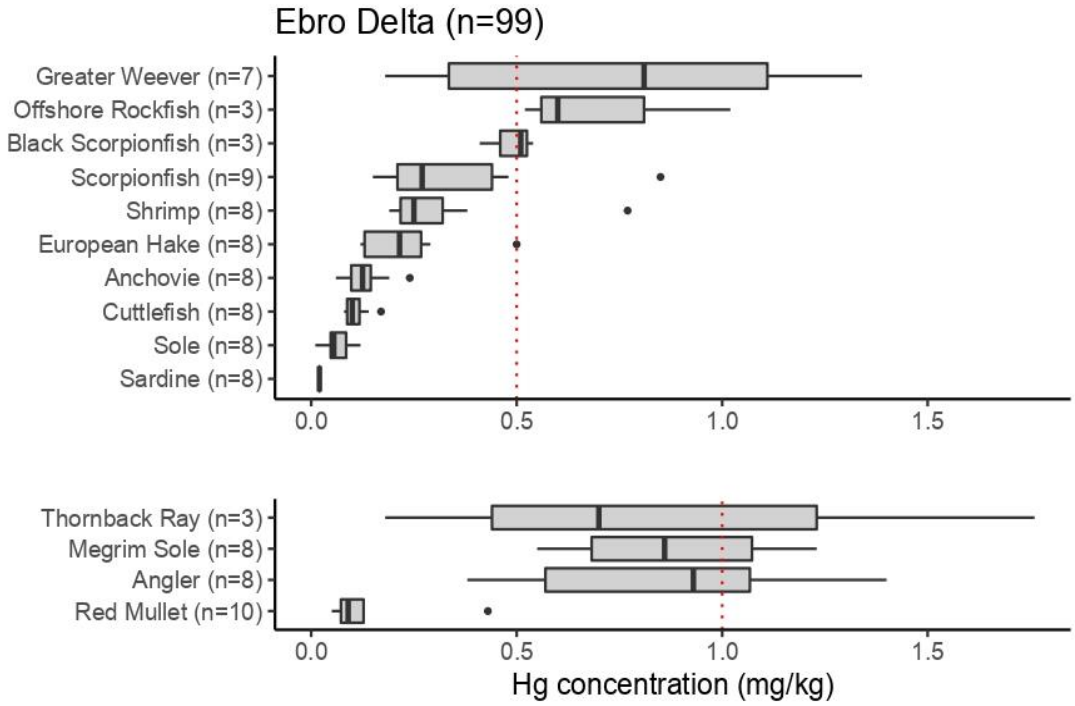


**Figure 4.3.** Total mercury concentrations in edible fish from Ebro Delta littoral. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according EU legislation.

In Spain, a total of 868 specimens were collected, of which 199 with mercury levels above human consumption limits. Thus, 23% of the total had an impactful level of mercury. The sampling campaigns were carried out in different periods, covering all the months of the year, highlighting a different availability of species throughout the calendar year. Although the places where the campaigns took place belonged to the same nation, eating habits were consistently different.

In Marseille, 192 specimens of 23 different species were collected (Figure 4.4). These samples were obtained after three sampling campaigns from October 2018 to February 2020. The median and mean of T-Hg reported were, respectively, 0.26 and 0.35 mg $kg^{-1}$ ww, between a minimum of 0.02 and 2.48 mg $kg^{-1}$ ww. The percentage of samples that had a quantity of T-Hg above the cut-off values was the 23% (44 specimens), while that which duplicated these guideline values was the 5% (10 specimens) of the total. All specimens of common seabream had Hg concentrations above the EU threshold for human consumption. Nearly 75% of the specimens of white seabream and atlantic horse mackerel had Hg concentrations above this level.

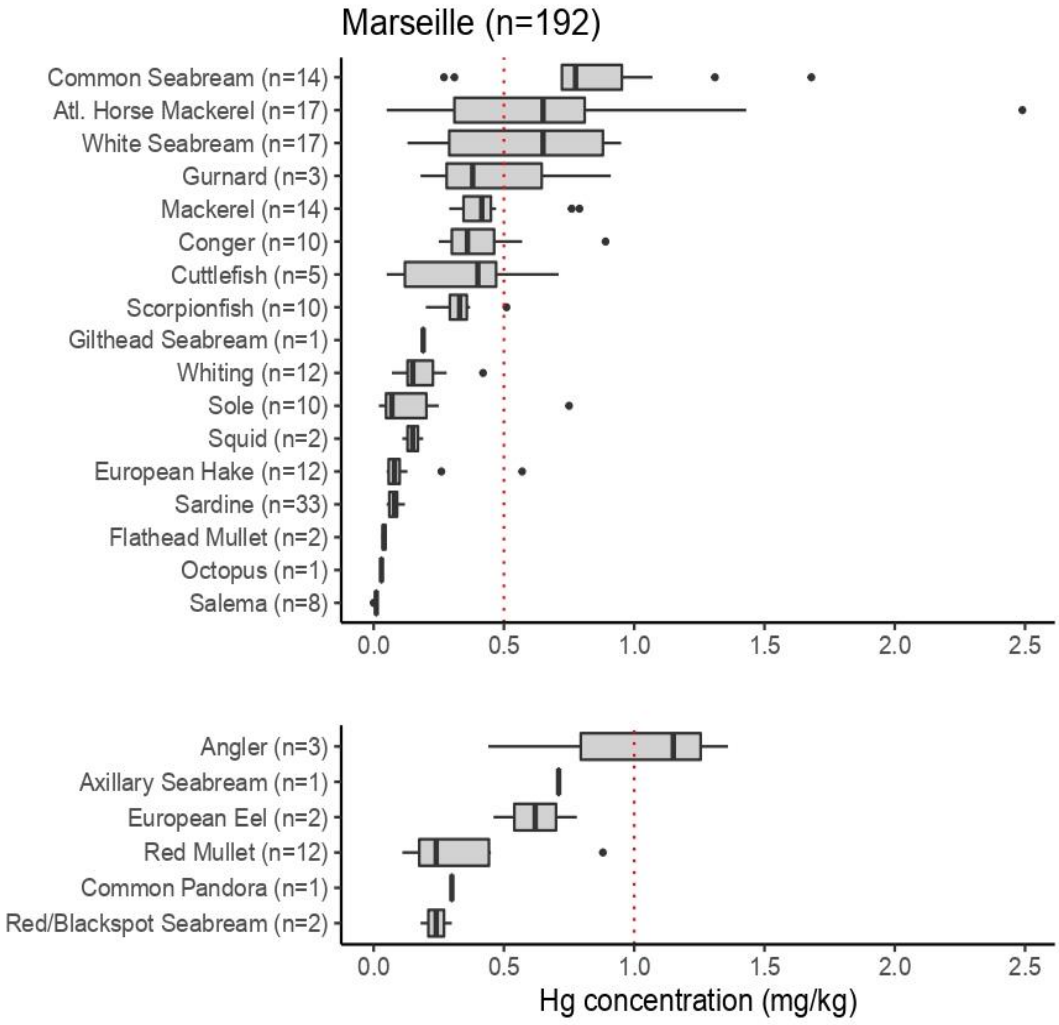


**Figure 4.4.** Total mercury concentrations in edible fish from Marseille. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

In France, these three sampling campaigns led to the purchase of 192 specimens in Marseille, the most important port city in the South of France. The campaigns were carried out in different seasons, outlining the different species available for consumption over the course of a calendar year.

In Civitavecchia, samples (126 specimens of 11 different species, as shown in Figure 4.5) were collected from June 2019 to May 2021. The concentration of T-Hg ranged from 0.01 to 2.45 mg $kg^{-1}$ ww, with a mean of 0.48 mg $kg^{-1}$ ww and a median of 0.39 mg $kg^{-1}$ ww. Thirty-three percent of the samples, 45 specimens, exceeded the limit values, while an 8%, 10 specimens, recorded values more than doubling the maximum permitted threshold. Nearly all comber specimens and a large proportion of scorpionfish had Hg values above this threshold.

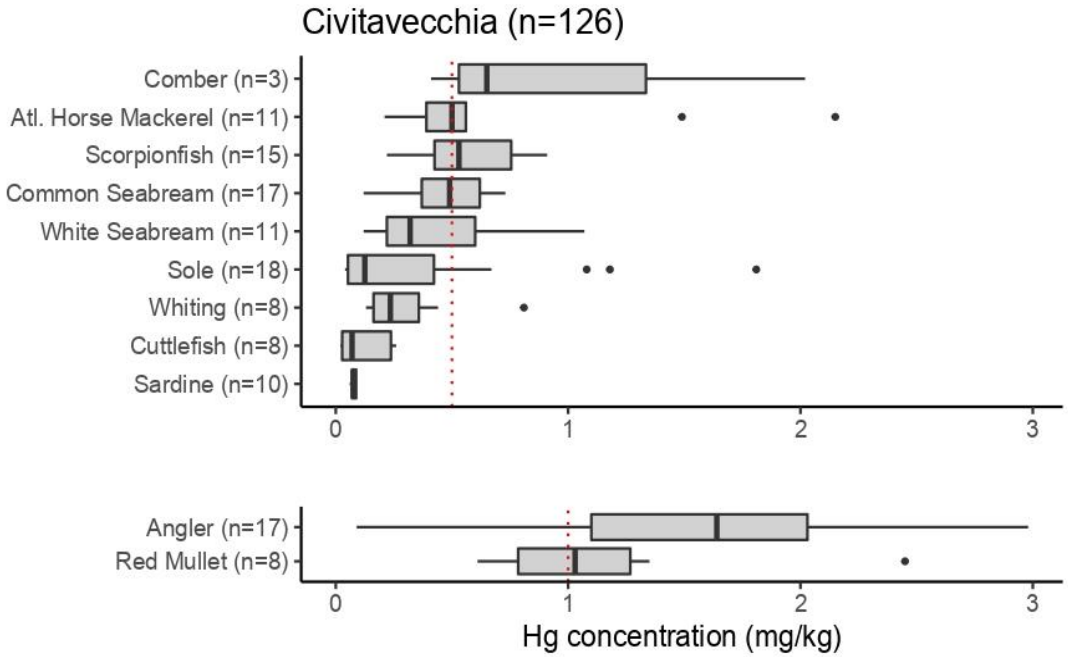


**Figure 4.5.** Total mercury concentrations in edible fish from Civitavecchia. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

In Genoa, 78 specimens belonging to 9 different species were obtained (Figure 4.6). T-Hg median was 0.21 mg $kg^{-1}$ ww and T-Hg mean was 0.35 mg $kg^{-1}$ ww. Values for mercury levels ranged from 0.01 to 1.66 mg $kg^{-1}$ ww. Thirteen samples (16%) were above the EU Hg cut-off, but only 2 samples exceeded these limit values more than double. Nearly all common seabream species had Hg concentrations above this limit.

In Alghero, 81 specimens were purchased (8 species, Figure 4.7) from June 2019 to April 2021. The median for T-Hg concentrations in Alghero's specimens was 0.32 mg $kg^{-1}$ ww and the average was 0.27 mg $kg^{-1}$ ww, from a value of 0.04 to 1.7 mg $kg^{-1}$ ww. In Alghero overall, 15 specimens (18%) were above the threshold values and only 2 were above double.

In Italy, 285 specimens were collected. Seventy-three specimens (25.5% of total) presented levels of T-Hg above the limit-threshold. Samples were collected in all months of the year and different eating habits were observed according to the country where the sampling took place.

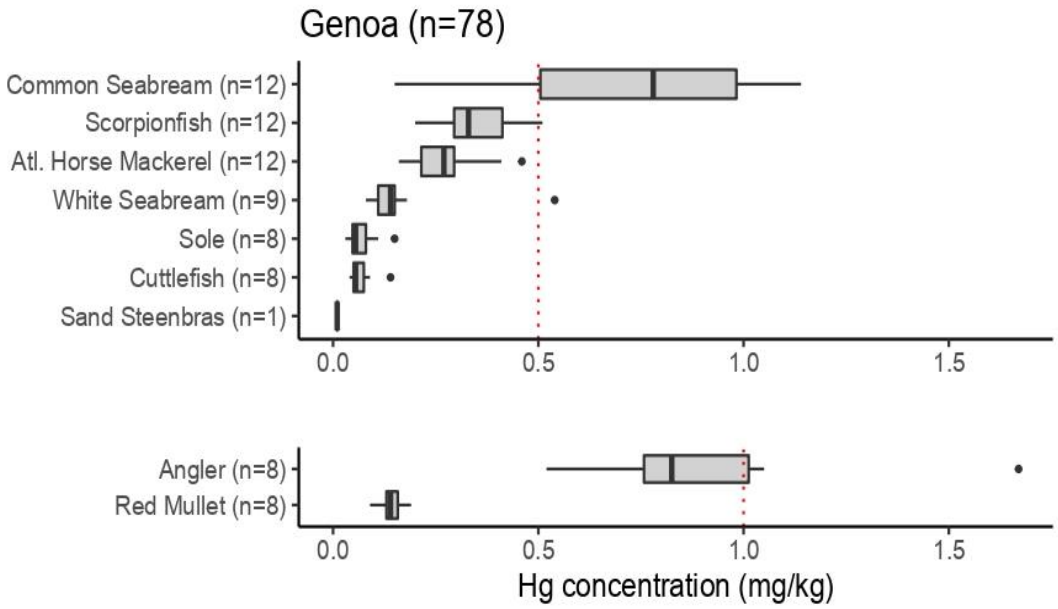


**Figure 4.6.** Total mercury concentrations in edible fish from Genoa. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

Overall, 1380 specimens of fish, crustaceans and molluscs were collected. Among them, 35 fish specimens were excluded, due to excessive small size or dubious origin. Finally, 1345 specimens from the Western Mediterranean were analysed. The median for T-Hg concentrations in all collected samples was 0.24 mg $kg^{-1}$ ww, while the mean was 0.4 mg $kg^{-1}$ ww. The number of species with T-Hg levels above the EU limits was 316 (23.5% of total; Figure 4.8).

With regard to benthic and bentho-pelagic species, blackspot seabream (*Pagellus bogaraveo*; mean 0.22 mg $kg^{-1}$ ww, range 0.11-0.3 mg $kg^{-1}$ ww), pearly razorfish (*Xyrichtys novacula*; mean 0.1 mg $kg^{-1}$ ww, range 0.09-0.13 mg $kg^{-1}$ ww), surmullet (*Mullus surmuletus*; mean 0.18 mg $kg^{-1}$ ww, range 0.062-0.49 mg $kg^{-1}$ ww), brown meagre (*Sciaena umbra*; mean 0.12 mg $kg^{-1}$ ww, range 0.09-0.23 mg $kg^{-1}$ ww) and painted comber (*Serranus scriba*; mean 0.28 mg $kg^{-1}$ ww, range 0.12-0.46 mg $kg^{-1}$ ww) did not exceed the EU T-Hg threshold values in any specimen.

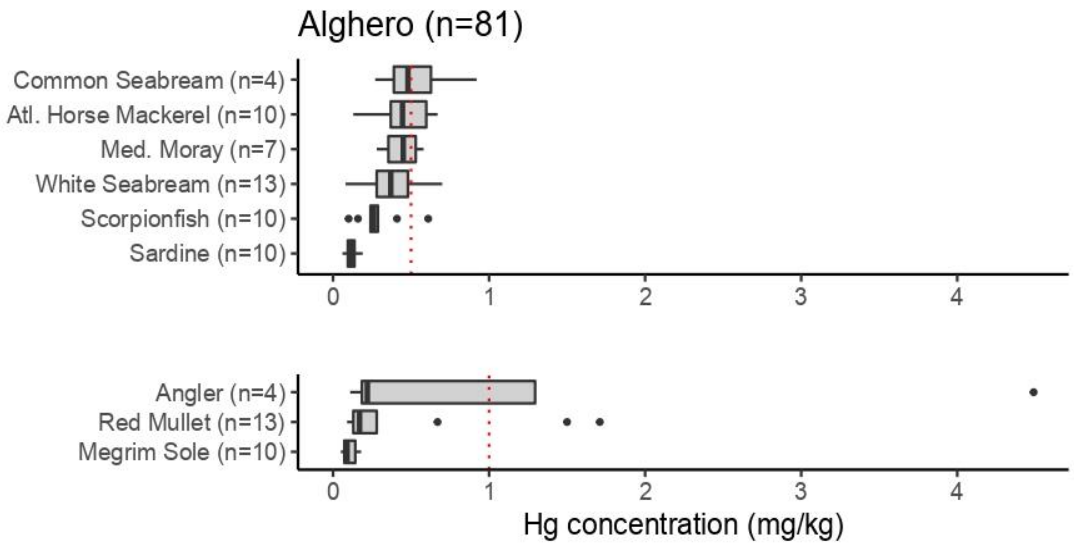


**Figure 4.7.** Total mercury concentrations in edible fish from Alghero. The red dotted line indicates the maximum limit for the level of mercury that can be consumed by humans without adverse health effects in fish according to EU legislation.

Common dolphinfish (*Coryphaena hippurus*) was the only great predator species that was entirely safe as regard mercury concentration. This species had very low T-Hg concentration mean and median, both attesting at 0.07 mg $kg^{-1}$ ww. Its T-Hg range was between 0.05 and 0.09 mg $kg^{-1}$ ww.

The only mollusk species found within the safe species for human consumption was squid (*Loligo vulgaris*), with a mercury mean value of 0.17 mg $kg^{-1}$ ww and a range of 0.02-0.36 mg $kg^{-1}$ ww. Only a single specimen of cuttlefish (*Sepia officinalis*) had T-Hg levels above the EU cut-off limit, while all the other specimens were well underneath. Therefore, it was not considered in the group of safe species.

None of the crustaceans' species stayed within the safe species. They showed several specimens with very high Hg amounts.

## 4.4. Discussion

All results are reported in total mercury concentration. More than 90% of mercury in fish muscle and seafood is present in the form of methylmercury. Thus, total mercury could be estimated assuming conservatively that all the mercury is in MeHg form (EFSA, 2004).

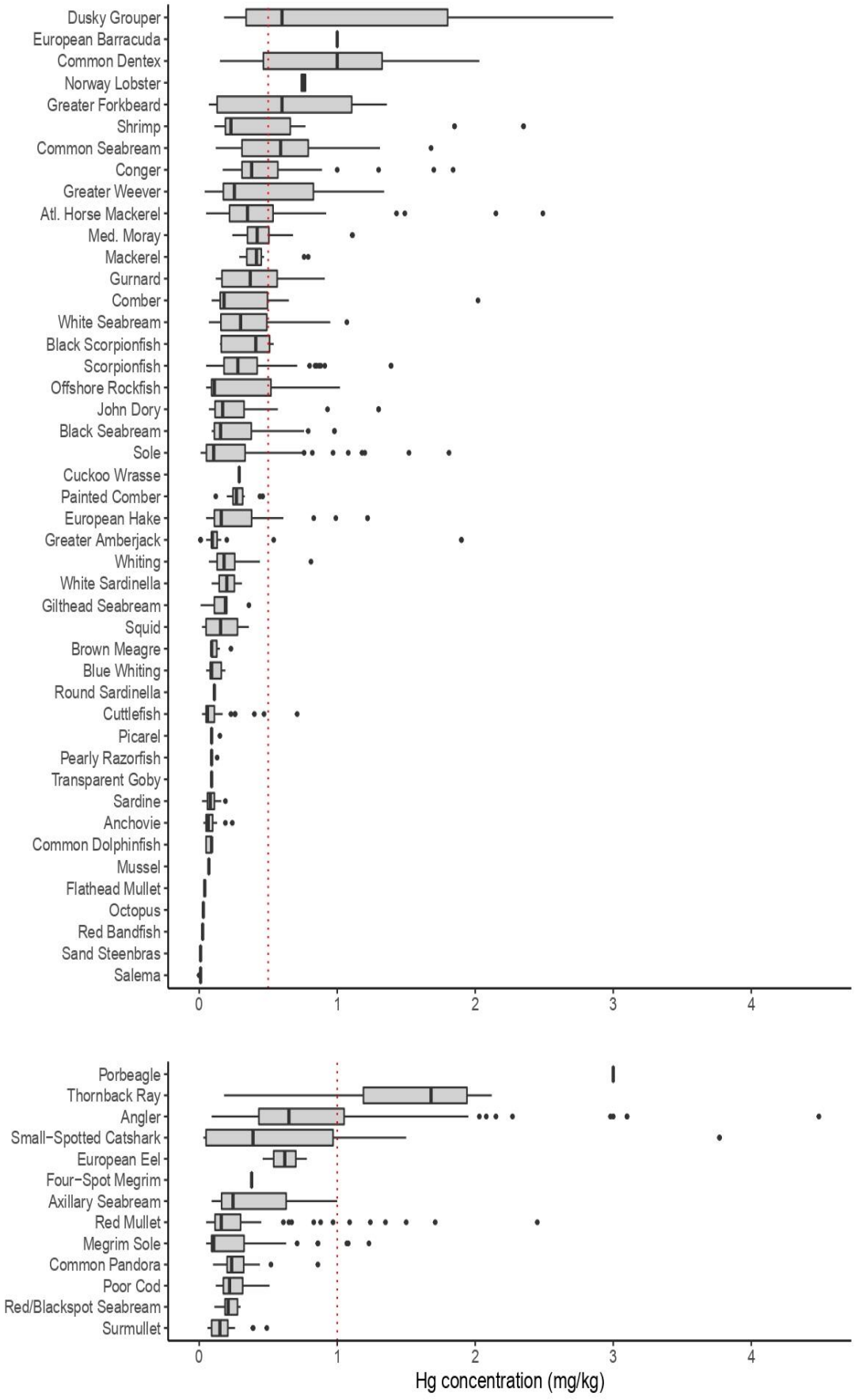


**Figure 4.8.** Total mercury concentrations in edible fish from the Western Mediterranean Sea.

**Table 4.1.** Species, mean weight (W), mean length (L), number (N), sites in which fish were purchased, mercury range, mean and median (mg $kg^{-1}$ ww) in fish safe species.

| Scientific name | Species | W | L | Min | Max | Mean±SD | Median | N | Site |
|---|---|---|---|---|---|---|---|---|---|
| *Sardina pilchardus* | Sardine | 24 | 15 | 0.06 | 0.19 | 0.12±0.04 | 0.12 | 10 | ALG[a] |
| | | 37 | 17 | 0.05 | 0.16 | 0.08±0.03 | 0.07 | 10 | AL[b] |
| | | 65 | 21 | 0.09 | 0.15 | 0.12±0.01 | 0.12 | 13 | BI[c] |
| | | 20 | 14 | 0.05 | 0.12 | 0.08±0.02 | 0.08 | 33 | M[d] |
| | | 15 | 13 | 0.06 | 0.09 | 0.08±0.01 | 0.08 | 10 | CI[e] |
| | | 31 | 15 | 0.02 | 0.02 | 0.02±0.01 | 0.02 | 7 | ED[f] |
| | | **21** | **5** | **0.01** | **0.19** | **0.084±0.04** | **0.079** | **84** | **Total** |
| *Engraulis encrasicolus* | Anchovie | 18 | 14 | 0.06 | 0.24 | 0.13±0.06 | 0.12 | 8 | ED[f] |
| | | 12 | 13 | 0.05 | 0.10 | 0.07±0.02 | 0.07 | 14 | BI[c] |
| | | 12 | 13 | 0.03 | 0.05 | 0.04±0.01 | 0.04 | 8 | AL[b] |
| | | **14** | **13.** | **0.03** | **0.24** | **0.077±0.05** | **0.064** | **30** | **Total** |
| *Loligo vulgaris* | Squid | - | - | 0.30 | 0.36 | 0.33±0.04 | 0.33 | 4 | BI[c] |
| | | 309 | 55 | 0.02 | 0.21 | 0.09±0.07 | 0.08 | 8 | AL[b] |
| | | 91 | 37 | 0.11 | 0.19 | 0.15±0.06 | 0.15 | 2 | M[d] |
| | | **180** | **42** | **0.02** | **0.36** | **0.17±0.12** | **0.16** | **14** | **Total** |
| *Mullus surmuletus* | Surmullet | 250 | 21 | 0.09 | 0.49 | 0.22±0.13 | 0.17 | 12 | BI[c] |
| | | 84 | 20 | 0.06 | 0.20 | 0.11±0.05 | 0.10 | 6 | AL[b] |
| | | **250** | **20** | **0.06** | **0.49** | **0.18±0.12** | **0.15** | **18** | **Total** |
| *Serranus scriba* | Painted comber | - | - | 0.20 | 0.46 | 0.28±0.08 | 0.27 | 8 | BI[c] |
| | | 81 | 20 | 0.12 | 0.44 | 0.30±0.16 | 0.33 | 3 | AL[b] |
| | | **262** | **15** | **0.12** | **0.46** | **0.28±0.1** | **0.27** | **11** | **Total** |

[a] Alghero; [b] Alacant; [c] Balearic Islands; [d] Marseille; [e] Civitavecchia; [f] L'Ampolla-Ametlla de Mar (Ebro Delta littoral).

**Table 4.1. (Cont.)** Species, mean weight (W), mean length (L), number (N), sites in which fish were purchased, mercury range, mean and median (mg $kg^{-1}$ ww) in fish safe species.

| Scientific name | Species | W | L | Min | Max | Mean±SD | Median | N | Site |
|---|---|---|---|---|---|---|---|---|---|
| *Pagellus bogaraveo* | Blackspot seabr. | - | - | 0.11 | 0.30 | 0.21±0.08 | 0.21 | 4 | BI[c] |
| | | 103 | 18 | 0.18 | 0.30 | 0.24±0.08 | 0.24 | 2 | M[d] |
| | | **181** | **18** | **0.11** | **0.3** | **0.22±0.07** | **0.21** | **6** | **Total** |
| *Micromesistius poutassou* | Blue whiting | **47** | **20** | **0.05** | **0.19** | **0.11±0.05** | **0.09** | **8** | **AL**[b] |
| *Sarpa salpa* | Salema | **547** | **34** | **0.00** | **0.01** | **0.01±0.002** | **0.01** | **8** | **M**[d] |
| *Sciaena umbra* | Brown meagre | **394** | **29** | **0.09** | **0.23** | **0.12±0.05** | **0.09** | **8** | **BI**[c] |
| *Spicara smaris* | Picarel | **620** | **14** | **0.09** | **0.15** | **0.10±0.02** | **0.09** | **7** | **BI**[c] |
| *Xyrichtys novacula* | Pearly razorfish | **-** | **-** | **0.09** | **0.13** | **0.10±0.01** | **0.09** | **7** | **BI**[c] |
| *Coryphaena hippurus* | Common dolphinfish | **1600** | **64** | **0.05** | **0.09** | **0.07±0.02** | **0.09** | **5** | **BI**[c] |

[a] Alghero; [b] Alacant; [c] Balearic Islands; [d] Marseille; [e] Civitavecchia; [f] L'Ampolla-Ametlla de Mar (Ebro Delta littoral).

The current study includes the most frequent fish species sold in local markets and consumed in the three main countries of the western Mediterranean area. Previous studies have also examined the concentrations of total mercury levels in fish intended for human consumption. However, they were mostly concerned with specific areas (Storelli et al., 2020; Barone et al., 2015; Yabanli, 2013; Carbonell et al., 2009) and none have been carried out as extensive analysis as in the current study in different countries. It is certainly not easy to handle data from three distinct populations that have different habits and consumption. In fact, to model a complete risk assessment related to the intake of mercury it is mandatory to consider more environmental and social factors, in addition to the specific fish consumption. However, estimating the absorption of mercury and methylmercury for the populations that consume local fish is certainly an excellent starting point to then arrive at a whole risk assessment, associated with mercury.

Assuming that almost all mercury present in fish is in the form of MeHg, an estimation weekly intake (EWI) has been calculated in Spain, France, and Italy. EWI values are slightly different as country fish consumption is varied. The EWI value has been calculated on an adult population (males and females) with an average weight of 68.48 kg. The highest EWI has been recorded in Spain with a value of 2.43 μg / kg body weight (bw) per week (w), then in France with 2.41 μg / kg bw w and finally in Italy with 2.31 μg / kg bw w. Considering that the limit for weekly sustainable consumption of methylmercury is 1.6 μg / kg bw w, all three countries have a higher mercury uptake than the allowable limits. Reporting the percentage of EWI compared to the Provisional Tolerable Weekly Intake (PTWI), following the equation 4.1, it is found that the percentage of weekly intake is 152%, 151% and 144% in Spain, France and Italy, respectively, in excess of PTWI. Obviously, these estimates may vary from region to region as it is known that consumption levels are not homogeneous, but they are higher in coastal areas. In any case, these data show a common trend for the three Western Mediterranean countries.

About 23.5% of the fish sold in the Mediterranean Sea markets have Hg levels above the EU recommendation for human consumption. Not all fish offered in fishmongers is collected from the Mediterranean Sea. Part of it comes from oceans or originate in the breeding facilities. The different origin involves differences in mercury concentrations. Fish specimens from the ocean have a lower quantity of mercury than in the Mediterranean Sea (Junqué et al., 2018; Cossa et al., 2012; Lahaye et al., 2006). A

similar argument can be made for farmed and wild fish species. Wild species have a higher amount of mercury than farmed ones (Di Lena et al., 2017; Kim et al., 2012; Jardine et al., 2009). The consumption of farmed fish species is regular in European countries (approximately 1.37 million tons per year; EUMOFA, 2019). On the other hand, the present study has not included oily fish, e.g. tuna, which is known to accumulate high concentrations of Hg and widely consumed by humans. Inclusion of the values of this fish species would likely increase the Hg burden. Thus, human consumption is more diverse and may easily include fish from these other origins. The above reported EWI excess are highest values assuming that people only consume wild fish from the Western Mediterranean. However, the present PhD dissertation is concerned with fish from this origin. The key question is to ascertain why Mediterranean wild fish has such mercury pollution and what could be the recommendations for human consumption and environmental management in view of this problem.

The fish specimen that has the highest amount of total mercury is the angler (*Lophius piscatorius*), reaching a value of 4.49 mg $kg^{-1}$ ww. The median of this species is 0.65 mg $kg^{-1}$ ww. This species is a top predator that lives purely in rocky and sandy bottoms (Figure 4.9). The EU has set the limit value for mercury in this species at 1 mg $kg^{-1}$ ww. For this reason, it was expected to have high levels of mercury for this species. The maximum amount of total mercury in the angler specimen is significantly higher than in other studies in the literature, while the median is lower or in line with the averages and medians of the mercury concentrations of this species in the Mediterranean area (Llull et al., 2017; Brambilla et al., 2013; Storelli & Marcotrigiano, 2000). The only study that disagrees with the current study is that of Junque et al., 2018, where the median level for total mercury is 1.9 mg $kg^{-1}$ ww. In the Atlantic Ocean, this species has a lower range, median and maximum value for total mercury (Afonso et al., 2008).

The second highest T-Hg concentration comes from a small-spotted catshark (*Scyliorhinus canicula*; Figure 4.10) with a value of 3.8 mg $kg^{-1}$ ww. It is surprising because this species is not considered a top predator fish in the food chain. This value is quite higher than others present in literature related to the same species (Coelho et al., 2010; Storelli et al., 2005). However, the median of T-Hg of this species, 0.39 mg $kg^{-1}$ ww, is not higher than those of many other species (Figure 4.8).

**Figure 4.9.** Angler / monkfish (*Lophius piscatorius)*.

**Figure 4.10.** Small-spotted catshark (*Scyliorhinus canicula)*. Image reprinted from www.fishbase.org

Among fish specimens with more than three samples, the species with highest median T-Hg concentration, 1.68 mg $kg^{-1}$ ww, is thornback ray (*Raja clavata*; Figure 4.11). Almost all of the specimens examined have mercury levels beyond the limit imposed for this species (75%, 6 samples out of 8). Mercury concentrations in thornback ray are higher in the Mediterranean Sea than in the Atlantic Ocean (Duarte et al., 2021; Dixon & Jones, 1994). The median value of the current study is higher than the mean reported in another work describing the mercury concentration of the Thornback ray in the Mediterranean (Storelli et al., 1998).

**Figure 4.11.** Thornback ray (*Raja clavata)*.

Another species with high median T-Hg concentrations, 1 mg $kg^{-1}$ ww, is common dentex (*Dentex dentex*; Figure 4.12). The mercury values of this fish are not very far from those reported in other studies (Llull et al., 2017; Brambilla et al., 2013), although the concentrations and median in this study are slightly higher. The median of this species and nearly all measured concentrations in the collected specimens are well above the EU threshold value (20 specimens out of 28, 71%).

**Figure 4.12.** Common dentex (*Dentex dentex)*. Images reprinted from www.ictioterm.es

Another species of extraordinary economic importance and present in all the sites where we collected fish is common seabream (*Pagrus pagrus*; Figure 4.13). This species has a very high median, 0.59 mg $kg^{-1}$ ww, which exceeds its mercury threshold value. More than half of the specimens analysed have mercury levels above the limit

(56%, 32 out of 57 specimens examined). The mean value for T-Hg content in this species, 0.6 mg $kg^{-1}$ ww, is similar (Shokr et al., 2019) or higher (Llull et al., 2017; Banana et al., 2016; Plessi et al., 2001) to other Mediterranean studies.

**Figure 4.13.** Common seabream (*Pagrus pagrus)*. Image reprinted from www.fishbase.org

The high number of T-Hg contaminated specimens within a species is a phenomenon related to several species, such as thornback ray (*Raja clavata*), dusky grouper (*Epinephelus marginatus*), common dentex (*Dentex dentex*), greater forkbeard (*Phycis blennoides*), and common seabream (*Pagrus pagrus*). More than half of each of these species have specimens with mercury levels beyond their limit. This is probably related to the temporal fluctuations in mercury levels affecting some specific sites in the Western Mediterranean. Porbeagle (*Lamna nasus*) and European barracuda (*Sphyraena sphyraena*) are not included in the list as not enough samples belonging to these fish species are collected to reach significance (one sample each species).

Six of the studied species are exclusive-pelagic. These fish species are anchovy (*Engraulis encrasicolus*), sardine (*Sardina pilchardus*), picarel (*Spicara smaris*), atlantic horse mackerel (*Trachurus trachurus*), blue whiting (*Micromistesius Poutassou*) and mackerel (*Scomber scombrus*). 194 pelagic specimens of these species were examined, and only 19 showed levels of mercury above the EU threshold of which 18 belong to atlantic horse mackerel, while the other is a mackerel. Previous papers reporting Hg concentrations in some pelagic fish from the Mediterranean area, and they are in agreement with the present study (Koker et al., 2021; Costa et al., 2020; Cammilleri et

al., 2019; Kuplulu et al., 2018). However, the amount of T-Hg concentration in Atlantic horse mackerel and mackerel is higher in the current study. Other papers have reported that the same pelagic species caught in the Atlantic Ocean show even lower mercury levels (Vieira et al., 2021; da Silva et al., 2020), further confirming the impact of the Mediterranean Sea on fish species in relation to mercury concentration. In any case, pelagic fish are the fish category that have a lower T-Hg concentration in relation to the overall fish species.

Fifteen species examined live almost barely on the seabed (the exclusive-benthic species): four-spot megrim (*Lepidorhombus boscii*), angler (*Lophius piscatorius*), red mullet (*Mullus barbatus*), surmullet (*Mullus surmuletus*), norway lobster (*Nephrops norvegicus*), thornback ray (*Raja clavata*), black scorpionfish (*Scorpaena porcus*), red scorpionfish (*Scorpaena scrofa*), small-spotted catshark (*Scyliorhinus canicula*), common sole (*Solea solea*), megrim sole (*Lepidorhombus whiffiagonis*), gurnard (*Chelidonichthys lucerna*), greater weever (*Trachinus draco*), shrimp (*Aristeus antennatus*), offshore rockfish (*Pontinus kuhlii*). Benthic fish show higher T-Hg concentrations than pelagic fish, confirming data from other studies (Grgec et al., 2020; Azevedo et al., 2019; Le Croizier et al., 2019; Arcagni et al., 2018; Li et al., 2017).

The consumption of seafood is constantly increasing. Therefore, it is essential to monitor not only the mercury present in fish, but also in seafood. The different species of molluscs and crustaceans with high consumption in the Mediterranean area were evaluated. The molluscs samples have very low mercury values with very few exceptions. The mean values of mercury concentrations were 0.17 mg $kg^{-1}$ ww for squids, 0.11 mg $kg^{-1}$ ww for cuttlefish, 0.03 mg $kg^{-1}$ ww for octopus, and 0.07 mg $kg^{-1}$ ww for mussels. In this case, the total mercury levels found are in agreement with those in the Mediterranean Sea and oceans mollusc studies in the literature (Barcia Garcìa et al., 2021; Minet et al., 2021; Uren et al., 2020; Ariano et al., 2019; Kalogeropoulos et al., 2012; Pastorelli et al., 2012; Pierce et al., 2008). Here, the T-Hg concentration values in the Mediterranean and in the oceans are similar. So, most likely the molluscs have developed a way to excrete mercury and / or to absorb it to a smaller extent than regular fish.

Crustaceans, on the other hand, have high levels of mercury. In fact, more than half of the specimens analysed have mercury levels above the threshold limit. The two most frequent species in the Mediterranean Sea, shrimp and norway lobster, also show

high average values for T-Hg concentrations, 0.8 mg $kg^{-1}$ ww and 0.76 mg $kg^{-1}$ ww respectively. These mercury levels are comparable to those reported in the study of Di Lena et al., 2018.

Among the different categories of fish, cartilaginous fish deserve a mention for their great presence in the Mediterranean Sea. There are only three species of cartilaginous fish in the present fish database: porbeagle (*Lamna nasus*), thornback ray (*Raja clavata*), and small-spotted catshark (*Scyliorhinus canicula*). This low representation is due to the fact that they are not sold in local markets probably due to their less tasty and towelling flesh. However, in some European kitchens their flesh is used for soup preparation. The T-Hg mean in small-spotted catshark is 0.71 mg $kg^{-1}$ ww, between 0.03 and 3.77 mg $kg^{-1}$ ww, in thornback ray is 1.46 mg $kg^{-1}$ ww from a value of 0.18 to 2.12 mg $kg^{-1}$ ww, and in porbeagle the only sample examined records a value of 3 mg $kg^{-1}$ ww. These results are comparable with other cartilaginous fish (Storelli et al., 2005, Storelli et al., 2002).

Another interesting feature of the examined database is that some species have high values of T-Hg concentration only in specific places (Figure 4.14). Although fish samples are purchased in different times, all the specimens have high T-Hg levels only in a specific place, while do not show this issue anywhere else. For example, it is clear that shrimp, common sole, squid, greater forkbeard, small-spotted catshark, thornback ray in Balearic Islands, Atlantic horse mackerel, axillary seabream, cuttlefish, white seabream, and common seabream in Marseille, angler, red mullet, scorpionfish, and comber in Civitavecchia, anchovies, megrim sole, black scorpionfish, greater weever, offshore rockfish in L'Ampolla and Ametlla de Mar, common seabream in Genoa have specimens with higher Hg concentration related to the other places.

These results suggest a species-specific effect linked to dietary habits in a given habitat that results in a high level of total mercury concentration (Gorbunov et al., 2016). Another reason could be a site-specific event that could have brought about a fluctuation in mercury levels (Da Silva et al., 2005) and that some species were more sensitive than others to these variations.

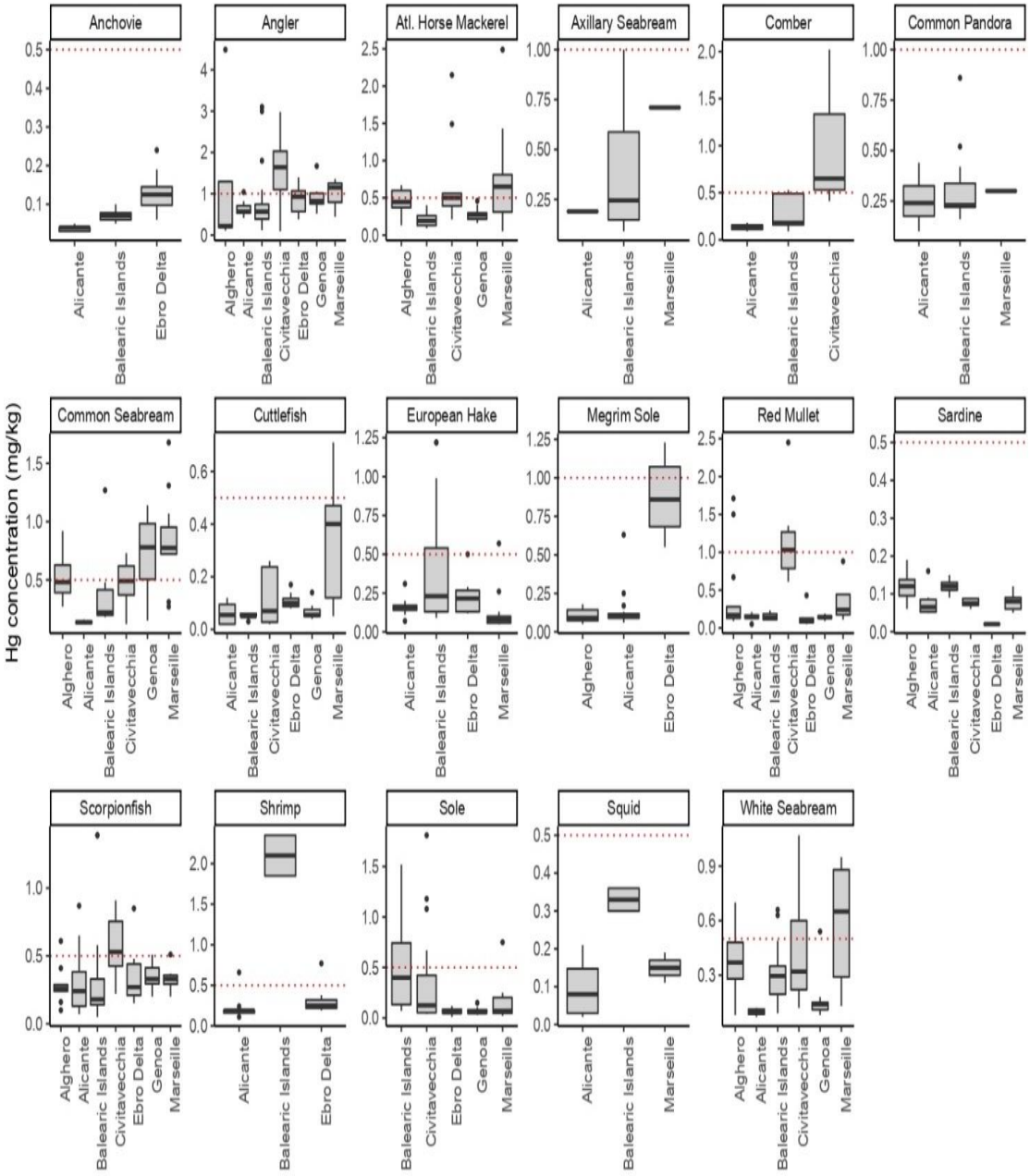


**Figure 4.14.** Mercury concentration differences of fish species in different places.

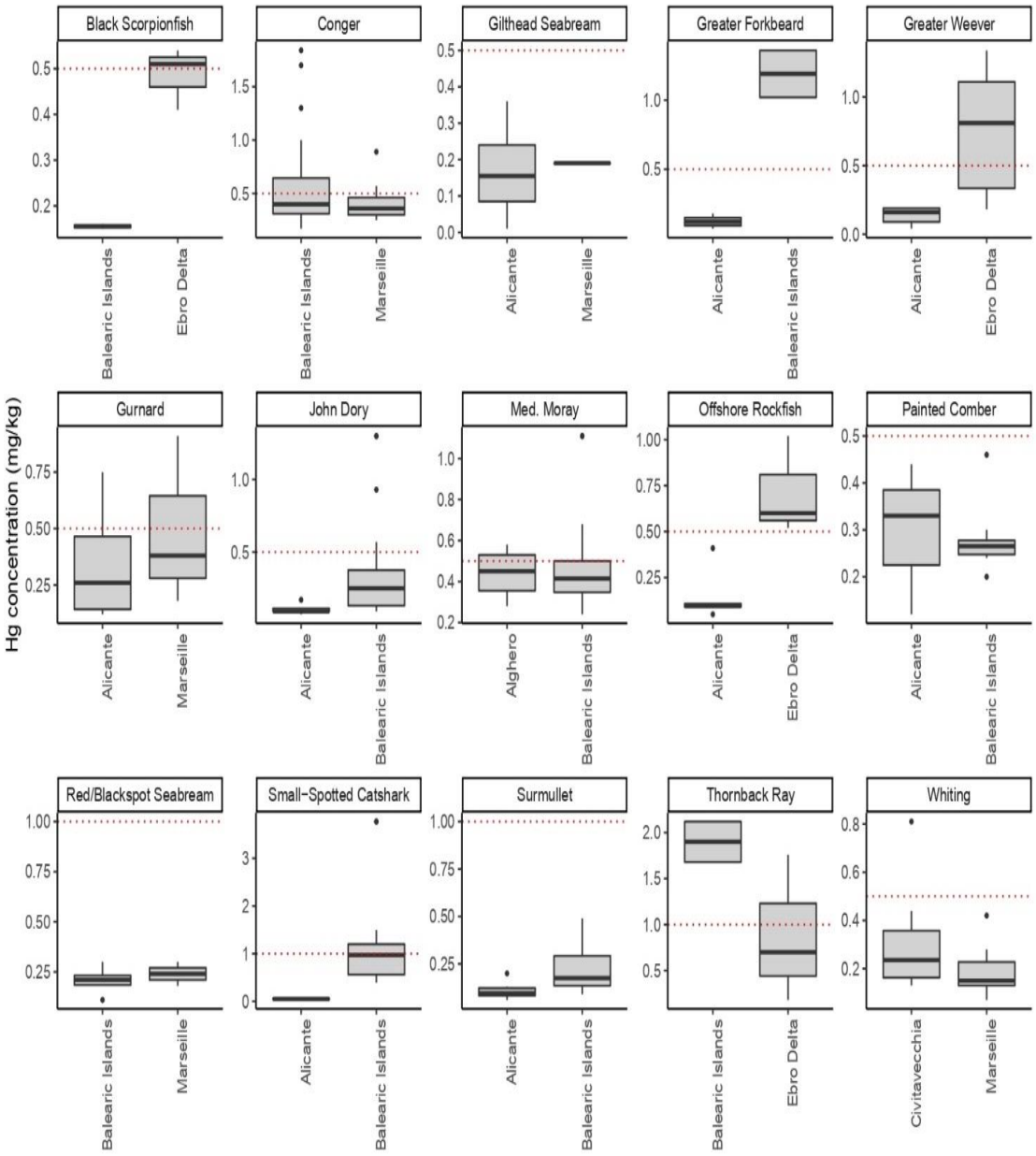


**Figure 4.14 (Cont.).** Mercury concentration differences of fish species in different places.

National advisories should also be emitted for the fish species mentioned above, in consideration of their higher levels of Hg contamination and associated variability. In light of these results, it would therefore be adequate to draw up a list of fish species at high risk concerning total mercury for each individual region and to issue restrictions on their consumption and sales, in order to avoid the absorption of high amounts of mercury.

This study therefore confirmed a critical level of total mercury in the western Mediterranean Sea. Fortunately, this phenomenon does not affect all species of fish, but is limited to a specific number of them. Critical levels of mercury in the Mediterranean Sea do not affect the entire aquatic biosystem, indeed some species show a lower capacity to absorb mercury than others, showing low concentrations. In light of this, the question arose as to which species could be totally safe for human consumption.

In the evaluation of the current fish and seafood database, it has been found only twelve species fulfil the EU threshold for human consumption in all cases: sardines (*Sardina pilchardus*), anchovies (*Engraulis encrasicolus*), squids (*Loligo vulgaris*), surmullets (*Mullus surmuletus*), painted comber (*Serranus scriba*), blackspot seabream (*Pagellus bogaraveo*), blue whiting (*Micromesistius poutassou*), salema (*Sarpa salpa*), brown meagre (*Sciaena umbra*), picarel (*Spicara smaris*), pearly razorfish (*Xyrichtys novacula*), common dolphinfish (*Coryphaena hippurus*). A thorough analysis of these species has been described below.

The pelagic species (sardine, anchovy, picarel, blue whiting; Figure 4.15) have similar mercury levels, starting from a minimum value of 0.015 mg $kg^{-1}$ ww, reaching a maximum of 0.24 mg $kg^{-1}$ ww. For their importance regarding a socio-economical point of view, there are several studies about pelagic fish concerning the western Mediterranean basin (Rumolo et al., 2016; Costalago et al., 2012; Navarro et al., 2011; Palomera et al., 2007), based on the trophic ecology using stomach content and stable isotope analyses. During larval and juvenile stages, pelagic species feed on plankton and show a similar feeding behaviour (Costalago et al., 2011; Ruiz et al., 2006). In the adult stage, pelagic fish manifest different specialized feeding strategy, habitat and seasonality (Costalago et al., 2014; Morote et al., 2010; van der Lingen et al., 2006). These fish species occupy a very low trophic level and feed mainly on zooplanckton, small crustaceans, small fish and cephalopods (Mir-Arguimbau et al., 2020; Albo-Puigserver et al., 2016; Karachle & Stergiou, 2014). The data available in the literature, show a general concordance with the data of the present study for the pelagic species. The only exceptions are given in the study of Bilandžić et al., 2011, in which the mercury levels of the picarel reach 2.06 mg $kg^{-1}$ ww and in that of Ozden, 2013, which reported high levels of mercury only at certain times of the year for sardines (2.15 mg $kg^{-1}$ ww in August and 0.77 mg $kg^{-1}$ ww in September) and anchovies (0.79 mg $kg^{-1}$ ww in March). In the current study, anchovies and sardines do not show any T-Hg mercury variability during March and September.

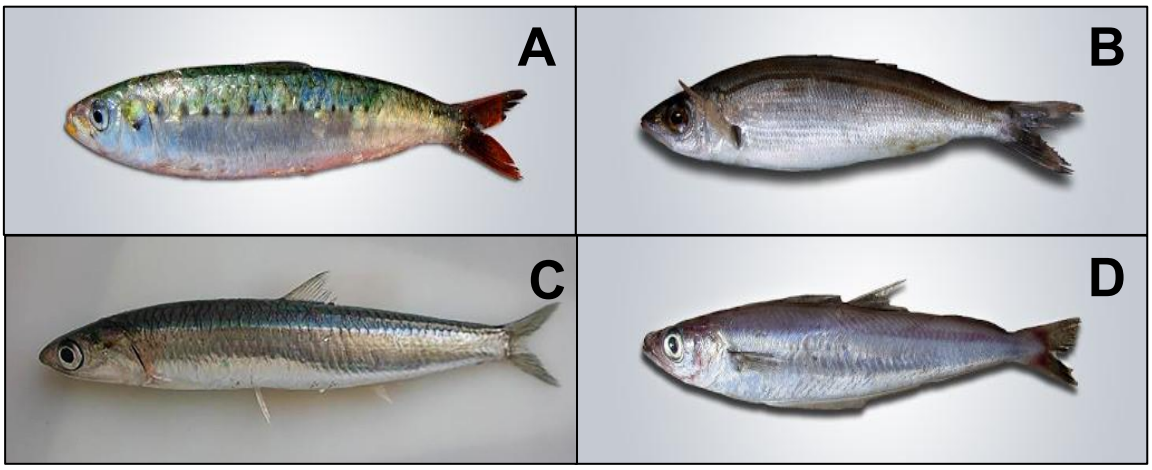


**Figure 4.15.** Pelagic fish. Sardines (*Sardina pilchardus*; A), picarel (*Spicara smaris*; B), anchovies (*Engraulis encrasicolus*; C), blue whiting (*Micromesistius poutassou*; D). Images reprinted from www.ictioterm.es

The specimens with the lowest Hg level are belonging to salema species (*Sarpa salpa*; 0.001 mg $kg^{-1}$ ww; Figure 4.16). Furthermore, with the median value of 0.015 mg $kg^{-1}$ ww, salema is the species with the lowest T-Hg content in Western Mediterranean Sea. The salema feeding habits justify these Hg low values (Ahmed et al., 2014). Indeed, it is the only exclusive-herbivorous species in the Mediterranean Sea. In fact, their primary source of supply is mainly constituted by macrophytes and seaweeds (Matic-Skoko et al., 2004). The data about this species in the current study are in accordance with others in the literature (Torres et al., 2014; Brambilla et al., 2013; Mezghani-Chaari et al., 2011; Storelli et al., 2003).

**Figure 4.16.** Salema (*Sarpa salpa*). Images taken from www.ictioterm.es

Benthic fish live in deep water, are good active predators with highly diversified diets and depend mainly on benthic and mesopelagic prey (Fanelli & Cartes, 2010; Mauchline & Gordon, 1986). In the Mediterranean Sea, the total mercury levels of fish increase with depth because they tend to incorporate concentrations of organic mercury dissolved in seawater and in the seabed as well as those related to their prey (Naccari et al., 2015; Choy et al., 2009). This trend occurs in this study. In this context, it is necessary to report an exception given by surmullet (*Mullus surmuletus*; Figure 4.17), where no specimen showed quantities of Hg above the threshold limits. This data also coincides with the average values of Hg relating to this species in literature (Ramon et al., 2021; Papetti & Rossi, 2009; Falcó et al., 2006).

**Figure 4.17.** Surmullet (*Mullus surmuletus*). Images taken from www.ictioterm.es

In the Mediterranean biodiversity there are not only pelagic or benthic fish, but also fish that have intermediate habits, spending a great part of their life on the seabed and a part in the open sea. These fish species are the Mediterranean fish majority. Among these bentho-pelagic fish species all specimens of blackspot seabream (*Pagellus bogaraveo*), brown meagre (*Sciaena umbra*), pearly razorfish (*Xyrichtys novacula*) and painted comber (*Serranus scriba*) are below the threshold value for mercury concentration (Figure 4.18).

Completely safe for human health are brown meagre and blackspot seabream which have low mercury concentrations; other studies are in agreement with the present work, with the only exception of the study of Brambilla et al., 2013 work, where the blackspot seabream mercury maximum value stands at 0.673 mg $kg^{-1}$ ww.

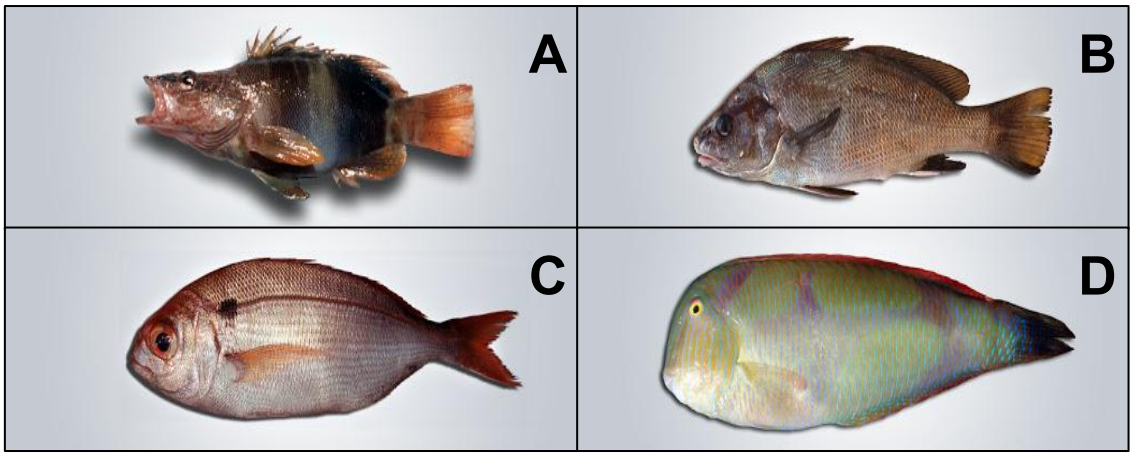


**Figure 4.18.** Bentho-pelagic safe fish. Painted comber (*Serranus scriba*; A), brown meagre (*Sciaena umbra*; B), blackspot seabream (*Pagellus bogaraveo*; C), pearly razorfish (*Xyrichtys novacula*; D). Images reprinted from www.ictioterm.es

In total disagreement with this work are the data collected with the levels of mercury detected in painted comber. In some papers, it has high levels of mercury (Gibičar et al., 2009; Giorgi et al., 2009). The papers just mentioned date back to a few years ago and the mercury levels could fluctuate from one year to another (Grieb et al., 2020). In fact, a more recent paper (Brambilla et al., 2013) aligns with the results of the present work and describes safe mercury levels for painted combers. There are no data in the literature for pearly razorfish. It is a rare fish that lives in clear and shallow areas with sandy bottoms, usually near seaweed and coral beds. As it is increasingly difficult to find a clean seabed, due to the continuous spills of waste into the sea, its habitat has shrunk, leading to a considerable decrease in the number of specimens. For this reason, it is difficult to find this species in European markets. It is consumed in certain regions of Spain and in Southern Italy where its flesh is highly appreciated.

Neither pelagic nor fully benthic, squid (*Loligo vulgaris*; Figure 4.19) inhabits circumlittoral zones. Even if its type of feeding is purely the predation of fish and sometimes on other cephalopods and crustaceans (Pierce et al., 1994), the squid does not register high levels of mercury in any specimen flesh. This species can also be considered the only mollusc species entirely safe. Other papers in the literature report the same trends (Barone et al., 2015; Storelli et al., 2009; Falcò et al., 2006).

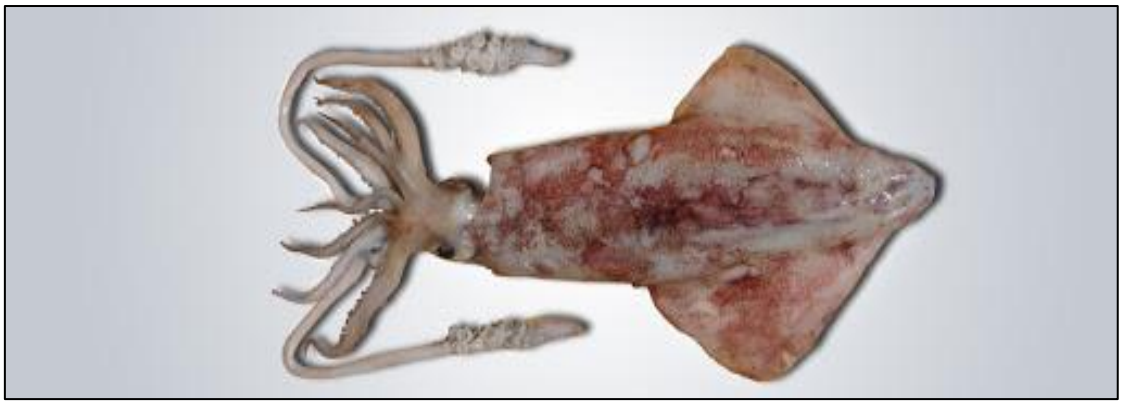

**Figure 4.19.** Squid (*Loligo vulgaris*). Images reprinted from www.ictioterm.es

Similar T-Hg values as squid have been found in cuttlefish (*Sepia officinalis*) in which, however, one specimen with high levels of mercury (0.71 mg kg-1 ww) was found in this study. The results of the other studies confirm those of the present study, as the means and medians of the mercury levels are very low for cuttlefish (Barone et al., 2015; Mezghani-Chaari et al., 2011; Storelli, 2009), but there are rarely some specimens that exceed the threshold value (Brambilla et al., 2013).

Among the large predator species of the Mediterranean Sea, only the common dolphinfish (*Coryphaena hippurus*) is safe for mercury levels. All common dolphinfish specimens (Figure 4.20) in this study have extremely low mercury levels. Being a predator with a large size and high speed, the common dolphinfish is considered a species at the top of the marine food webs (Kojadinovic et al., 2007). There are various studies on mercury levels on this species (Bergés-Tiznado et al., 2019; Araújo & Cedeño-Macias, 2016; Teffer et al., 2014; Kojadinovic et al., 2006), but none of them refer to specimens captured in the Mediterranean Sea. It is possible to capture some specimens in this sea because it is a highly migratory species (Mahon & Oxenford, 1999). This fish travels along many kilometres, but it is very rare. However, this work is the only one that reports the mercury concentrations of common dolphinfish captured in the Mediterranean Sea. Very likely the captured specimens have spent a limited time in the Mediterranean Sea. This could be one of the causes of its low mercury level.

The data of the twelve safe species are robust, as all specimens were acquired in different places and times. For this reason, in a large pool of possible species that could be purchased in the markets of the Western Mediterranean Sea coasts, these twelve could

**Figure 4.20.** Common dolphinfish (*Coryphaena hippurus*). Images reprinted from www.ictioterm.es

prove to be safe choices for human health, even in sensitive categories such as pregnant women and children.

Inter and intra-specific variations in mercury levels in these fish species have been linked to biotic factors (Hajeb et al., 2009; de Marco et al., 2006) and abiotic factors (Choy et al., 2009; Storelli et al., 2005).

## 4.5. Conclusions

The beneficial effects of fish consumption are known, but the deleterious effects that mercury may have on human health should be monitored. The situation of edible fish in the Western Mediterranean Sea is not very rosy. On average, one in five fish individuals has mercury levels above the EU Hg threshold limit. Obviously, there is variability in terms of species and places, but the Hg concentration is an effect that should not be underestimated. More attention should be paid to local species. Despite they are not consumed as much as the best-known fish species, such as *Thunnus albacares, Salmo salar,* and *Xiphias gladius,* they are equally consumed by local populations. There should be precise and constant monitoring of fish species in every single region in order not to risk making a large absorption of Hg in the population and not to manifest effects related to mercury toxicity. For example, a series of warnings should be issued soon for the consumption of some fish species that in some places have high levels of mercury.

This study could suggest which fish species in the Mediterranean Sea are risky and safe for human consumption. Cataloging the species to be avoided and the "friendly" species for human health has an important advantage for the consumers who would know how to move in the purchase of edible fish. Therefore, the purchase of the species

considered safe for human health could bring benefits due to the consumption of fish, minimizing the risks due to the intake of mercury in the diet. Avoiding the purchase of fish species with high amounts of mercury would decrease the high absorption of mercury. Basically, we will be able to target the purchase of fish to those species with less impact as regards mercury concentration.

An attempt is made to combine the eating habits of a country and the mercury level in fish. These estimates give a first glimpse into the real risk associated with fish consumption. In the future, with the addition of the data on mercury consumption of aquaculture and imported fish, the total Hg absorption and excretion rates, with the addition of socio-economic data of the population, the results presented in this chapter could provide a complete view of the mercury risk assessment for the entire population of the Western Mediterranean.

Greater rigor should be aimed at all those anthropogenic activities that dump their mercury-rich waste directly into the Mediterranean Sea or into rivers with access to this sea. The study's troubling data suggests that further remediation is needed to limit the high level of mercury already present in the Western Mediterranean compartments and biota. Some intervention aimed at decreasing the Hg in the Mediterranean Sea is strictly necessary, if we do not want to arrive in the near future to another Minamata.

Since the Minamata episodes, enormous progress has been made but it is necessary to continue in this way to avoid the dispersion of mercury in all its forms and limit the damage caused by mercury as this pollutant could still cause irreversible damage to people and the environment today.

# CHAPTER 5

GENERAL CONCLUSIONS

# CHAPTER 5

# GENERAL CONCLUSIONS

Mercury is a toxic element that can affect humans and animals. The problem of the spread of mercury in the environment should not be underestimated, given its adverse effects on nervous, immunological, and hormonal systems, already mentioned above.

Pollutant contamination starts being a local issue, but if it is extensively released, it turns into a wide context, magnifying the extent of the problem, namely in the case of chemically stable compounds such as mercury. The spread of this metal is clear as outlined in chapter 2, where it has been shown that the spillages of mercury from a chlor-alkali plant could reach 100 km downriver and pollute the marine environment. This has been evidenced from the distribution of this metal in the Flix reservoir and downriver and in the good agreement between some features of the mercury isotopic composition such as odd-MIFs. They showed the same values in the otter and fish of the Flix meander, and in the angler fish collected in L'Ampolla and Ametlla de Mar. The similarity of values is observed despite the different fish species examined, including freshwater and marine fish.

Furthermore, the mercury composition in an otter sample collected nearby Flix reservoir is also characterized by high mercury content in the muscle and liver. This mammal has a very close composition of Hg odd-MIFs to those in fish also confirms the predominant chlor-alkali source for this metal. This chlor-alkali input is affecting even an animal species that lives between terrestrial and aquatic environments.

Similar arguments can be proposed for nearly all other metal analysed. The occurrence of chromium, zinc, selenium, nickel, copper, cadmium, and arsenic in the sediments of the last stretch of the Ebro River is mostly due to spills from the chlor-alkali plant in the Flix reservoir. The distribution of lead is an exception as it reflects local sources from villages located at the shore of the Ebro River. The biota examined in the area, fish and the otter also shows a predominance of metals originating from the chlor-alkali plant, even in the case of the studied Mediterranean fish. However, in this last case, the concentrations of arsenic are not related to inputs from the Flix reservoir but to intake from marine sources.

Mercury spills from chlor-alkali plants have not only a local impact, but they affect wide environments, such as the Western Mediterranean. The present PhD dissertation has shown that mercury spills by chlor-alkali installations constitute the dominant source of this metal in this marine environment. In the locations where there was a chlor-alkali plant spill nearby, the mercury isotopic signatures of odd-MIFs in fish were very similar. Furthermore, the chlor-alkali contributions can also be observed on the fish samples from areas far from these industrial plants. A method for calculating the percentage of influence of individual sources on the mercury concentration on fish samples of the Western Mediterranean area has been elaborated. Using this method, the percentage of the contributions by the chlor-alkali plants and by atmospheric deposition plus the background on mercury in fish was calculated. The method has shown that the Mediterranean fish are affected by atmospheric deposition and background in a proportion of 0-37%, while the contributions from chlor-alkali plants range between 63% and 100%. This mass balance shows consistent results even for fish captured in the Western Mediterranean thousands of km away from chlor-alkali plants.

Having in mind these findings, the concentrations of mercury have been examined in fish from the Mediterranean Sea, destined for human consumption. Nearly 1,400 specimens from the major European countries bordering the western Mediterranean have been analysed. About one out of five fish was having mercury levels above the threshold limits indicated by the European Union as suitable for human consumption. This study also allowed to draw up a list of fish whose examined specimens fulfilled the EU recommendations in all cases. These species were: sardine, anchovie, squid, surmullet, painted comber, blackspot seabream, blue whiting, salema, brown meagre, picarel, pearly razorfish, and common dolphinfish.

On the contrary, the species showing a high percentage of individuals not fulfilling the EU recommendations were dusky grouper, european barracuda, common dentex, norway lobster, greater forkbeard, common seabream, porbeagle, and thornback ray. Another group of species with a lower but significant number of individuals not fulfilling the EU recommendations were shrimp, conger, greater weever, atlantic horse mackerel, angler, and red mullet.

The estimated weekly intake (EWI) of MeHg was calculated for the average populations of Spain, France, and Italy. All three exceed MeHg EWI levels over 144%. This estimate was based on the assumption that only local Western Mediterranean fish

was consumed by the population of these countries. It is therefore a first raw step for a complete risk assessment associated with mercury intake.

Exposure to mercury should not be underestimated, as even today it could cause irreversible damage to people and environments.

# BIBLIOGRAPHY

REFERENCES

REFERENCES


Abass, K., Huusko, A., Knutsen, H. K., Nieminen, P., Myllynen, P., Meltzer, H. M., Vahakangas, K., & Rautio, A. (2018). Quantitative estimation of mercury intake by toxicokinetic modelling based on total mercury levels in humans. *Environment International*, *114*, 1–11. https://doi.org/10.1016/j.envint.2018.02.028

Abbott, L. C., & Nigussie, F. (2021). Mercury toxicity and neurogenesis in the mammalian brain. *International Journal of Molecular Sciences*, *22 (14)*, 7520-7535. MDPI AG. https://doi.org/10.3390/ijms22147520

Ackerman, J. T., Fleck, J. A., Eagles-Smith, C. A., Marvin-DiPasquale, M., Windham-Myers, L., Herzog, M. P., & McQuillen, H. L. (2019). Wetland Management Strategy to Reduce Mercury in Water and Bioaccumulation in Fish. *Environmental Toxicology and Chemistry*, *38 (10)*, 2178–2196. https://doi.org/10.1002/etc.4535

Adams, D. H., Tremain, D. M., & Evans, D. W. (2018). Large-scale assessment of mercury in sentinel estuarine fishes of the Florida Everglades and adjacent coastal ecosystems. *Bulletin of Marine Science*, *94 (4)*, 1413–1427. https://doi.org/10.5343/bms.2017.1160

Adrian, M. I., & Delibes, M. (1987). Food habits of the otter (Lutra lutra) in two habitats of the Doñana National Park, SW Spain. *Journal of Zoology*, *212 (3)*, 399-406. https://doi.org/10.1111/j.1469-7998.1987.tb02911.x

AESAN – Agencia Española de Seguridad Alimentaria. (2006). Modelo de dieta española para la determinaciòn de la exposiciòn del consumidor a sustancias quimicas (Spanish text). *Rev. 1, 30 May 2006*, 1-33.

Afonso, C., Lourenço, H. M., Pereira, C., Martins, M. F., Carvalho, M. L., Castro, M., & Nunes, M. L. (2008). Total and organic mercury, selenium and α-tocopherol in some deep-water fish species. *Journal of the Science of Food and Agriculture*, *88 (14)*, 2543–2550. https://doi.org/10.1002/jsfa.3379

Ahmed, A. I., El-Etreby, S. G., Alwany, M. A., & Ali, R. A. (2014). Food and Feeding Habits of Sarpa salpa Salema (family: Sparidae) in the Libyan Coast of the Mediterranean Sea. *Egyptian Journal of Aquatic Biology & Fisheries*, *18 (4)*. www.ejabf.eg.net

Albo-Puigserver, M., Navarro, J., Coll, M., Layman, C. A., & Palomera, I. (2016). Trophic structure of pelagic species in the northwestern Mediterranean Sea. *Journal of Sea Research*, *117*, 27–35. https://doi.org/10.1016/j.seares.2016.09.003

Alcaraz, C., Caiola, N., & Ibáñez, C. (2011). Bioaccumulation of pollutants in the zebra mussel from hazardous industrial waste and evaluation of spatial distribution using

GAMs. *Science of the Total Environment*, *409 (5)*, 898–904. https://doi.org/10.1016/j.scitotenv.2010.11.015

Amoatey, P., & Baawain, M. S. (2019). Effects of pollution on freshwater aquatic organisms. *Water Environment Research*, *91*, 1272-1287. DOI: 10.1002/wer.1221

Amyot, M., Greg Mierle, T. S., Lean, D. R., & McQueen, D. J. (1994). Sunlight-Induced Formation of Dissolved Gaseous Mercury in Lake Waters. *Environmental Science Technology*, *28*, 2366-2371. https://pubs.acs.org/sharingguidelines

Andreji, J., Stránai, I., Massányi, P., & Valent, M. (2005). Concentration of selected metals in muscle of various fish species. *Journal of Environmental Science and Health - Part A Toxic/Hazardous Substances and Environmental Engineering*, *40 (4)*, 899–912. https://doi.org/10.1081/ESE-200048297

Andersen, A., Julshamn, K., Ringdal, O., & Morkore, J. (1987). Trace elements intake in the Faroe Islands II. Intake of mercury and other elements by consumption of pilot whales (Globicephalus meleanus). *The Science of the Total Environment*, *65*, 63-68.

Araújo, C. V. M., & Cedeño-Macias, L. A. (2016). Heavy metals in yellowfin tuna (Thunnus albacares) and common dolphinfish (Coryphaena hippurus) landed on the Ecuadorian coast. *Science of the Total Environment*, *541*, 149–154. https://doi.org/10.1016/j.scitotenv.2015.09.090

Arcagni, M., Juncos, R., Rizzo, A., Pavlin, M., Fajon, V., Arribére, M. A., Horvat, M., & Ribeiro Guevara, S. (2018). Species- and habitat-specific bioaccumulation of total mercury and methylmercury in the food web of a deep oligotrophic lake. *Science of the Total Environment*, *612*, 1311–1319. https://doi.org/10.1016/j.scitotenv.2017.08.260

Ariano, A., Marrone, R., Andreini, R., Smaldone, G., Velotto, S., Montagnaro, S., Anastasio, A., & Severino, L. (2019). Metal concentration in muscle and digestive gland of common octopus (Octopus vulgaris) from two coastal site in Southern Tyrrhenian Sea (Italy). *Molecules*, *24 (13)*. https://doi.org/10.3390/molecules24132401

Aschner, M., & Aschner, J. L. (1990). Mercury Neurotoxicity: Mechanisms of Blood-Brain Barrier Transport. *Neuroscience & Biobehavioral Reviews*, *14*, 169-176.

Aslam, S., & Yousafzai, A. M. (2017). Chromium toxicity in fish: A review article. *Journal of Entomology and Zoology Studies*, *5 (3)*, 1483–1488.

Aucott, M., McLinden, M., & Winka, M. (2003). Release of mercury from broken fluorescent bulbs. *Journal of the Air and Waste Management Association*, *53 (2)*, 143–151. https://doi.org/10.1080/10473289.2003.10466132

Azevedo, L. S., Pestana, I. A., da Costa Nery, A. F., Bastos, W. R., & Souza, C. M. M. (2019). Variation in Hg accumulation between demersal and pelagic fish from

Puruzinho Lake, Brazilian Amazon. *Ecotoxicology, 28 (10)*, 1143–1149. https://doi.org/10.1007/s10646-019-02118-x

Bagnato, E., Aiuppa, A., Parello, F., Calabrese, S., D'Alessandro, W., Mather, T. A., McGonigle, A. J. S., Pyle, D. M., & Wängberg, I. (2007). Degassing of gaseous (elemental and reactive) and particulate mercury from Mount Etna volcano (Southern Italy). *Atmospheric Environment, 41 (35)*, 7377–7388. https://doi.org/10.1016/j.atmosenv.2007.05.060

Baldi, F., & Bargagli, R. (1984). Mercury pollution in marine sediments near a chlor-alkali plant: distribution and availability of the metal. *The Science of the Total Environment, 39*, 15-26.

Baldi, F., & D'Amato, M. L. (1986). Mercury pollution in marine sediment cores near cinnabar deposits and a chlor-alkali plant. *The Science of the Total Environment, 57*, 111–120. https://doi.org/10.1016/0048-9697(86)90016-1

Banana, A. A. S., Mohamed, R. M. S. R., & Al-Gheethi, A. A. S. (2016). Mercury pollution for marine environment at Farwa Island, Libya. *Journal of Environmental Health Science and Engineering, 14 (1)*. https://doi.org/10.1186/s40201-016-0246-y

Barcia García, L., Pinzone, M., Lepoint, G., Pau, C., Das, K., & Kiszka, J. J. (2021). Factors affecting mercury concentrations in two oceanic cephalopods of commercial interest from the southern Caribbean. *Marine Pollution Bulletin, 168*, 1-7. https://doi.org/10.1016/j.marpolbul.2021.112408

Bargagli, R., Battisti, E., Focardi, S., & Formichi, P. (1993). Preliminary data on environmental distribution of mercury in northern victoria land, antarctica. *Antarctic Science, 5 (1)*, 3–8. https://doi.org/10.1017/S0954102093000021

Barkay, T., Miller, S. M., & Summers, A. O. (2003). Bacterial mercury resistance from atoms to ecosystems. *FEMS Microbiology Reviews, 27 (2–3)*, 355–384. https://doi.org/10.1016/S0168-6445(03)00046-9

Barone, G., Storelli, A., Garofalo, R., Busco, V. P., Quaglia, N. C., Centrone, G., & Storelli, M. M. (2015). Assessment of mercury and cadmium via seafood consumption in Italy: estimated dietary intake (EWI) and target hazard quotient (THQ). *Food Additives and Contaminants - Part A Chemistry, Analysis, Control, Exposure and Risk Assessment, 32 (8)*, 1277–1286. https://doi.org/10.1080/19440049.2015.1055594

Barst, B. D., Hammerschmidt, C. R., Chumchal, M. M., Muir, D. C. G., Smith, J. D., Roberts, A. P., Rainwater, T. R., & Drevnick, P. E. (2013). Determination of mercury speciation in fish tissue with a direct mercury analyzer. *Environmental Toxicology and Chemistry, 32 (6)*, 1237–1241. https://doi.org/10.1002/etc.2184

Beal, S. A., Osterberg, E. C., Zdanowicz, C. M., & Fisher, D. A. (2015). Ice Core Perspective on Mercury Pollution during the Past 600 Years. *Environmental Science and Technology*, *49 (13)*, 7641–7647. https://doi.org/10.1021/acs.est.5b01033

Bears, H., Richards, J. G., & Schulte, P. M. (2006). Arsenic exposure alters hepatic arsenic species composition and stress-mediated gene expression in the common killifish (Fundulus heteroclitus). *Aquatic Toxicology*, *77 (3)*, 257–266. https://doi.org/10.1016/j.aquatox.2005.12.008

Bełdowska, M., Jędruch, A., Łęczyński, L., Saniewska, D., & Kwasigroch, U. (2016). Coastal erosion as a source of mercury into the marine environment along the Polish Baltic shore. *Environmental Science and Pollution Research*, *23 (16)*, 16372–16382. https://doi.org/10.1007/s11356-016-6753-7

Belzile, N., & Chen, Y. W. (2017). Thallium in the environment: A critical review focused on natural waters, soils, sediments and airborne particles. *Applied Geochemistry*, *84*, 218–243. https://doi.org/10.1016/j.apgeochem.2017.06.013

Bemrah, N., Sirot, V., Leblanc, J. C., & Volatier, J. L. (2009). Fish and seafood consumption and omega 3 intake in French coastal populations: CALIPSO survey. *Public Health Nutrition*, *12 (5)*, 599–608. https://doi.org/10.1017/S1368980008002681

Bergés-Tiznado, M. E., Fernando Márquez-Farías, J., Cristina Osuna-Martínez, C., Torres-Rojas, Y. E., Galván-Magaña, F., & Páez-Osuna, F. (2019). Patterns of mercury and selenium in tissues and stomach contents of the dolphinfish Coryphaena hippurus from the SE Gulf of California, Mexico: Concentrations, biomagnification and dietary intake. *Marine Pollution Bulletin*, *138*, 84–92. https://doi.org/10.1016/j.marpolbul.2018.11.023

Bergquist, B. A., & Blum, J. D. (2007). Mass-Dependent and-Independent Fractionation of Hg Isotopes by Photoreduction in Aquatic Systems. *Science*, *318*, 417-420. http://science.sciencemag.org/

Bergquist, B. A., & Blum, J. D. (2009). The Odds and Evens of Mercury Isotopes: Applications of Mass-Dependent and Mass-Independent Isotope Fractionation. *Elements*, *5 (6)*, 353–357. DOI: https://doi.org/10.2113/gselements.5.6.353

Bernhoft, R. A. (2012). Mercury toxicity and treatment: A review of the literature. *Journal of Environmental and Public Health*, *2012*, 1-10. https://doi.org/10.1155/2012/460508

Bhave, P., & Shrestha, R. (2018). Total mercury status in an urban water body, Mithi River, Mumbai and analysis of the relation between total mercury and other pollution parameters. *Environmental Monitoring and Assessment*, *190 (12)*, 711-721. https://doi.org/10.1007/s10661-018-7080-x

Biester, H., Muller, G., Scholer, H. F., & Scholer, S. (2002). Binding and mobility of mercury in soils contaminated by emissions from chlor-alkali plants. *The Science of the Total Environment*, *284*, 191-203.

Bilandžić, N., Dokić, M., & Sedak, M. (2011). Metal content determination in four fish species from the Adriatic Sea. *Food Chemistry*, *124 (3)*, 1005–1010. https://doi.org/10.1016/j.foodchem.2010.07.060

Biswas, A., Blum, J. D., Bergquist, B. A., Keeler, G. J., & Xie, Z. (2008). Natural mercury isotope variation in coal deposits and organic soils. *Environmental Science and Technology*, *42 (22)*, 8303–8309. https://doi.org/10.1021/es801444b

Bjørklund, G., Dadar, M., Mutter, J., & Aaseth, J. (2017). The toxicology of mercury: Current research and emerging trends. *Environmental Research*, *159*, 545–554. https://doi.org/10.1016/j.envres.2017.08.051

Bloom, N.S. (1992). On the chemical form of mercury in edible fish and marine invertebrate tissue. *Canadian Journal of Fisheries and Aquatic Science*, *49*, 1010–1017.

Blum, J. D., & Bergquist, B. A. (2007). Reporting of variations in the natural isotopic composition of mercury. *Analytical and Bioanalytical Chemistry*, *388 (2)*, 353–359. https://doi.org/10.1007/s00216-007-1236-9

Blum, J. D., Popp, B. N., Drazen, J. C., Anela Choy, C., & Johnson, M. W. (2013). Methylmercury production below the mixed layer in the North Pacific Ocean. *Nature Geoscience*, *6 (10)*, 879–884. https://doi.org/10.1038/ngeo1918

Blum, J. D., Drazen, J. C., Johnson, M. W., Popp, B. N., Motta, L. C., & Jamieson, A. J. (2020). Mercury isotopes identify near-surface marine mercury in deep-sea trench biota. *PNAS*, *117 (47)*, 29292-29298. www.pnas.org/cgi/doi/10.1073/pnas.2012773117

Bolaños-Álvarez, Y., Alonso-Hernández, C. M., Morabito, R., Díaz-Asencio, M., Pinto, V., & Gómez-Batista, M. (2016). Mercury contamination of riverine sediments in the vicinity of a mercury cell chlor-alkali plant in Sagua River, Cuba. *Chemosphere*, *152*, 376–382. https://doi.org/10.1016/j.chemosphere.2016.03.025

Bonsignore, M., Tamburrino, S., Oliveri, E., Marchetti, A., Durante, C., Berni, A., Quinci, E., & Sprovieri, M. (2015). Tracing mercury pathways in Augusta Bay (southern Italy) by total concentration and isotope determination. *Environmental Pollution*, *205*, 178–185. https://doi.org/10.1016/j.envpol.2015.05.033

Bonsignore, M., Andolfi, N., Barra, M., Madeddu, A., Tisano, F., Ingallinella, V., Castorina, M., & Sprovieri, M. (2016). Assessment of mercury exposure in human populations: A status report from Augusta Bay (southern Italy). *Environmental Research*, *150*, 592–599. https://doi.org/10.1016/j.envres.2016.01.016

Bonsignore, M., Manta, D. S., Barsanti, M., Conte, F., Delbono, I., Horvat, M., Quinci, E. M., Schirone, A., Shlyapnikov, Y., & Sprovieri, M. (2020). Mercury isotope signatures in sediments and marine organisms as tracers of historical industrial pollution. *Chemosphere, 258*, 1-13. https://doi.org/10.1016/j.chemosphere.2020.127435

Bose-O'Reilly, S., McCarty, K. M., Steckling, N., & Lettmeier, B. (2010). Mercury exposure and children's health. *Current Problems in Pediatric and Adolescent Health Care, 40 (8)*, 186–215. https://doi.org/10.1016/j.cppeds.2010.07.002

Bose-O'Reilly, S., Schierl, R., Nowak, D., Siebert, U., William, J. F., Owi, F. T., & Ir, Y. I. (2016). A preliminary study on health effects in villagers exposed to mercury in a small-scale artisanal gold mining area in Indonesia. *Environmental Research, 149*, 274–281. https://doi.org/10.1016/j.envres.2016.04.007

Bowman, K. L., Hammerschmidt, C. R., Lamborg, C. H., & Swarr, G. (2015). Mercury in the North Atlantic Ocean: The U.S. GEOTRACES zonal and meridional sections. *Deep-Sea Research Part II: Topical Studies in Oceanography, 116*, 251–261. https://doi.org/10.1016/j.dsr2.2014.07.004

Brambilla, G., Abete, M. C., Binato, G., Chiaravalle, E., Cossu, M., Dellatte, E., Miniero, R., Orletti, R., Piras, P., Roncarati, A., Ubaldi, A., & Chessa, G. (2013). Mercury occurrence in Italian seafood from the Mediterranean Sea and possible intake scenarios of the Italian coastal population. *Regulatory Toxicology and Pharmacology, 65 (2)*, 269–277. https://doi.org/10.1016/j.yrtph.2012.12.009

Brand, A. F., Hynes, J., Walker, L. A., Glória Pereira, M., Lawlor, A. J., Williams, R. J., Shore, R. F., & Chadwick, E. A. (2020). Biological and anthropogenic predictors of metal concentration in the Eurasian otter, a sentinel of freshwater ecosystems. *Environmental Pollution, 266*, 1-9. https://doi.org/10.1016/j.envpol.2020.115280

Breder, R., & Flucht, R. (1984). Mercury levels in the atmosphere of various regions and locations in Italy. *The Science of the Total Environment, 40*, 231-244.

Bridges, K. N., Soulen, B. K., Overturf, C. L., Drevnick, P. E., & Roberts, A. P. (2016). Embryotoxicity of maternally transferred methylmercury to fathead minnows (Pimephales promelas). *Environmental Toxicology and Chemistry, 35 (6)*, 1436–1441. https://doi.org/10.1002/etc.3282

Brown, K. W., (2001). Workers' Health and Colonial Mercury Mining at Huancavelica, Peru. *Cambrigde University Press, 57 (4)*, 467-496. https://www.jstor.org/stable/1007830

Buchachenko, A. L., Ivanov, V. L., Roznyatovskii, V. A., Vorob'Ev, A. K., & Ustynyuk, Y. A. (2008). Inversion of the sign of the magnetic isotope effect of mercury in photolysis of substituted dibenzylmercury. *Doklady Physical Chemistry, 420 (1)*, 85–87. https://doi.org/10.1134/S0012501608050011

Burger, J., Gaines, K. F., Boring, C. S., Stephens, W. L., Snodgrass, J., & Gochfeld, M. (2001). Mercury and selenium in fish from the Savannah River: Species, trophic level, and locational differences. *Environmental Research*, *87 (2)*, 108–118. https://doi.org/10.1006/enrs.2001.4294

Burns, D. A., & Riva-Murray, K. (2018). Variation in fish mercury concentrations in streams of the Adirondack region, New York: A simplified screening approach using chemical metrics. *Ecological Indicators*, *84*, 648–661. https://doi.org/10.1016/j.ecolind.2017.09.031

Cai, X., Cai, B., Zhang, H., Chen, L., Zheng, C., Tong, P., Lin, H., Zhang, Q., Liu, M., Tong, Y., & Wang, X. (2020). Establishment of High-Resolution Atmospheric Mercury Emission Inventories for Chinese Cement Plants Based on the Mass Balance Method. *Environmental Science and Technology*, *54 (21)*, 13399–13408. https://doi.org/10.1021/acs.est.0c02963

Cairns, W. R., Turetta, C., Maffezzoli, N., Magand, O., Araujo, B. F., Angot, H., Segato, D., Cristofanelli, P., Sprovieri, F., Scarchilli, C., Grigioni, P., Ciardini, V., Barbante, C., Dommergue, A., & Spolaor, A. (2021). Mercury in precipitated and surface snow at Dome C and a first estimate of mercury depositional fluxes during the Austral summer on the high Antarctic plateau. *Atmospheric Environment*, *262*, 1-11. https://doi.org/10.1016/j.atmosenv.2021.118634

Cammilleri, G., Galluzzo, F. G., Fazio, F., Pulvirenti, A., Vella, A., Dico, G. M. lo, Macaluso, A., Ciaccio, G., & Ferrantelli, V. (2019). Mercury detection in benthic and pelagic fish collected from western sicily (Southern Italy). *Animals*, *9 (9)*, 594-599. https://doi.org/10.3390/ani9090594

Campara, F., D'andrea, P., Micciolò, R., Savonitto, C., Tansellà, M., & Zimmermann-Tansella, C. (1984). Psychological performance of workers with blood-lead concentration below the current threshold limit value. *International Archives of Occupational and Environmental Health*, *53*, 233-246.

Carbonell, G., Bravo, J. C., Fernández, C., & Tarazona, J. V. (2009). A new method for total mercury and methyl mercury analysis in muscle of seawater fish. *Bulletin of Environmental Contamination and Toxicology*, *83 (2)*, 210–213. https://doi.org/10.1007/s00128-009-9720-x

Carignan, J., Estrade, N., Sonke, J. E., & Donard, O. F. X. (2009). Odd isotope deficits in atmospheric Hg measured in lichens. *Environmental Science and Technology*, *43 (15)*, 5660–5664. https://doi.org/10.1021/es900578v

Carrasco, L., Díez, S., Soto, D. X., Catalan, J., & Bayona, J. M. (2008). Assessment of mercury and methylmercury pollution with zebra mussel (Dreissena polymorpha) in the Ebro River (NE Spain) impacted by industrial hazardous dumps. *Science of the Total Environment*, *407 (1)*, 178–184. https://doi.org/10.1016/j.scitotenv.2008.07.031

Carrasco, L., Bayona, J. M., & Díez, S. (2010). Mercury in Aquatic Organisms of the Ebro River Basin. *The Handbook of the Environmental Chemistry*, *13*, 239–258. https://doi.org/10.1007/698_2010_71

Carrasco, L., Barata, C., García-Berthou, E., Tobias, A., Bayona, J. M., & Díez, S. (2011). Patterns of mercury and methylmercury bioaccumulation in fish species downstream of a long-term mercury-contaminated site in the lower Ebro River (NE Spain). *Chemosphere*, *84 (11)*, 1642–1649. https://doi.org/10.1016/j.chemosphere.2011.05.022

Castilhos, Z., Rodrigues-Filho, S., Cesar, R., Rodrigues, A. P., Villas-Boas, R., de Jesus, I., Lima, M., Faial, K., Miranda, A., Brabo, E., Beinhoff, C., & Santos, E. (2017). Human exposure and risk assessment associated with mercury contamination in artisanal gold mining areas in the Brazilian Amazon. *Environmental Science Pollution Research*, *22*, 11255-11264. DOI: 10.1007/s11356-015-4340-y

Cebalho, E. C., Díez, S., dos Santos Filho, M., Muniz, C. C., Lázaro, W., Malm, O., & Ignácio, A. R. A. (2017). Effects of small hydropower plants on mercury concentrations in fish. *Environmental Science and Pollution Research*, *24 (28)*, 22709–22716. https://doi.org/10.1007/s11356-017-9747-1

Celo, V., Lean, D. R. S., & Scott, S. L. (2006). Abiotic methylation of mercury in the aquatic environment. *Science of the Total Environment*, *368 (1)*, 126–137. https://doi.org/10.1016/j.scitotenv.2005.09.043

Chang, L. W. (1977). Neurotoxic Effects of Mercury - A Review. *Environmental Research*, *14*, 329-373.

Cheng, H., & Hu, Y. (2010). Lead (Pb) isotopic fingerprinting and its applications in lead pollution studies in China: A review. *Environmental Pollution*, *158 (5)*, 1134–1146. https://doi.org/10.1016/j.envpol.2009.12.028

Chowdhury, T. R., Kumar Basu, G., Kumar Mandal, B., Kumar Biswas, B., Samanta, G., Kumar Chowdhury, U., Ranjan Chanda, C., Lodh, D., Lal Roy, S., Saha, C., Roy, S., Kabir, S., Quamruzzaman, Q., & Chakraborti, D. (1999). Arsenic poisoning in the Ganges delta. *Nature*, *401*, 545-546.

Choy, C. A., Popp, B. N., Kaneko, J. J., & Drazen, J. C. (2009). The influence of depth on mercury levels in pelagic fishes and their prey. *PNAS*, *106 (33)*, 13865-13869. www.pnas.org/cgi/content/full/

Cinnirella, S., Bruno, D. E., Pirrone, N., Horvat, M., Živković, I., Evers, D. C., Johnson, S., & Sunderland, E. M. (2019). Mercury concentrations in biota in the Mediterranean Sea, a compilation of 40 years of surveys. *Scientific Data*, *6(1)*, 1-11. https://doi.org/10.1038/s41597-019-0219-y

Cizdziel, J. V., Hinners, T. A., Pollard, J. E., Heithmar, E. M., & Cross, C. L. (2002). Mercury concentrations in fish from Lake Mead, USA, related to fish size,

condition, trophic level, location, and consumption risk. *Archives of Environmental Contamination and Toxicology*, *43 (3)*, 309–317. https://doi.org/10.1007/s00244-002-1191-6

Clarkson, T. W., & Magos, L. (2006). The Toxicology of Mercury and Its Chemical Compounds. *Critical Reviews in Toxicology*, *39*, 609-662. DOI: 10.1080/10408440600845619

Clearwater, S. J., Farag, A. M., & Meyer, J. S. (2002). Bioavailability and toxicity of dietborne copper and zinc to fish. *Comparative Biochemistry and Physiology Part C*, *132*, 269-313.

Coelho, J. P., Santos, H., Reis, A. T., Falcão, J., Rodrigues, E. T., Pereira, M. E., Duarte, A. C., & Pardal, M. A. (2010). Mercury bioaccumulation in the spotted dogfish (Scyliorhinus canicula) from the Atlantic Ocean. *Marine Pollution Bulletin*, *60 (8)*, 1372–1375. https://doi.org/10.1016/j.marpolbul.2010.05.008

Copat, C., Bella, F., Castaing, M., Fallico, R., Sciacca, S., & Ferrante, M. (2012). Heavy metals concentrations in fish from Sicily (Mediterranean Sea) and evaluation of possible health risks to consumers. *Bulletin of Environmental Contamination and Toxicology*, *88 (1)*, 78–83. https://doi.org/10.1007/s00128-011-0433-6

Cossa, D., & Martin, J. M. (1991). Mercury in the Rhône delta and adjacent marine areas. *Marine Chemistry*, *36 (1–4)*, 291–302. https://doi.org/10.1016/S0304-4203(09)90067-6

Cossa, D., Harmelin-Vivien, M., Mellon-Duval, C., Loizeau, V., Averty, B., Crochet, S., Chou, L., & Cadiou, J. F. (2012). Influences of bioavailability, trophic position, and growth on methylmercury in hakes (Merluccius merluccius) from Northwestern Mediterranean and Northeastern Atlantic. *Environmental Science and Technology*, *46 (9)*, 4885–4893. https://doi.org/10.1021/es204269w

Costa, F., Coelho, J. P., Baptista, J., Martinho, F., Pereira, M. E., & Pardal, M. A. (2020). Mercury accumulation in fish species along the Portuguese coast: Are there potential risks to human health? *Marine Pollution Bulletin*, *150*, 1-8. https://doi.org/10.1016/j.marpolbul.2019.110740

Costalago, D., Tecchio, S., Palomera, I., Álvarez-Calleja, I., Ospina-Álvarez, A., & Raicevich, S. (2011). Ecological understanding for fishery management: Condition and growth of anchovy late larvae during different seasons in the Northwestern Mediterranean. *Estuarine, Coastal and Shelf Science*, *93 (4)*, 350–358. https://doi.org/10.1016/j.ecss.2011.05.005

Costalago, D., Navarro, J., Álvarez-Calleja, I., & Palomera, I. (2012). Ontogenetic and seasonal changes in the feeding habits and trophic levels of two small pelagic fish species. *Marine Ecology Progress Series*, *460*, 169–181. https://doi.org/10.3354/meps09751

Costalago, D., Palomera, I., & Tirelli, V. (2014). Seasonal comparison of the diets of juvenile European anchovy Engraulis encrasicolus and sardine Sardina pilchardus in the Gulf of Lions. *Journal of Sea Research, 89,* 64–72. https://doi.org/10.1016/j.seares.2014.02.008

Covelli, S., Acquavita, A., Piani, R., Predonzani, S., & de Vittor, C. (2009). Recent contamination of mercury in an estuarine environment (Marano lagoon, Northern Adriatic, Italy). *Estuarine, Coastal and Shelf Science, 82 (2),* 273–284. https://doi.org/10.1016/j.ecss.2009.01.021

Cransveld, A., Amouroux, D., Tessier, E., Koutrakis, E., Ozturk, A. A., Bettoso, N., Mieiro, C. L., Bérail, S., Barre, J. P. G., Sturaro, N., Schnitzler, J., & Das, K. (2017). Mercury Stable Isotopes Discriminate Different Populations of European Seabass and Trace Potential Hg Sources around Europe. *Environmental Science and Technology, 51 (21),* 12219–12228. https://doi.org/10.1021/acs.est.7b01307

Craw, D., Chappell, D., & Reay, A. (2000). Environmental mercury and arsenic sources in fossil hydrothermal systems, Northland, New Zealand. *Environmental Geology, 39 (8),* 875-887.

Craw, D. (2005). Potential anthropogenic mobilisation of mercury and arsenic from soils on mineralised rocks, Northland, New Zealand. *Journal of Environmental Management, 74 (3),* 283–292. https://doi.org/10.1016/j.jenvman.2004.10.005

Da Silva, D. S., Lucotte, M., Roulet, M., Poirier, H., Mergler, D., Oliveira Santos, E., & Crossa, M. (2005). Trophic structure and bioaccumulation of mercury in fish of three natural lakes of the Brazilian amazon. *Water, Air, and Soil Pollution, 165 (1–4),* 77–94. https://doi.org/10.1007/s11270-005-4811-8

da Silva, J. M., Alves, L. M. F., Laranjeiro, M. I., Silva, A., Angélico, M. M., Norte, A. C., Lemos, M. F. L., Ramos, J. A., Novais, S. C., & Ceia, F. R. (2020). Mercury levels in commercial mid-trophic level fishes along the Portuguese coast – Relationships with trophic niche and oxidative damage. *Ecological Indicators, 116,* 1-11. https://doi.org/10.1016/j.ecolind.2020.106500

de Marco, S. G., Botté, S. E., & Marcovecchio, J. E. (2006). Mercury distribution in abiotic and biological compartments within several estuarine systems from Argentina: 1980-2005 period. *Chemosphere, 65 (2),* 213–223. https://doi.org/10.1016/j.chemosphere.2006.02.059

de Matos, L. S., Silva Correa, A. S. A., da Silva, S. A. A., Muniz, C. C., & Alves Ignacio, A. R. (2021). Mercury concentrations in fish and human health assessment in preflood phase of a hydro dam in Teles Pires River, Southern Brazilian Amazon. *Elementa, 9 (1),* art. 9, 1-13. https://doi.org/10.1525/elementa.2021.020

di Lena, G., Casini, I., Caproni, R., Fusari, A., & Orban, E. (2017). Total mercury levels in commercial fish species from Italian fishery and aquaculture. *Food Additives and*

*Contaminants: Part B Surveillance*, *10 (2)*, 118–127. https://doi.org/10.1080/19393210.2017.1281353

di Lena, G., Casini, I., Caproni, R., & Orban, E. (2018). Total mercury levels in crustacean species from Italian fishery. *Food Additives and Contaminants: Part B Surveillance*, *11 (3)*, 175–182. https://doi.org/10.1080/19393210.2018.1450302

Dixon, R., & Jones, B. (1994). Mercury Concentrations in Stomach Contents and Muscle of Five Fish Species from the North East Coast of England. *Marine Pollution Bulletin*, *28 (12)*, 741-745.

Dreiem, A., & Seegal, R. F. (2007). Methylmercury-induced changes in mitochondrial function in striatal synaptosomes are calcium-dependent and ROS-independent. *NeuroToxicology*, *28 (4)*, 720–726. https://doi.org/10.1016/j.neuro.2007.03.004

Duan, P., Khan, S., Ali, N., Shereen, M. A., Siddique, R., Ali, B., Iqbal, H. M. N., Nabi, G., Sajjad, W., & Bilal, M. (2020). Biotransformation fate and sustainable mitigation of a potentially toxic element of mercury from environmental matrices. *Arabian Journal of Chemistry*, *13 (9)*, 6949–6965. https://doi.org/10.1016/j.arabjc.2020.06.041

Duarte, B., Duarte, I. A., Caçador, I., Reis-Santos, P., Vasconcelos, R. P., Gameiro, C., Tanner, S. E., & Fonseca, V. F. (2021). Elemental fingerprinting of thornback ray (Raja clavata) muscle tissue as a tracer for provenance and food safety assessment. *Food Control*, *133*, 108592, 1-12. https://doi.org/10.1016/j.foodcont.2021.108592

EEA – European Environment Agency. (2016). Seafood in Europe. A food system approach for sustainability. *EEA report*, *No 25/2016*, 1-58.

EFSA – European Food Safety Authority. (2004). Opinion of the Scientific Panel on Contaminants in the Food Chain on a request from the Commission related to mercury and methylmercury in food. *The EFSA Journal (2004)*, *34*, 1-14.

Ehrlich, R., Robins, T., Jordaan, E. E., Miller, S., Mbuli, S., Selby, P., Wynchank, S., Cantrell, A., de Broe, M., D'Haese, P., Todd, A., & Landrigan, P. (1998). Lead absorption and renal dysfunction in a South African battery factory. *Occupational and Environmental Medicine*, *55 (7)*, 453–460. https://doi.org/10.1136/oem.55.7.453

Epov, V., Bérail, S., Pécheyran, C., Amouroux, D., & Donard, O.F.X. (2012). Isotopic Analysis via Multi-Collector Inductively Coupled Plasma Mass Spectrometry in Elemental Speciation. *Wiley-VCH*, 495-517. DOI: 10.1002/9783527650484.ch17

Ericksen, J. A., & Gustin, M. S. (2004). Foliar exchange of mercury as a function of soil and air mercury concentrations. *Science of the Total Environment*, *324 (1–3)*, 271–279. https://doi.org/10.1016/j.scitotenv.2003.10.034

EUMOFA - European Market Observatory for Fisheries and Aquaculture Products. (2019). EU Fish market. Edition 2019. 1-112. ISSN 2363-4170 ISBN 978-92-76-12178-7; DOI:10.2771/4556

EWG – Environmental Working Group (2016). U.S. Fish advice may expose babies to too much mercury. 1-20. https://ewg.org

Falcó, G., Llobet, J. M., Bocio, A., & Domingo, J. L. (2006). Daily intake of arsenic, cadmium, mercury, and lead by consumption of edible marine species. *Journal of Agricultural and Food Chemistry*, *54 (16)*, 6106–6112. https://doi.org/10.1021/jf0610110

Fanelli, E., & Cartes, J. E. (2010). Temporal variations in the feeding habits and trophic levels of three deep-sea demersal fishes from the western Mediterranean Sea, based on stomach contents and stable isotope analyses. *Marine Ecology Progress Series*, *402*, 213–232. https://doi.org/10.3354/meps08421

Fantozzi, L., Guerrieri, N., Manca, G., Orrù, A., & Marziali, L. (2021). An integrated investigation of atmospheric gaseous elemental mercury transport and dispersion around a chlor-alkali plant in the ossola valley (Italian central alps). *Toxics*, *9 (7)*, 172- 188. https://doi.org/10.3390/toxics9070172

FAO – Food and Agriculture Organization of the United Nations. (1986). Meeting on the Biogeochemical Cycle of Mercury in the Mediterranean. *FAO Fisheries Report No. 325*, *Supplement*, *FIPL/R325*.

FAO – Food and Agriculture Organization of the United Nations. (2020). The state of world fisheries and aquaculture. Sustainability in action. 1-224. https://doi.org/10.4060/ca9229en

FAO / WHO - Food and Agriculture Organization of the United Nations and World Health Organization (2011). Joint FAO / WHO expert consultation on the risks and benefits of fish consumption. *FAO Fisheries and Aquaculture Report No. 978 FIPM/R978,* 1-63.

Fard, J. H. N., Zare Javid, A., Ravanbakhsh, M., Ramezani, Z., Ahmadi, M., Angali, K. A., & Ardeshirzadeh, S. (2017). Determination of nickel and thallium concentration in Cynoglossus arel fish in Musa estuary, Persian Gulf, Iran. *Environmental Science and Pollution Research*, *24 (3)*, 2936–2945. https://doi.org/10.1007/s11356-016-8055-5

Farina, M., Brandão, R., Lara, F., Soares, F., Souza, D., & Rocha, J. (2003). Mechanisms of the inhibitory effects of selenium and mercury on the activity of d-aminolevulinate dehydratase from mouse liver, kidney and brain. *Toxicology Letters*, *139*, 55-66. www.elsevier.com/locate/toxlet

Ferrara, R., Mazzolai, B., Lanzillotta, E., Nucaro, E., & Pirrone, N. (2000). Volcanoes as emission sources of atmospheric mercury in the Mediterranean basin. *The Science of the Total Environment*, *259*, 115-121.

Ferrara, R., Lanzillotta, E., & Ceccarini, C. (2001). Dissolved gaseous mercury concentration and mercury evasional flux from seawater in front of a chlor-alkali plant. *Environmental Technology (United Kingdom)*, *22 (8)*, 971–978. https://doi.org/10.1080/09593332208618233

Ferrario, C., Finizio, A., & Villa, S. (2017). Legacy and emerging contaminants in meltwater of three Alpine glaciers. *Science of the Total Environment*, *574*, 350–357. https://doi.org/10.1016/j.scitotenv.2016.09.067

Flemming, C. A., & Trevors, J. T. (1989). Copper toxicity and chemistry in the environment: a review. *Water, Air, and Soil Pollution*, *44*, 143-158.

Foucher, D., Ogrinc, N., & Hintelmann, H. (2009). Tracing mercury contamination from the Idrija mining region (slovenia) to the gulf of trieste using Hg isotope ratio measurements. *Environmental Science and Technology*, *43 (1)*, 33–39. https://doi.org/10.1021/es801772b

Freire, C., Ramos, R., Lopez-Espinosa, M. J., Díez, S., Vioque, J., Ballester, F., & Fernández, M. F. (2010). Hair mercury levels, fish consumption, and cognitive development in preschool children from Granada, Spain. *Environmental Research*, *110 (1)*, 96–104. https://doi.org/10.1016/j.envres.2009.10.005

Friedli, H. R., Arellano, A. F., Cinnirella, S., & Pirrone, N. (2009). Initial estimates of mercury emissions to the atmosphere from global biomass burning. *Environmental Science and Technology*, *43 (10)*, 3507–3513. https://doi.org/10.1021/es802703g

Gabriel, M. C., Williamson, D. G., Brooks, S., & Lindberg, S. (2005). Atmospheric speciation of mercury in two contrasting Southeastern US airsheds. *Atmospheric Environment*, *39 (27)*, 4947–4958. https://doi.org/10.1016/j.atmosenv.2005.05.003

Gaetke, L. M., & Chow, C. K. (2003). Copper toxicity, oxidative stress, and antioxidant nutrients. *Toxicology*, *189 (1–2)*,147–163. https://doi.org/10.1016/S0300-483X(03)00159-8

Ganjavi, M., Ezzatpanah, H., Givianrad, M. H., & Shams, A. (2010). Effect of canned tuna fish processing steps on lead and cadmium contents of Iranian tuna fish. *Food Chemistry*, *118 (3)*, 525–528. https://doi.org/10.1016/j.foodchem.2009.05.018

Gantner, N., Hintelmann, H., Zheng, W., & Muir, D. C. (2009). Variations in stable isotope fractionation of Hg in food webs of Arctic lakes. *Environmental Science and Technology*, *43 (24)*, 9148–9154. https://doi.org/10.1021/es901771r

Gardner, R. M., & Nyland, J. F. (2016). Immunotoxic effects of Mercury. *Environmental Influences on the Immune System. Springer*, 273-305. DOI: 10.1007/978-3-7091-1890-0

Garg, T. K., & Chang, J. Y. (2006). Methylmercury causes oxidative stress and cytotoxicity in microglia: Attenuation by 15-deoxy-delta 12, 14-prostaglandin J2. *Journal of Neuroimmunology, 171 (1–2),* 17–28. https://doi.org/10.1016/j.jneuroim.2005.09.007

Garí, M., Grimalt, J. O., Torrent, M., & Sunyer, J. (2013). Influence of socio-demographic and diet determinants on the levels of mercury in preschool children from a Mediterranean island. *Environmental Pollution, 182,* 291–298. https://doi.org/10.1016/j.envpol.2013.07.022

Garrett, R. G. (2000). Natural sources of metals to the environment. *Human and Ecological Risk Assessment (HERA), 6 (6),* 945–963. https://doi.org/10.1080/10807030091124383

Gehrke, G. E., Blum, J. D., Slotton, D. G., & Greenfield, B. K. (2011). Mercury isotopes link mercury in san francisco bay forage fish to surface sediments. *Environmental Science and Technology, 45 (4),* 1264–1270. https://doi.org/10.1021/es103053y

Genchi, G., Sinicropi, M. S., Carocci, A., Lauria, G., & Catalano, A. (2017). Mercury exposure and heart diseases. *International Journal of Environmental Research and Public Health, 14 (1),* 74-87. https://doi.org/10.3390/ijerph14010074

Gerson, J. R., Driscoll, C. T., Hsu-Kim, H., & Bernhardt, E. S. (2018). Senegalese artisanal gold mining leads to elevated total mercury and methylmercury concentrations in soils, sediments, and rivers. *Elementa, 6,* art. 11, 1-14. https://doi.org/10.1525/elementa.274

Ghosh, N., & Bhattachara, S. (1992). Thyrotoxicity of chlorides of cadmium and mercury in rabbit. *Biomedical Environmental Science, 5,* 236-240.

Gibičar, D., Horvat, M., Logar, M., Fajon, V., Falnoga, I., Ferrara, R., Lanzillotta, E., Ceccarini, C., Mazzolai, B., Denby, B., & Pacyna, J. (2009). Human exposure to mercury in the vicinity of chlor-alkali plant. *Environmental Research, 109 (4),* 355–367. https://doi.org/10.1016/j.envres.2009.01.008

Gidlow, D. A. (2015). Lead toxicity. *Occupational Medicine, 65 (5),* 348–356. https://doi.org/10.1093/occmed/kqv018

Gilmour, C. C., Podar, M., Bullock, A. L., Graham, A. M., Brown, S. D., Somenahally, A. C., Johs, A., Hurt, R. A., Bailey, K. L., & Elias, D. A. (2013). Mercury methylation by novel microorganisms from new environments. *Environmental Science and Technology, 47 (20),* 11810–11820. https://doi.org/10.1021/es403075t

Giorgi, I., Abete, M. C., Squadrone, S., Tarasco, R., Arsieni, P., Pellegrino, M., Leogrande, M., & Prearo, M. (2009). Contaminazione da metalli pesanti nel pescato del Mar Ligure (Heavy metals contamination in fish if the Ligurian Sea (Italian text). *Associazione Italiana Veterinari Igienisti (AIVI), 6,* 68-72.

Gonzalez, D. J. X., Arain, A., & Fernandez, L. E. (2019). Mercury exposure, risk factors, and perceptions among women of childbearing age in an artisanal gold mining region of the Peruvian Amazon. *Environmental Research*, *179*, 108786, 1-9. https://doi.org/10.1016/j.envres.2019.108786

Gorbunov, A. V., Ermolaev, B. V., Lyapunov, S. M., Okina, O. I., Pavlov, S. S., & Frontasyeva, M. V. (2016). Estimation of Mercury Intake from Consumption of Fish and Seafood in Russia. *Food and Nutrition Sciences*, *7 (7)*, 516–523. https://doi.org/10.4236/fns.2016.77053

Grandjean, P., & Herz, K. T. (2011). Methylmercury and brain development: Imprecision and underestimation of developmental neurotoxicity in humans. *Mount Sinai Journal of Medicine, 78 (1)*, 107–118. https://doi.org/10.1002/msj.20228

Grgec, A. S., Kljaković-Gašpić, Z., Orct, T., Tičina, V., Sekovanić, A., Jurasović, J., & Piasek, M. (2020). Mercury and selenium in fish from the eastern part of the Adriatic Sea: A risk-benefit assessment in vulnerable population groups. *Chemosphere*, *261*, 127742, 1-9. https://doi.org/10.1016/j.chemosphere.2020.127742

Grieb, T. M., Fisher, N. S., Karimi, R., & Levin, L. (2020). An assessment of temporal trends in mercury concentrations in fish. *Ecotoxicology*, *29 (10)*, 1739–1749. https://doi.org/10.1007/s10646-019-02112-3

Grimalt, J.O., Sanchez-Cabeza, J.A., & Palanques, A. (2003). *Catalan J. ACA/CIRIT*, final report.

Gump, B. B., Dykas, M. J., MacKenzie, J. A., Dumas, A. K., Hruska, B., Ewart, C. K., Parsons, P. J., Palmer, C. D., & Bendinskas, K. (2017). Background lead and mercury exposures: Psychological and behavioral problems in children. *Environmental Research*, *158*, 576–582. https://doi.org/10.1016/j.envres.2017.06.033

Gustin, M. S. (2003). Are mercury emissions from geologic sources significant? A status report. *The Science of the Total Environment*, *304*, 153-167.

Gyamfi, O., Sørensen, P. B., Darko, G., Ansah, E., Vorkamp, K., & Bak, J. L. (2021). Contamination, exposure and risk assessment of mercury in the soils of an artisanal gold mining community in Ghana. *Chemosphere*, *267*, 128910, 1-11. https://doi.org/10.1016/j.chemosphere.2020.128910

Ha, E., Basu, N., Bose-O'Reilly, S., Dórea, J. G., McSorley, E., Sakamoto, M., & Chan, H. M. (2017). Current progress on understanding the impact of mercury on human health. *Environmental Research*, *152*, 419–433. https://doi.org/10.1016/j.envres.2016.06.042

Hajeb, P., Jinap, S., Ismail, A., Fatimah, A. B., Jamilah, B., & Abdul Rahim, M. (2009). Assessment of mercury level in commonly consumed marine fishes in Malaysia. *Food Control, 20 (1)*, 79–84. https://doi.org/10.1016/j.foodcont.2008.02.012

Hammerschmidt, C. R., & Fitzgerald, W. F. (2006). Methylmercury in freshwater fish linked to atmospheric mercury deposition. *Environmental Science and Technology, 40 (24)*, 7764–7770. https://doi.org/10.1021/es061480i

Han, D., Fu, Q., Gao, S., Zhang, X., Feng, J., Chen, X., Huang, X., Liao, H., Cheng, J., & Wang, W. (2019). Investigate the impact of local iron–steel industrial emission on atmospheric mercury concentration in Yangtze River Delta, China. *Environmental Science and Pollution Research, 26 (6)*, 5862–5872. https://doi.org/10.1007/s11356-018-3978-7

Harris, R. C., M Rudd, J. W., Amyot, M., Babiarz, C. L., Beaty, K. G., Blanchfield, P. J., Bodaly, R. A., Branfireun, B. A., Gilmour, C. C., Graydon, J. A., Heyes, A., Hintelmann, H., Hurley, J. P., Kelly, C. A., Krabbenhoft, D. P., Lindberg, S. E., Mason, R. P., Paterson, M. J., Podemski, C. L., … Tate, M. T. (2007). Whole-ecosystem study shows rapid fish-mercury response to changes in mercury deposition. *PNAS, 104 (42)*, 16586-16591. www.pnas.orgcgidoi10.1073pnas.0704186104

Has-Schön, E., Bogut, I., Kralik, G., Bogut, S., Horvatić, J., & Ĉaĉiĉ, I. (2008). Heavy metal concentration in fish tissues inhabiting waters of "Buŝko Blato" reservoar (Bosnia and Herzegovina). *Environmental Monitoring and Assessment, 144 (1–3)*, 15–22. https://doi.org/10.1007/s10661-007-9627-0

Havarinasab, S., Häggqvist, B., Björn, E., Pollard, K. M., & Hultman, P. (2005). Immunosuppressive and autoimmune effects of thimerosal in mice. *Toxicology and Applied Pharmacology, 204 (2)*, 109–121. https://doi.org/10.1016/j.taap.2004.08.019

Havelková, M., Dušek, L., Némethová, D., Poleszczuk, G., & Svobodová, Z. (2008). Comparison of mercury distribution between liver and muscle - A biomonitoring of fish from lightly and heavily contaminated localities. *Sensors, 8 (7)*, 4095–4109. https://doi.org/10.3390/s8074095

Hedgecock, I. M., Pirrone, N., Trunfio, G. A., & Sprovieri, F. (2006). Integrated mercury cycling, transport, and air-water exchange (MECAWEx) model. *Journal of Geophysical Research Atmospheres, 111 (20)*, D20302, 1-13. https://doi.org/10.1029/2006JD007117

Hogstedt, C., Hane, M., Agrell, A., & Bodin, L. (1983). Neuropsychological test results and symptoms among workers with well-defined long-term exposure to lead. *British Journal of Industrial Medicine, 40*, 99-105.

Horvat, M., Kotnik, J., Logar, M., Fajon, V., Zvonarić, T., & Pirrone, N. (2003). Speciation of mercury in surface and deep-sea waters in the Mediterranean Sea.

*Atmospheric Environment, 37 (SUPPL. 1)*, 93–108. https://doi.org/10.1016/S1352-2310(03)00249-8

Hu, J., Sun, Q., & Zhang, J. H. (2020). Critical temperature for rapid release of mercury from coal after high temperature: A review. *Journal of Cleaner Production*, *267*, 122166, 1-10. https://doi.org/10.1016/j.jclepro.2020.122166

Hylander, L. D., & Herbert, R. B. (2008). Global emission and production of mercury during the pyrometallurgical extraction of nonferrous sulfide ores. *Environmental Science and Technology*, *42 (16)*, 5971–5977. https://doi.org/10.1021/es800495g

Hyman, M. (2004). The impact of mercury on human health and the environment. *70 ALTERNATIVE THERAPIES*, *10 (6)*, 70-75. www.melisa.org

IARC - Working Group on the Evaluation of Carcinogenic Risks to Humans., World Health Organization., & International Agency for Research on Cancer. (2006). Inorganic and organic lead compunds. International Agency for Research on Cancer. *Vol. 87*, 1-529.

Ishaque, A., Ishaque, S., Arif, A., & Abbas, H. (2020). Toxic effects of lead on fish and human. *Biological and Clinical Sciences Research Journal*, *2020 (45)*, 1-7. https://doi.org/10.54112/bcsrj.v2020i1.47

ISMEA – Istituto di Servizi per il Mercato Agricolo Alimentare. (2011). Il pesce a tavola: percezioni e stili di consumo degli italiani. Perception and consume attitudes of Italian people (Italian text). Maggio 2011, 1-25. https://www.ismea.it/flex/cm/pages/ServeBLOB.php/L/IT/IDPagina/6191

Jadán-Piedra, C., Alcántara, C., Monedero, V., Zúñiga, M., Vélez, D., & Devesa, V. (2017). The use of lactic acid bacteria to reduce mercury bioaccessibility. *Food Chemistry*, *228*, 158–166. https://doi.org/10.1016/j.foodchem.2017.01.157

Jaffe, D., Prestbo, E., Swartzendruber, P., Weiss-Penzias, P., Kato, S., Takami, A., Hatakeyama, S., & Kajii, Y. (2005). Export of atmospheric mercury from Asia. *Atmospheric Environment*, *39 (17)*, 3029–3038. https://doi.org/10.1016/j.atmosenv.2005.01.030

Janssen, S. E., Schaefer, J. K., Barkay, T., & Reinfelder, J. R. (2016). Fractionation of Mercury Stable Isotopes during Microbial Methylmercury Production by Iron- and Sulfate-Reducing Bacteria. *Environmental Science and Technology*, *50 (15)*, 8077–8083. https://doi.org/10.1021/acs.est.6b00854

Jardine, L. B., Burt, M. D. B., Arp, P. A., & Diamond, A. W. (2009). Mercury comparisons between farmed and wild Atlantic salmon (Salmo salar L.) and atlantic cod (Gadus morhua L.). *Aquaculture Research*, *40 (10)*, 1148–1159. https://doi.org/10.1111/j.1365-2109.2009.02211.x

Jenssen, M. T. S., Brantsæter, A. L., Haugen, M., Meltzer, H. M., Larssen, T., Kvalem, H. E., Birgisdottir, B. E., Thomassen, Y., Ellingsen, D., Alexander, J., & Knutsen,

H. K. (2012). Dietary mercury exposure in a population with a wide range of fish consumption - Self-capture of fish and regional differences are important determinants of mercury in blood. *Science of the Total Environment*, *439*, 220–229. https://doi.org/10.1016/j.scitotenv.2012.09.024

Johansson, K., Bergbäck, B. & Tyler, G. (2001). Impact of Atmospheric Long Range Transport of Lead, Mercury and Cadmium on the Swedish Forest Environment. *Water, Air, & Soil Pollution: Focus*, *1*, 279–297. https://doi.org/10.1023/A:1017528826641

Jiskra, M., Wiederhold, J. G., Skyllberg, U., Kronberg, R. M., & Kretzschmar, R. (2017). Source tracing of natural organic matter bound mercury in boreal forest runoff with mercury stable isotopes. *Environmental Science: Processes and Impacts*, *19 (10)*, 1235–1248. https://doi.org/10.1039/c7em00245a

Joiris, C. R., Ali, I. B., Holsbeek, L., Bossicart, M., & Tapia, G. (1995). Total and Organic Mercury in Barents Sea Pelagic Fish. *Bulletin Environmental Contamination Toxicology*, *55*, 674-681.

Jordao, C. P., Pereira, J. L., & Jham, G. N. (1997). Chromium contamination in sediment, vegetation and fish caused by tanneries in the State of Minas Gerais, Brazil. *The Science of the Total Environment*, *207*, 1-11.

Junqué, E., Garí, M., Arce, A., Torrent, M., Sunyer, J., & Grimalt, J. O. (2017). Integrated assessment of infant exposure to persistent organic pollutants and mercury via dietary intake in a central western Mediterranean site (Menorca Island). *Environmental Research*, *156*, 714–724. https://doi.org/10.1016/j.envres.2017.04.030

Junqué, E., Garí, M., Llull, R. M., & Grimalt, J. O. (2018). Drivers of the accumulation of mercury and organochlorine pollutants in Mediterranean lean fish and dietary significance. *Science of the Total Environment*, *634*, 170–180. https://doi.org/10.1016/j.scitotenv.2018.03.335

Kalogeropoulos, N., Karavoltsos, S., Sakellari, A., Avramidou, S., Dassenakis, M., & Scoullos, M. (2012). Heavy metals in raw, fried and grilled Mediterranean finfish and shellfish. *Food and Chemical Toxicology*, *50 (10)*, 3702–3708. https://doi.org/10.1016/j.fct.2012.07.012

Karachle, P. K., & Stergiou, K. I. (2014). Diet and feeding habits of Spicara maena and S. smaris in the North Aegean Sea. *Acta Adriatica*, *55 (1)*, 75–84.

Karagas, M. R., Choi, A. L., Oken, E., Horvat, M., Schoeny, R., Kamai, E., Cowell, W., Grandjean, P., & Korrick, S. (2012). Evidence on the human health effects of low-level methylmercury exposure. *Environmental Health Perspectives*, *120 (6)*, 799–806. https://doi.org/10.1289/ehp.1104494

Keskin, Y., Baskaya, R., Özyaral, O., Yurdun, T., Lüleci, N. E., & Hayran, O. (2007). Cadmium, lead, mercury and copper in fish from the Marmara Sea, Turkey. *Bulletin of Environmental Contamination and Toxicology*, *78 (3–4)*, 258–261. https://doi.org/10.1007/s00128-007-9123-9

Kim, J. H., & Kang, J. C. (2015). The lead accumulation and hematological findings in juvenile rock fish Sebastes schlegelii exposed to the dietary lead (II) concentrations. *Ecotoxicology and Environmental Safety*, *115*, 33–39. https://doi.org/10.1016/j.ecoenv.2015.02.009

Kim, J. H., & Kang, J. C. (2016). The immune responses in juvenile rockfish, Sebastes schlegelii for the stress by the exposure to the dietary lead (II). *Environmental Toxicology and Pharmacology*, *46*, 211–216. https://doi.org/10.1016/j.etap.2016.07.022

Kim, K. H., & Kim, M. Y. (2002). Mercury emissions as landfill gas from a large-scale abandoned landfill site in Seoul. *Atmospheric Environment*, *36*, 4919-4928.

Kim, C. K., Lee, T. W., Lee, K. T., Lee, J. H., & Lee, C. B. (2012). Nationwide monitoring of mercury in wild and farmed fish from fresh and coastal waters of Korea. *Chemosphere*, *89 (11)*, 1360–1368. https://doi.org/10.1016/j.chemosphere.2012.05.093

Kim, S. A., Kwon, Y. M., Kim, S., & Joung, H. (2016). Assessment of dietary mercury intake and blood mercury levels in the Korean population: Results from the Korean National Environmental Health Survey 2012-2014. *International Journal of Environmental Research and Public Health*, *13 (9)*, 877-890. https://doi.org/10.3390/ijerph13090877

Kogut, K., Górecki, J., & Burmistrz, P. (2021). Opportunities for reducing mercury emissions in the cement industry. *Journal of Cleaner Production*, *293*, 126053, 1-11. https://doi.org/10.1016/j.jclepro.2021.126053

Kojadinovic, J., Potier, M., le Corre, M., Cosson, R. P., & Bustamante, P. (2006). Mercury content in commercial pelagic fish and its risk assessment in the Western Indian Ocean. *Science of the Total Environment*, *366 (2–3)*, 688–700. https://doi.org/10.1016/j.scitotenv.2006.02.006

Kojadinovic, J., Potier, M., le Corre, M., Cosson, R. P., & Bustamante, P. (2007). Bioaccumulation of trace elements in pelagic fish from the Western Indian Ocean. *Environmental Pollution*, *146 (2)*, 548–566. https://doi.org/10.1016/j.envpol.2006.07.015

Köker, L., Aydın, F., Gaygusuz, Ö., Akçaalan, R., Çamur, D., İlter, H., Ayoğlu, F. N., Altın, A., Topbaş, M., & Albay, M. (2021). Heavy Metal Concentrations in Trachurus Mediterraneus and Merlangius Merlangus Captured from Marmara Sea, Turkey and Associated Health Risks. *Environmental Management*, *67 (3)*, 522–531. https://doi.org/10.1007/s00267-020-01352-y

Kotnik, J., Horvat, M., Tessier, E., Ogrinc, N., Monperrus, M., Amouroux, D., Fajon, V., Gibičar, D., Žižek, S., Sprovieri, F., & Pirrone, N. (2007). Mercury speciation in surface and deep waters of the Mediterranean Sea. *Marine Chemistry, 107 (1)*, 13–30. https://doi.org/10.1016/j.marchem.2007.02.012

Kritee, K., Blum, J. D., Johnson, M. W., Bergquist, B. A., & Barkay, T. (2007). Mercury stable isotope fractionation during reduction of Hg(II) to Hg(0) by Mercury resistant microorganisms. *Environmental Science and Technology, 41 (6)*, 1889–1895. https://doi.org/10.1021/es062019t

Kumar, P., & Singh, A. (2010). Cadmium toxicity in fish: An overview. *GERF Bulletin of Biosciences, 1 (1)*, 41-47. www.gerfbb.com

Kumari, B., Kumar, V., Sinha, A. K., Ahsan, J., Ghosh, A. K., Wang, H., & DeBoeck, G. (2017). Toxicology of arsenic in fish and aquatic systems. *Environmental Chemistry Letters, 15 (1)*, 43–64. https://doi.org/10.1007/s10311-016-0588-9

Kuplulu, O., Iplikcioglu Cil, G., Korkmaz, S. D., Aykut, O., & Ozansoy G. (2018). Determination of Metal Contamination in Seafood from the Black, Marmara, Aegean and Mediterranean Sea Metal Contamination in Seafood. *Journal of the Hellenic Veterinary Medical Society, 69 (1)*, 749-758.

Kuss, J., Krüger, S., Ruickoldt, J., & Wlost, K. P. (2018). High-resolution measurements of elemental mercury in surface water for an improved quantitative understanding of the Baltic Sea as a source of atmospheric mercury. *Atmospheric Chemistry and Physics, 18 (6)*, 4361–4376. https://doi.org/10.5194/acp-18-4361-2018

Kwasigroch, U., Bełdowska, M., Jędruch, A., & Saniewska, D. (2018). Coastal erosion—a "new" land-based source of labile mercury to the marine environment. *Environmental Science and Pollution Research, 25 (28)*, 28682–28694. https://doi.org/10.1007/s11356-018-2856-7

Kwon, S. Y., Blum, J. D., Carvan, M. J., Basu, N., Head, J. A., Madenjian, C. P., & David, S. R. (2012). Absence of fractionation of mercury isotopes during trophic transfer of methylmercury to freshwater fish in captivity. *Environmental Science and Technology, 46 (14)*, 7527–7534. https://doi.org/10.1021/es300794q

Kwon, S. Y., Blum, J. D., Chirby, M. A., & Chesney, E. J. (2013). Application of mercury isotopes for tracing trophic transfer and internal distribution of mercury in marine fish feeding experiments. *Environmental Toxicology and Chemistry, 32 (10)*, 2322–2330. https://doi.org/10.1002/etc.2313

Kwon, S. Y., Blum, J. D., Chen, C. Y., Meattey, D. E., & Mason, R. P. (2014). Mercury isotope study of sources and exposure pathways of methylmercury in estuarine food webs in the northeastern U.S. *Environmental Science and Technology, 48 (17)*, 10089–10097. https://doi.org/10.1021/es5020554

Kwon, S. Y., Blum, J. D., Yin, R., Tsui, M. T. K., Yang, Y. H., & Choi, J. W. (2020). Mercury stable isotopes for monitoring the effectiveness of the Minamata Convention on Mercury. *Earth-Science Reviews, 203*, 103111, 1-22. https://doi.org/10.1016/j.earscirev.2020.103111

La Colla, N. S., Botté, S. E., Simonetti, P., Negrin, V. L., Serra, A. V., & Marcovecchio, J. E. (2021). Water, sediments and fishes: First multi compartment assessment of metal pollution in a coastal environment from the SW Atlantic. *Chemosphere, 282*, 131131, 1-11. https://doi.org/10.1016/j.chemosphere.2021.131131

Laffont, L., Sonke, J. E., Maurice, L., Hintelmann, H., Pouilly, M., Bacarreza, Y. S., Perez, T., & Behra, P. (2009). Anomalous mercury isotopic compositions of fish and human hair in the Bolivian amazon. *Environmental Science and Technology, 43 (23)*, 8985–8990. https://doi.org/10.1021/es9019518

Lahaye, V., Bustamante, P., Dabin, W., van Canneyt, O., Dhermain, F., Cesarini, C., Pierce, G. J., & Caurant, F. (2006). New insights from age determination on toxic element accumulation in striped and bottlenose dolphins from Atlantic and Mediterranean waters. *Marine Pollution Bulletin, 52 (10)*, 1219–1230. https://doi.org/10.1016/j.marpolbul.2006.02.020

Lai, B., Murthy, R. C., Anand, M., Chandra, S. V., Kumar, R., Tripathi, O., & Srimal, R. C. (1991). Cardiotoxicity and Hypertension in Rats After Oral Lead Exposure, *Drug and Chemical Toxicology, 14:3*, 305-318. DOI: 10.3109/01480549109002192

Lamborg, C. H., Fitzgerald, W. F., O'donnell, J., & Torgersen, T. (2002). A non-steady-state compartmental model of global-scale mercury biogeochemistry with interhemispheric atmospheric gradients. *Geochimica et Cosmochimica Acta, 66 (7)*, 1105-1118.

Lamperti, A., & Niewenhuis, R. (1976). The Effects of Mercury on the Structure and Function of the Hypothalamo-Pituitary Axis in the Hamster. *Cell and Tissue Research, 170*, 315-324.

Larsen, E. H., & Francesconi, K. A. (2003). Arsenic concentrations correlate with salinity for fish taken from the North Sea and Baltic waters. *Journal of the Marine Biological Association of the United Kingdom, 83 (2)*, 283–284. https://doi.org/10.1017/S0025315403007082h

Latif, M. A., Bodaly, R. A., Johnston, T. A., & Fudge, R. J. P. (2001). E□ects of environmental and maternally derived methylmercury on the embryonic and larval stages of walleye (Stizostedion vitreum). *Environmental Pollution, 111*, 139-148.

Le Croizier, G., Schaal, G., Point, D., le Loc'h, F., Machu, E., Fall, M., Munaron, J. M., Boyé, A., Walter, P., Laë, R., & Tito De Morais, L. (2019). Stable isotope analyses revealed the influence of foraging habitat on mercury accumulation in tropical

coastal marine fish. *Science of the Total Environment, 650*, 2129–2140. https://doi.org/10.1016/j.scitotenv.2018.09.330

Lee, B. J., Kwon, S. Y., Yin, R., Li, M., Jung, S., Lim, S. H., Lee, J. H., Kim, K. W., Kim, K. D., & Jang, J. W. (2020). Internal dynamics of inorganic and methylmercury in a marine fish: Insights from mercury stable isotopes. *Environmental Pollution, 267*, 115588, 1-8. https://doi.org/10.1016/j.envpol.2020.115588

Lehnherr, I., & st. Louis, V. L. (2009). Importance of ultraviolet radiation in the photodemethylation of methylmercury in freshwater ecosystems. *Environmental Science and Technology, 43 (15)*, 5692–5698. https://doi.org/10.1021/es9002923

Lemly, A. D. (1993). Metabolic stress during winter increases the toxicity of selenium to fish. *Aquatic Toxicology, 27*, 133-158.

Lepak, R. F., Janssen, S. E., Yin, R., Krabbenhoft, D. P., Ogorek, J. M., Dewild, J. F., Tate, M. T., Holsen, T. M., & Hurley, J. P. (2018). Factors Affecting Mercury Stable Isotopic Distribution in Piscivorous Fish of the Laurentian Great Lakes. *Environmental Science and Technology, 52 (5)*, 2768–2776. https://doi.org/10.1021/acs.est.7b06120

Lescord, G. L., Johnston, T. A., Branfireun, B. A., & Gunn, J. M. (2018). Percentage of methylmercury in the muscle tissue of freshwater fish varies with body size and age and among species. *Environmental Toxicology and Chemistry, 37 (10)*, 2682–2691. https://doi.org/10.1002/etc.4233

Li, M., Schartup, A. T., Valberg, A. P., Ewald, J. D., Krabbenhoft, D. P., Yin, R., Balcom, P. H., & Sunderland, E. M. (2016). Environmental Origins of Methylmercury Accumulated in Subarctic Estuarine Fish Indicated by Mercury Stable Isotopes. *Environmental Science and Technology, 50 (21)*, 11559–11568. https://doi.org/10.1021/acs.est.6b03206

Li, G., Wu, Q., Wang, S., Li, Z., Liang, H., Tang, Y., Zhao, M., Chen, L., Liu, K., & Wang, F. (2017). The influence of flue gas components and activated carbon injection on mercury capture of municipal solid waste incineration in China. *Chemical Engineering Journal, 326*, 561–569. https://doi.org/10.1016/j.cej.2017.05.099

Liao, Y., Xu, H., Liu, W., Ni, H., Zhang, X., Zhai, A., Quan, Z., Qu, Z., & Yan, N. (2019). One Step Interface Activation of ZnS Using Cupric Ions for Mercury Recovery from Nonferrous Smelting Flue Gas. *Environmental Science and Technology, 53 (8)*, 4511–4518. https://doi.org/10.1021/acs.est.9b01310

Lis, J., Pasieczna, A., Karbowska, B., Zembrzuski, W., & Lukaszewski, Z. (2003). Thallium in soils and stream sediments of a Zn-Pb mining and smelting area. *Environmental Science and Technology, 37 (20)*, 4569–4572. https://doi.org/10.1021/es0346936

Liu, J., Xu, X., Yu, S., Cheng, H., Hong, Y., & Feng, X. (2014). Mercury pollution in fish from South China Sea: Levels, species-specific accumulation, and possible sources. *Environmental Research*, *131*, 160–164. https://doi.org/10.1016/j.envres.2014.03.004

Liu, K., Wu, Q., Wang, L., Wang, S., Liu, T., Ding, D., Tang, Y., Li, G., Tian, H., Duan, L., Wang, X., Fu, X., Feng, X., & Hao, J. (2019). Measure-Specific Effectiveness of Air Pollution Control on China's Atmospheric Mercury Concentration and Deposition during 2013-2017. *Environmental Science and Technology*, *53 (15)*, 8938–8946. https://doi.org/10.1021/acs.est.9b02428

Llop, S., Ballester, F., & Broberg, K. (2015). Effect of Gene-Mercury Interactions on Mercury Toxicokinetics and Neurotoxicity. *Current Environmental Health Reports*, *2 (2)*, 179–194. https://doi.org/10.1007/s40572-015-0047-y

Llull, R. M., Garí, M., Canals, M., Rey-Maquieira, T., & Grimalt, J. O. (2017). Mercury concentrations in lean fish from the Western Mediterranean Sea: Dietary exposure and risk assessment in the population of the Balearic Islands. *Environmental Research*, *158*, 16–23. https://doi.org/10.1016/j.envres.2017.05.033

Maas, C., Bruck, W., Haffner, H.T., & Schweinsberg, F. (1996). Study on the significance of mercury accumulation in the brain from dental amalgam fillings through direct mouth-nose-brain transport. *Zentralbl Hyg Umweltmed*, *198(3)*, 275–291.

Madigan, D. J., Li, M., Yin, R., Baumann, H., Snodgrass, O. E., Dewar, H., Krabbenhoft, D. P., Baumann, Z., Fisher, N. S., Balcom, P., & Sunderland, E. M. (2018). Mercury Stable Isotopes Reveal Influence of Foraging Depth on Mercury Concentrations and Growth in Pacific Bluefin Tuna. *Environmental Science and Technology*, *52 (11)*, 6256–6264. https://doi.org/10.1021/acs.est.7b06429

Mahon, R., & Oxenford, H. A. (1999). Precautionary assessment and management of dolphinfish in the Caribbean. *Scientia Marina*, *63 (3-4)*, 429-438.

Maruyama, K., Yorifuji, T., Tsuda, T., Sekikawa, T., Nakadaira, H., & Saito, H. (2012). Methyl mercury exposure at Niigata, Japan: Results of neurological examinations of 103 adults. *Journal of Biomedicine and Biotechnology*, *2012*, 635075, 1-8. https://doi.org/10.1155/2012/635075

Marvin-DiPasquale, M. C., & Oremland, R. S. (1998). Bacterial Methylmercury Degradation in Florida Everglades Peat Sediment. *Environmental Science Technology*, *32*, 2556-2563.

Maserti, B. E., & Ferrara, R. (1991). Mercury in plants, soil and atmosphere near a chlor-alkali complex. *Water, Air, and Soil Pollution*, *56*, 15-20.

Mason, C. F., & Stephenson, A. (2001). Metals in tissues of European otters (Lutra lutra) from Denmark, Great Britain and Ireland. *Chemosphere*, *44*, 351-353.

Mason, R. P., & Sheu, G. R. (2002). Role of the ocean in the global mercury cycle. *Global Biogeochemical Cycles, 16 (4),* 40, 1-14. https://doi.org/10.1029/2001gb001440

Mason, R. P., Fitzgerald, W. F., & Morel, F. M. M. (1994). The biogeochemical cycling of elemental mercury: Anthropogenic influences. *Geochimica et Cosmochimica Acta, 58 (15),* 3191-3198.

Mason, R. P., Abbott, M. L., Bodaly, R. A., Bullock jr, O. R., Driscoll, C. T., Evers, D., Lindberg, S. E., Murray, M., & Swain, E. B. (2005). Monitoring the Response to Changing Mercury Deposition. *Environmental Science and Technology, January 1, 2005,* 14-22.

Mason, R. P., Choi, A. L., Fitzgerald, W. F., Hammerschmidt, C. R., Lamborg, C. H., Soerensen, A. L., & Sunderland, E. M. (2012). Mercury biogeochemical cycling in the ocean and policy implications. *Environmental Research, 119,* 101–117. https://doi.org/10.1016/j.envres.2012.03.013

Matic-Skoko, S., Kraljevic, M., Dulcic, J., & Pallaoro, A. (2004). Growth of juvenile salema, Sarpa salpa (Teleostei: Sparidae), in the Kornati Archipelago, eastern Adriatic Sea. *Scientia Marina, 68 (3),* 411-417.

Mauchline, J., & Gordon, J. D. M. (1986). Foraging strategies of deep-sea fish. *Marine ecology – progress series, 27,* 227-238.

Maurice-Bourgoin, L., Quiroga, I., Chincheros, J., & Courau, P. (2000). Mercury distribution in waters and fishes of the upper Madeira rivers and mercury exposure in riparian Amazonian populations. *The Science of the Total Environment, 260,* 73-86.

Mazumder, D. N. G. (2008). Chronic arsenic toxicity & human health. *Indian Journal Medical Research, 128,* 436-447.

Melendez-Perez, J. J., Fostier, A. H., Carvalho, J. A., Windmöller, C. C., Santos, J. C., & Carpi, A. (2014). Soil and biomass mercury emissions during a prescribed fire in the Amazonian rain forest. *Atmospheric Environment, 96,* 415–422. https://doi.org/10.1016/j.atmosenv.2014.06.032

Mezghani-Chaari, S., Hamza, A., & Hamza-Chaffai, A. (2011). Mercury contamination in human hair and some marine species from Sfax coasts of Tunisia: Levels and risk assessment. *Environmental Monitoring and Assessment, 180 (1–4),* 477–487. https://doi.org/10.1007/s10661-010-1800-1

Mieiro, C. L., Pacheco, M., Pereira, M. E., & Duarte, A. C. (2009). Mercury distribution in key tissues of fish (Liza aurata) inhabiting a contaminated estuary - Implications for human and ecosystem health risk assessment. *Journal of Environmental Monitoring, 11 (5),* 1004–1012. https://doi.org/10.1039/b821253h

Milanov, D. R., Krstić, P. M., Marković, V. R., Jovanović, A. D., Baltić, M. B., Ivanović, S. J., Jovetić, M., & Baltić, M. (2016). Analysis of heavy metals concentration in

tissues of three different fish species included in human diet from Danube River, in the Belgrade Region, Serbia. *Acta Veterinaria, 66 (1)*, 89–102. https://doi.org/10.1515/acve-2016-0007

Minet, A., Manceau, A., Valada-Mennuni, A., Brault-Favrou, M., Churlaud, C., Fort, J., Nguyen, T., Spitz, J., Bustamante, P., & Lacoue-Labarthe, T. (2021). Mercury in the tissues of five cephalopods species: First data on the nervous system. *Science of the Total Environment*, *759*. https://doi.org/10.1016/j.scitotenv.2020.143907

Mir-Arguimbau, J., Navarro, J., Balcells, M., Martín, P., & Sabatés, A. (2020). Feeding ecology of blue whiting (Micromesistius poutassou) in the NW Mediterranean: The important role of Myctophidae. *Deep-Sea Research Part I: Oceanographic Research Papers*, *166*, 1-16. https://doi.org/10.1016/j.dsr.2020.103404

Moller-Madsen, B., & Thorlacius-Ussing, O. (1986). Accumulation of mercury in the anterior pituitary of rats following oral intraperitoneal administration of methyl mercury. *Virchows Arch [Cell Pathology]*, *51*, 303-311.

Monperrus, M., Tessier, E., Amouroux, D., Leynaert, A., Huonnic, P., & Donard, O. F. X. (2007). Mercury methylation, demethylation and reduction rates in coastal and marine surface waters of the Mediterranean Sea. *Marine Chemistry*, *107 (1)*, 49–63. https://doi.org/10.1016/j.marchem.2007.01.018

Morel, F. M. M., Kraepiel, A. M. L., & Amyot, M. (1998). The chemical cycle and bioaccumulation of mercury. *Annual Review of Ecology, Evolution and Systematics*, *29*, 543-566.

Morote, E., Pilar Olivar, M., Villate, F., & Uriarte, I. (2010). A comparison of anchovy (Engraulis encrasicolus) and sardine (Sardina pilchardus) larvae feeding in the Northwest Mediterranean: influence of prey availability and ontogeny. *International Council for the Exploration of the Sea. Oxford Journals*, 897- 908. DOI:10.1093/icesjms/fsp302

Motta, L. C., Blum, J. D., Popp, B. N., Drazen, J. C., & Close, H. G. (2020). Mercury stable isotopes in flying fish as a monitor of photochemical degradation of methylmercury in the Atlantic and Pacific Oceans. *Marine Chemistry*, *223*, 103790, 1-8. https://doi.org/10.1016/j.marchem.2020.103790

Mukherjee, D., Kumar, V., & Chakraborti, P. (1994). Effect of mercury chloride and cadmium chloride on gonadal function and its regulation in sexually mature common carp Cyprinus carpio. *Biomedical Environmental Science*, *7*, 13-24.

Mukherjee, A. B., Zevenhoven, R., Bhattacharya, P., Sajwan, K. S., & Kikuchi, R. (2008). Mercury flow via coal and coal utilization by-products: A global perspective. *Resources, Conservation and Recycling*, *52(4)*, 571–591. https://doi.org/10.1016/j.resconrec.2007.09.002

Muyssen, B. T. A., Brix, K. V., DeForest, D. K., & Janssen, C. R. (2004). Nickel essentiality and homeostasis in aquatic organisms. *Environmental Reviews*, *12 (2)*, 113–131. https://doi.org/10.1139/A04-004

Naccari, C., Cicero, N., Ferrantelli, V., Giangrosso, G., Vella, A., Macaluso, A., Naccari, F., & Dugo, G. (2015). Toxic Metals in Pelagic, Benthic and Demersal Fish Species from Mediterranean FAO Zones 37. *Bulletin of the Environmental Contamination and Toxicology*, *95*, 567-573. DOI 10.1007/s00128-015-1585-6

Nacht, D. M., Gustin, M. S., Engle, M. A., Zehner, R. E., & Giglini, A. D. (2004). Atmospheric Mercury Emissions and Speciation at the Sulphur Bank Mercury Mine Superfund Site, Northern California. *Environmental Science and Technology*, *38 (7)*, 1977–1983. https://doi.org/10.1021/es0304244

Nagorski, S. A., Vermilyea, A. W., & Lamborg, C. H. (2021). Mercury export from glacierized Alaskan watersheds as influenced by bedrock geology, watershed processes, and atmospheric deposition. *Geochimica et Cosmochimica Acta*, *304*, 32–49. https://doi.org/10.1016/j.gca.2021.04.003

Navarro, A., Quirós, L., Casado, M., Faria, M., Carrasco, L., Benejam, L., Benito, J., Díez, S., Raldúa, D., Barata, C., Bayona, J. M., & Piña, B. (2009). Physiological responses to mercury in feral carp populations inhabiting the low Ebro River (NE Spain), a historically contaminated site. *Aquatic Toxicology*, *93 (2–3)*, 150–157. https://doi.org/10.1016/j.aquatox.2009.04.009

Navarro, J., Coll, M., Louzao, M., Palomera, I., Delgado, A., & Forero, M. G. (2011). Comparison of ecosystem modelling and isotopic approach as ecological tools to investigate food webs in the NW Mediterranean Sea. *Journal of Experimental Marine Biology and Ecology*, *401 (1–2)*, 97–104. https://doi.org/10.1016/j.jembe.2011.02.040

Navrátil, T., Nováková, T., Shanley, J. B., Rohovec, J., & Vaňková, M. (2021). Distribution and pools of mercury in forest soils near recent and historical mercury emission sources in the central Czech Republic. *Journal of Geochemical Exploration*, *226*, 106782, 1-13. https://doi.org/10.1016/j.gexplo.2021.106782

Ng, T.B., & Liu, W.K. (1990). Toxic effect of heavy metals on cells isolated from the rat adrenal and testis. *In Vitro Cellular Biology*, *26*, 24-28.

NIH - National Institutes of Health. (2021). https://ods.od.nih.gov/factsheets/Selenium-HealthProfessional/#h7

NIVA – Norwegian Institute for Water Research. (2017). Spatial and temporal trends of mercury in freshwater fish in Fennoscandia (1965-2015). *ICP Waters report 132/2017, No. 7179-2017*, 1-66. www.niva.no

Nriagu, J., & Becker, C. (2003). Volcanic emissions of mercury to the atmosphere: global and regional inventories. *The Science of the Total Environment*, *304*, 3-12.

Nylander, M., & Weiner, J. (1989). Relation between mercury and selenium in pituitary glands of dental staff. *British journal of industrial medicine*, *46 (10)*, 751-752. ncbi.nlm.nih.gov

O'Flaherty, E. J. (1995). Physiologically Based Models for Bone-Seeking Elements. *Toxicology and applied pharmacology*, *131*, 297-305.

Ogrinc, N., Monperrus, M., Kotnik, J., Fajon, V., Vidimova, K., Amouroux, D., Kocman, D., Tessier, E., Zizek, S., & Horvat, M. (2007). Distribution of mercury and methylmercury in deep-sea superficial sediments of the Mediterranean Sea. *Marine Chemistry*, *107*, 31-48.

Ogrinc, N., Hintelmann, H., Kotnik, J., Horvat, M., & Pirrone, N. (2019). Sources of mercury in deep-sea sediments of the Mediterranean Sea as revealed by mercury stable isotopes. *Scientific Reports*, *9 (1)*, 11626, 1-9. https://doi.org/10.1038/s41598-019-48061-z

OJEU – Official Journal of the European Union (2006). Commission regulation (EC) No 1881/2006. *L 364/5*, *20.12.2006*, 1-20. https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A32006R1881

Okada, I. A., Sakuma, A. M., Maio, F. D., & Dovidauskas Odair Zenebon, S. (1997). Evaluation of lead and cadmium levels in milk due to environmental contamination in the Paraiba Valley region of Southeastern Brazil (Portuguese text). *Revista de Saùde Pùblica*, *31 (2)*, 140-143.

Okocha, R., & Adedeji, O. (2011). Overview of cadmium toxicity in fish. *Journal of Applied Sciences Research*, *7 (7)*, 1195-1207. https://www.researchgate.net/publication/279602220

Ou, Y.C., White, C.C., Krejsa, C.M., Ponce, R.A., Kavanagh, T.J., & Faustman, E.M. (1999). The role of intracellular glutathione in methylmercury-induced toxicity in embryonic neuronal cells. *NeuroToxicology*, *20(5)*, 793–804.

Özden, Ö. (2013). Monitoring programme on toxic metal in bluefish (Pomatomus saltatrix), anchovy (Engraulis encrasicolus) and sardine (Sardina pilchardus) from Istanbul, Turkey: Levels and estimated weekly intake. *Bulletin of Environmental Contamination and Toxicology*, *90 (5)*, 542–551. https://doi.org/10.1007/s00128-012-0958-3

Pacyna, E. G., Pacyna, J. M., Steenhuisen, F., & Wilson, S. (2006). Global anthropogenic mercury emission inventory for 2000. *Atmospheric Environment*, *40 (22)*, 4048–4063. https://doi.org/10.1016/j.atmosenv.2006.03.041

Palanques, A., Puig, P., Guillen, J., Querol, X., & Alastuey, A. (1999). Zinc contamination in the bottom and suspended sediments of the Guadalquivir estuary after the Aznalcollar spill (south-western Spain). Control of hydrodynamic processes. *The Science of the Total Environment*, *242*, 211-220.

Palanques, A., Guillén, J., Puig, P., & Grimalt, J. O. (2020). Effects of flushing flows on the transport of mercury-polluted particulate matter from the Flix Reservoir to the Ebro Estuary. *Journal of Environmental Management*, *260*, 110028, 1-14. https://doi.org/10.1016/j.jenvman.2019.110028

Palomera, I., Olivar, M. P., Salat, J., Sabatés, A., Coll, M., García, A., & Morales-Nin, B. (2007). Small pelagic fish in the NW Mediterranean Sea: An ecological review. *Progress in Oceanography*, *74 (2–3)*, 377–396. https://doi.org/10.1016/j.pocean.2007.04.012

Panagos, P., Jiskra, M., Borrelli, P., Liakos, L., & Ballabio, C. (2021). Mercury in European topsoils: Anthropogenic sources, stocks and fluxes. *Environmental Research*, *201*, 111556, 1-11. https://doi.org/10.1016/j.envres.2021.111556

Papetti, P., & Rossi, G. (2009). Heavy metals in the fishery products of low Lazio and the use of metallothionein as a biomarker of contamination. *Environmental Monitoring and Assessment*, *159 (1–4)*, 589–598. https://doi.org/10.1007/s10661-008-0725-4

Park, J. D., & Zheng, W. (2012). Human exposure and health effects of inorganic and elemental mercury. *Journal of Preventive Medicine and Public Health*, *45 (6)*, 344–352. https://doi.org/10.3961/jpmph.2012.45.6.344

Pastorelli, A. A., Baldini, M., Stacchini, P., Baldini, G., Morelli, S., Sagratella, E., Zaza, S., & Ciardullo, S. (2012). Human exposure to lead, cadmium and mercury through fish and seafood product consumption in Italy: a pilot evaluation. *Food Additives and Contaminants - Part A Chemistry, Analysis, Control, Exposure and Risk Assessment*, *29 (12)*, 1913–1921. https://doi.org/10.1080/19440049.2012.719644

Perrot, V., Epov, V. N., Valentina, M. V. P., Grebenshchikova, I., Zouiten, C., Sonke, J. E., Husted, S., Donard, O. F. X., & Amouroux, D. (2010). Tracing sources and bioaccumulation of mercury in fish of Lake Baikal - Angara River using Hg isotopic composition. *Environmental Science and Technology*, *44 (21)*, 8030–8037. https://doi.org/10.1021/es101898e

Perrot, V., Pastukhov, M. v., Epov, V. N., Husted, S., Donard, O. F. X., & Amouroux, D. (2012). Higher mass-independent isotope fractionation of methylmercury in the pelagic food web of Lake Baikal (Russia). *Environmental Science and Technology*, *46 (11)*, 5902–5911. https://doi.org/10.1021/es204572g

Perrot, V., Jimenez-Moreno, M., Berail, S., Epov, V. N., Monperrus, M., & Amouroux, D. (2013). Successive methylation and demethylation of methylated mercury species (MeHg and DMeHg) induce mass dependent fractionation of mercury isotopes. *Chemical Geology*, *355*, 153–162. https://doi.org/10.1016/j.chemgeo.2013.07.011

Perrot, V., Landing, W. M., Grubbs, R. D., & Salters, V. J. M. (2019). Mercury bioaccumulation in tilefish from the northeastern Gulf of Mexico 2 years after the Deepwater Horizon oil spill: Insights from Hg, C, N and S stable isotopes. *Science*

*of the Total Environment, 666*, 828–838. https://doi.org/10.1016/j.scitotenv.2019.02.295

Pierce, G. J., Boyle, P. R., Hastie, L. C., & Santos, M. B. (1994). Diets of squid Loligo forbesi and Loligo vulgaris in the northeast Atlantic. *Fisheries Research, 21 (1–2)*, 149–163. https://doi.org/10.1016/0165-7836(94)90101-5

Pierce, G. J., Stowasser, G., Hastie, L. C., & Bustamante, P. (2008). Geographic, seasonal and ontogenetic variation in cadmium and mercury concentrations in squid (Cephalopoda: Teuthoidea) from UK waters. *Ecotoxicology and Environmental Safety, 70 (3)*, 422–432. https://doi.org/10.1016/j.ecoenv.2007.07.007

Pirrone, N., Keeler, G. J., & Nriagu, J. O. (1996). Regional differences in worldwide emissions of mercury to the atmosphere. *Atmospheric Environment, 30 (17)*, 2981-2987.

Pirrone, N., Costa, P., Pacyna, J. M., & Ferrara, R. (2001). Mercury emissions to the atmosphere from natural and anthropogenic sources in the Mediterranean region. *Atmospheric Environment*, *35*, 2997-3006.

Pirrone, N., & Mason, R. (2009). Mercury fate and transport in the global atmosphere: Emissions, measurements and models. *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements and Models*. *Springer US*, 1-637. https://doi.org/10.1007/978-0-387-93958-2

Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R. B., Friedli, H. R., Leaner, J., Mason, R., Mukherjee, A. B., Stracher, G. B., Streets, D. G., & Telmer, K. (2010). Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmospheric Chemistry and Physics, 10 (13)*, 5951–5964. https://doi.org/10.5194/acp-10-5951-2010

Plessi, M., Bertelli, D., & Monzani, A. (2001). Mercury and Selenium Content in Selected Seafood. *Journal of food composition and* analysis, *14*, 461–467. https://doi.org/10.006/jfca.2001.1003

Poissant, L., Pilote, M., Beauvais, C., Constant, P., & Zhang, H. H. (2005). A year of continuous measurements of three atmospheric mercury species (GEM, RGM and Hgp) in southern Québec, Canada. *Atmospheric Environment, 39 (7)*, 1275–1287. https://doi.org/10.1016/j.atmosenv.2004.11.007

Pyle, D. M., & Mather, T. A. (2003). The importance of volcanic emissions for the global atmospheric mercury cycle. *Atmospheric Environment, 37 (36)*, 5115–5124. https://doi.org/10.1016/j.atmosenv.2003.07.011

Pyle, G., & Couture, P. (2011). Nickel. *Fish Physiology, Homeostasis and Toxicology of Essential Metals, vol. 31 (PART A)*, 253-289. https://doi.org/10.1016/S1546-5098(11)31005-9

Rajar, R., Četina, M., Horvat, M., & Žagar, D. (2007). Mass balance of mercury in the Mediterranean Sea. *Marine Chemistry, 107 (1),* 89–102. https://doi.org/10.1016/j.marchem.2006.10.001

Ramon, R., Murcia, M., Aguinagalde, X., Amurrio, A., Llop, S., Ibarluzea, J., Lertxundi, A., Alvarez-Pedrerol, M., Casas, M., Vioque, J., Sunyer, J., Tardon, A., Martinez-Arguelles, B., & Ballester, F. (2011). Prenatal mercury exposure in a multicenter cohort study in Spain. *Environment International, 37 (3),* 597–604. https://doi.org/10.1016/j.envint.2010.12.004

Ramon, D., Morick, D., Croot, P., Berzak, R., Scheinin, A., Tchernov, D., Davidovich, N., & Britzi, M. (2021). A survey of arsenic, mercury, cadmium, and lead residues in seafood (fish, crustaceans, and cephalopods) from the south-eastern Mediterranean Sea. *Journal of Food Science, 86 (3),* 1153–1161. https://doi.org/10.1111/1750-3841.15627

Ramos, L., Fernández, M. A., González, M. J., & Hernández, L. M. (1999). Heavy Metal Pollution in Water, Sediments, and Earthworms from the Ebro River, Spain. *Bulletin of the Environmental Contamination and Toxicology, 63,* 305-311.

Rashed, M. N. (2001). Cadmium and lead levels in fish (Tilapia nilotica) tissues as biological indicator for lake water pollution. *Environmental Monitoring and Assessment, 68,* 75-89.

Rasmussen, B.L., & Thorlacius-Ussing, O. (1987). Ultrastructural localization of mercury in adrenals from rats exposed to methyl mercury. *Virchows Arch [B-Cell Pathology Including Molecular Pathology], 52,* 529-538.

Razavi, N. R., Halfman, J. D., Cushman, S. F., Massey, T., Beutner, R., Foust, J., Gilman, B., & Cleckner, L. B. (2020). Mercury concentrations in fish and invertebrates of the Finger Lakes in central New York, USA. *Ecotoxicology, 29 (10),* 1673–1685. https://doi.org/10.1007/s10646-019-02132-z

Rea, A. W., Lindberg, S. E., Scherbatskoy, T., & Keeler, G. J. (2002). Mercury accumulation in foliage over time in two northern mixed-hardwood forests. *Water, Air, and Soil Prollution, 133,* 49-67.

Reinfelder, J. R., & Janssen, S. E. (2019). Tracking legacy mercury in the Hackensack River estuary using mercury stable isotopes. *Journal of Hazardous Materials, 375,* 121–129. https://doi.org/10.1016/j.jhazmat.2019.04.074

Reuben, A., Frischtak, H., Berky, A., Ortiz, E. J., Morales, A. M., Hsu-Kim, H., Pendergast, L. L., & Pan, W. K. (2020). Elevated Hair Mercury Levels Are Associated With Neurodevelopmental Deficits in Children Living Near Artisanal and Small-Scale Gold Mining in Peru. *GeoHealth, 4 (5),* 1-14. https://doi.org/10.1029/2019GH000222

Risher, J.F., & Tucker, P. (2017). Alkyl Mercury-Induced Toxicity: Multiple Mechanisms of Action. *Reviews of environmental contamination and toxicology*, *240*, 105-1491.

Rodrigues, P. d. A., Ferrari, R. G., dos Santos, L. N., & Conte jr, C. A. (2019). Mercury in aquatic fauna contamination: A systematic reviewon its dynamics and potential health risks. *Journal of Environmental Science*, *84*, 205-218. https://doi.org/10.1016/j.jes.2019.02.018

Rodŕiguez-Gonźalez, P., Epov, V. N., Bridou, R., Tessier, E., Guyoneaud, R., Monperrus, M., & Amouroux, D. (2009). Species-specific stable isotope fractionation of mercury during Hg(II) methylation by an anaerobic bacteria (Desulfobulbus propionicus) under dark conditions. *Environmental Science and Technology, 43 (24)*, 9183–9188. https://doi.org/10.1021/es902206j

Rua-Ibarz, A., Bolea-Fernandez, E., Maage, A., Frantzen, S., Sanden, M., & Vanhaecke, F. (2019). Tracing Mercury Pollution along the Norwegian Coast via Elemental, Speciation, and Isotopic Analysis of Liver and Muscle Tissue of Deep-Water Marine Fish (Brosme brosme). *Environmental Science and Technology*, *53 (4)*, 1776–1785. https://doi.org/10.1021/acs.est.8b04706

Rudd, J. W. M., Bodaly, R. A., Fisher, N. S., Kelly, C. A., Kopec, D., & Whipple, C. (2018). Fifty years after its discharge, methylation of legacy mercury trapped in the Penobscot Estuary sustains high mercury in biota. *Science of the Total Environment*, *642*, 1340–1352. https://doi.org/10.1016/j.scitotenv.2018.06.060

Ruelas-Inzunza, J., Meza-López, G., & Páez-Osuna, F. (2008). Mercury in fish that are of dietary importance from the coasts of Sinaloa (SE Gulf of California). *Journal of Food Composition and Analysis*, *21 (3)*, 211–218. https://doi.org/10.1016/j.jfca.2007.11.004

Ruiz, J., Garcia-Isarch, E., Emma Huertas, I., Prieto, L., Juárez, A., Muñoz, J. L., Sánchez-Lamadrid, A., Rodríguez-Gálvez, S., Naranjo, J. M., & Baldó, F. (2006). Meteorological and oceanographic factors influencing Engraulis encrasicolus early life stages and catches in the Gulf of Cádiz. *Deep-Sea Research Part II: Topical Studies in Oceanography*, *53 (11–13)*, 1363–1376. https://doi.org/10.1016/j.dsr2.2006.04.007

Ruiz-Fernández, A. C., Rangel-García, M., Pérez-Bernal, L. H., López-Mendoza, P. G., Gracia, A., Schwing, P., Hollander, D., Páez-Osuna, F., Cardoso-Mohedano, J. G., Cuellar-Martinez, T., & Sanchez-Cabeza, J. A. (2019). Mercury in sediment cores from the southern Gulf of Mexico: Preindustrial levels and temporal enrichment trends. *Marine Pollution Bulletin*, *149*, 110498, 1-13. https://doi.org/10.1016/j.marpolbul.2019.110498

Rumolo, P., Bonanno, A., Barra, M., Fanelli, E., Calabrò, M., Genovese, S., Ferreri, R., Mazzola, S., & Basilone, G. (2016). Spatial variations in feeding habits and trophic

levels of two small pelagic fish species in the central Mediterranean Sea. *Marine Environmental Research*, *115*, 65–77. https://doi.org/10.1016/j.marenvres.2016.02.004

Rutkiewicz, J., Nam, D. H., Cooley, T., Neumann, K., Padilla, I. B., Route, W., Strom, S., & Basu, N. (2011). Mercury exposure and neurochemical impacts in bald eagles across several Great Lakes states. *Ecotoxicology*, *20 (7)*, 1669–1676. https://doi.org/10.1007/s10646-011-0730-1

Rutter, A. P., Schauer, J. J., Shafer, M. M., Creswell, J. E., Olson, M. R., Robinson, M., Collins, R. M., Parman, A. M., Katzman, T. L., & Mallek, J. L. (2011). Dry deposition of gaseous elemental mercury to plants and soils using mercury stable isotopes in a controlled environment. *Atmospheric Environment*, *45 (4)*, 848–855. https://doi.org/10.1016/j.atmosenv.2010.11.025

Sabo, S., Vukmirović, S., Suđi, J., Juriš, P., Tomić, Z., Bjelović, S., Tomić, L., & Sabo, A. (2021). Pesticide and toxic metal pollution in waters, fish and wild animals in Vojvodina, Serbia. *Sustainability (Switzerland)*, *13 (17)*, 9809, 1-12. https://doi.org/10.3390/su13179809

Sakai, T. (2000). Biomarkers of Lead Exposure. *Industrial Health*, *38*, 127-142.

Salazar-Camacho, C., Salas-Moreno, M., Paternina-Uribe, R., Marrugo-Negrete, J., & Díez, S. (2021). Mercury species in fish from a tropical river highly impacted by gold mining at the Colombian Pacific region. *Chemosphere*, *264 (2)*, 128478, 1-10. https://doi.org/10.1016/j.chemosphere.2020.128478

Sanders, C. W., Pacifici, K., Hess, G. R., Olfenbuttel, C., & DePerno, C. S. (2020). Metal contamination of river otters in North Carolina. *Environmental Monitoring and Assessment*, *192 (2)*, 146-163. https://doi.org/10.1007/s10661-020-8106-8

Santos, L. N., Neves, R. A. F., Koureiche, A. C., & Lailson-Brito, J. (2021). Mercury concentration in the sentinel fish species Orthopristis ruber: Effects of environmental and biological factors and human risk assessment. *Marine Pollution Bulletin*, *169*, 112508, 1-6. https://doi.org/10.1016/j.marpolbul.2021.112508

Santos-Lima, C. dos, Mourão, D. de S., Carvalho, C. F. de, Souza-Marques, B., Vega, C. M., Gonçalves, R. A., Argollo, N., Menezes-Filho, J. A., Abreu, N., & Hacon, S. de S. (2020). Neuropsychological Effects of Mercury Exposure in Children and Adolescents of the Amazon Region, Brazil. *NeuroToxicology*, *79*, 48–57. https://doi.org/10.1016/j.neuro.2020.04.004

Sato, T., Ose, Y., & Sakai, T. (1980). Toxicological effect of selenium on fish. *Environmental Pollution*, *21*, 217-224.

Schaefer, A. M., Jensen, E. L., Bossart, G. D., & Reif, J. S. (2014). Hair mercury concentrations and fish consumption patterns in Florida residents. *International*

*Journal of Environmental Research and Public Health*, *11 (7)*, 6709–6726. https://doi.org/10.3390/ijerph110706709

Schauble, E. A. (2007). Role of nuclear volume in driving equilibrium stable isotope fractionation of mercury, thallium, and other very heavy elements. *Geochimica et Cosmochimica Acta*, *71 (9)*, 2170–2189. https://doi.org/10.1016/j.gca.2007.02.004

Schaumloffel, D., Cornelis, R., Crews, H., Caruso, J., & Heumann, K.G. (2005). Speciation of nickel. *Handbook of Elemental Speciation II: Species in the Environment, Food, Medicine, and Occupational Health*, John Wiley and Sons, New York, pp. 310-326.

Schwesig, D., & Krebs, O. (2003). The role of ground vegetation in the uptake of mercury and methylmercury in a forest ecosystem. *Plant and Soil, 253*, 445-455.

Seigneur, C., Karamchandani, P., Lohman, K., Vijayaraghavan, K., & Shia, R. L. (2001). Multiscale modeling of the atmospheric fate and transport of mercury. *Journal of Geophysical Research Atmospheres*, *106 (D21)*, 27795–27809. https://doi.org/10.1029/2000JD000273

Senn, D. B., Chesney, E. J., Blum, J. D., Bank, M. S., Maage, A., & Shine, J. P. (2010). Stable isotope (N, C, Hg) study of methylmercury sources and trophic transfer in the northern Gulf of Mexico. *Environmental Science and Technology*, *44 (5)*, 1630–1637. https://doi.org/10.1021/es902361j

Sharma, B. M., Nizzetto, L., Bharat, G. K., Tayal, S., Melymuk, L., Sáňka, O., Přibylová, P., Audy, O., & Larssen, T. (2015). Melting Himalayan glaciers contaminated by legacy atmospheric depositions are important sources of PCBs and high-molecular-weight PAHs for the Ganges floodplain during dry periods the authors dedicate this paper to the memory of their dear friend and colleague, Maneesh Manjunath. *Environmental Pollution*, *206*, 588–596. https://doi.org/10.1016/j.envpol.2015.08.012

Shenker, B. J., Rooney, C., Vitale, L., & Shapiro, I. M. (1992). Immunotoxic effects of mercuric compounds on human lymphocytes and monocytes. I. Suppression of T-cell activation. *Immunopharmacology and Immunotoxicology*, *14(3)*, 539–553.

Shenker, B. J., Rooney, C., Vitale, L., De Bolt, K. & Shapiro, I. M., (1993). Immunotoxic effects of mercuric compounds on human lymphocytes and monocytes. III. Alterations in B-cell function and viability. *Immunopharmacology and Immunotoxicology*, *15(1)*, 87–112.

Sherman, L. S., Blum, J. D., Johnson, K. P., Keeler, G. J., Barres, J. A., & Douglas, T. A. (2010). Mass-independent fractionation of mercury isotopes in Arctic snow driven by sunlight. *Nature Geoscience, 3 (3)*, 173–177. https://doi.org/10.1038/ngeo758

Shokr, L. A., Hassan, M. A., & Fawzy Elbahy, E. (2019). Heavy Metals Residues (Mercury and lead) Contaminating Nile and Marine Fishes. *Benha Veterinary Medical Journal, 36 (2)*, 40-48. http://www.bvmj.bu.edu.eg

Shore, R. F., Glória, M., Lee, P., Walker, A., & Thompson, D. R. (2011). Mercury in Nonmarine Birds and Mammals. *Environmental Contaminants in Biota, Chapter 18*, 609-624. www.graphpad.com

Sin, Y. M., & Teh, W. F. (1992). Effect of Long-Term Uptake of Mercuric Sulphide on Thyroid Hormones and Glutathione in Mice. *Bulletin of the Environmental Contamination and Toxicology, 49*, 847-854.

Slemr, F., Brunke, E. G., Ebinghaus, R., Temme, C., Munthe, J., Wängberg, I., Schroeder, W., Steffen, A., & Berg, T. (2003). Worldwide trend of atmospheric mercury since 1977. *Geophysical Research Letters, 30 (10)*, 1516, 23-27. https://doi.org/10.1029/2003gl016954

Smedley, P. L., & Kinniburgh, D. G. (2002). A review of the source, behaviour and distribution of arsenic in natural waters. *Applied Geochemistry, 17*, 517-568.

Smith, E., Smith, J., Smith, L., Biswas, T., Correll, R., & Naidu, R. (2003). Arsenic in Australian environment: An overview. *Journal of Environmental Science and Health - Part A Toxic/Hazardous Substances and Environmental Engineering, 38 (1)*, 223–239. https://doi.org/10.1081/ESE-120016891

Smith, C. N., Kesler, S. E., Blum, J. D., & Rytuba, J. J. (2008). Isotope geochemistry of mercury in source rocks, mineral deposits and spring deposits of the California Coast Ranges, USA. *Earth and Planetary Science Letters, 269 (3–4)*, 399–407. https://doi.org/10.1016/j.epsl.2008.02.029

Smolders, R., den Hond, E., Koppen, G., Govarts, E., Willems, H., Casteleyn, L., Kolossa-Gehring, M., Fiddicke, U., Castaño, A., Koch, H. M., Angerer, J., Esteban, M., Sepai, O., Exley, K., Bloemen, L., Horvat, M., Knudsen, L. E., Joas, A., Joas, R., … Schoeters, G. (2015). Interpreting biomarker data from the COPHES/DEMOCOPHES twin projects: Using external exposure data to understand biomarker differences among countries. *Environmental Research, 141*, 86–95. https://doi.org/10.1016/j.envres.2014.08.016

Soto, D. X., Roig, R., Gacia, E., & Catalan, J. (2011). Differential accumulation of mercury and other trace metals in the food web components of a reservoir impacted by a chlor-alkali plant (Flix, Ebro River, Spain): Implications for biomonitoring. *Environmental Pollution, 159 (6)*, 1481–1489. https://doi.org/10.1016/j.envpol.2011.03.017

Spolaor, A., Barbaro, E., Cappelletti, D., Turetta, C., Mazzola, M., Björkman, M. P., Lucchetta, F., Dallo, F., Aspmo Pfaffhuber, K., Angot, H., Dommergue, A., Maturilli, M., Saiz-Lopez, A., Barbante, C., & Cairns, W. R. (2019). Diurnal cycle of iodine, bromine and mercury concentrations in Svalbard surface snow.

*Atmospheric Chemistry and Physics*, *19*, 13325-13339. https://doi.org/10.5194/acp-19-13325-2019

Stadnicka, A. (1980). Localization of mercury in the rat ovary after oral administration of mercuric chloride. *Acta Histochemica*, *67*, 227-233.

Storelli, M. M., Stuffler, R. G., & Marcotrigiano, G. O. (1998). Total mercury in muscle of benthic and pelagic fish from the South Adriatic Sea (Italy). *Food Additives and Contaminants*, *15 (8)*, 876–883. https://doi.org/10.1080/02652039809374724

Storelli, M. M., & Marcotrigiano, G. O. (2000). Fish for human consumption: Risk of contamination by mercury. *Food Additives and Contaminants*, *17 (12)*, 1007–1011. https://doi.org/10.1080/02652030050207792

Storelli, M. M., Giacominelli-Stuffler, R., & Marcotrigiano, G. O. (2002). Total and methylmercury residues in cartilaginous fish from Mediterranean Sea. *Marine Pollution Bulletin*, *44*, 1354-1358.

Storelli, M. M., Giacominelli-Stuffler, R., Storelli, A., D'Addabbo, R., Palermo, C., & Marcotrigiano, G. O. (2003). Survey of total mercury and methylmercury levels in edible fish from the Adriatic Sea. *Food Additives and Contaminants*, *20 (12)*, 1114–1119. https://doi.org/10.1080/02652030310001622773

Storelli, M. M., Busco, V. P., & Marcotrigiano, G. O. (2005). Mercury and arsenic speciation in the muscle tissue of Scyliorhinus canicula from the mediterranean sea. *Bulletin of Environmental Contamination and Toxicology*, *75 (1)*, 81–88. https://doi.org/10.1007/s00128-005-0721-0

Storelli, M. M. (2009). Intake of Essential Minerals and Metals via Consumption of Seafood from the Mediterranean Sea. *Journal of Food Protection*, *72 (5)*, 1116-1120. http://meridian.allenpress.com/jfp/article-pdf/72/5/1116/1677838/0362-028x-72_5_1116.pdf

Storelli, A., Barone, G., Dambrosio, A., Garofalo, R., Busco, A., & Storelli, M. M. (2020). Occurrence of trace metals in fish from South Italy: Assessment risk to consumer's health. *Journal of Food Composition and Analysis*, *90*, 103487, 1-9. https://doi.org/10.1016/j.jfca.2020.103487

Stratakis, N., Conti, D. v., Borras, E., Sabido, E., Roumeliotaki, T., Papadopoulou, E., Agier, L., Basagana, X., Bustamante, M., Casas, M., Farzan, S. F., Fossati, S., Gonzalez, J. R., Grazuleviciene, R., Heude, B., Maitre, L., McEachan, R. R. C., Theologidis, I., Urquiza, J., … Chatzi, L. (2020). Association of Fish Consumption and Mercury Exposure During Pregnancy With Metabolic Health and Inflammatory Biomarkers in Children. *JAMA Network Open*, *Nutrition, Obesity, and Exercise*, *3 (3)*, 1-16. e201007. https://doi.org/10.1001/jamanetworkopen.2020.1007

Streets, D. G., Lu, Z., Levin, L., ter Schure, A. F. H., & Sunderland, E. M. (2018). Historical releases of mercury to air, land, and water from coal combustion. *Science*

*of the Total Environment, 615*, 131–140. https://doi.org/10.1016/j.scitotenv.2017.09.207

Suárez-Serrano, A., Alcaraz, C., Ibáñez, C., Trobajo, R., & Barata, C. (2010). Procambarus clarkii as a bioindicator of heavy metal pollution sources in the lower Ebro River and Delta. *Ecotoxicology and Environmental Safety, 73 (3)*, 280–286. https://doi.org/10.1016/j.ecoenv.2009.11.001

Suda, I., & Hirayama, K. (1992). Degradation of methyl and ethyl mercury into inorganic mercury by hydroxyl radical produced from rat liver microsomes. *Archives of Toxicology*, *66*, 398-402.

Suda, I., Totokil, S., Uchida, T., & Takahashil, H. (1992). Degradation of methyl and ethyl mercury into inorganic mercury by various phagocytic cells. In *Archives of Toxicology*, *66*, 40-44.

Sun, R., Streets, D. G., Horowitz, H. M., Amos, H. M., Liu, G., Perrot, V., Toutain, J. P., Hintelmann, H., Sunderland, E. M., & Sonke, J. E. (2016). Historical (1850–2010) mercury stable isotope inventory from anthropogenic sources to the atmosphere. *Elementa: Science of the Anthropocene*, *4*, 91. DOI: https://doi.org/10.12952/journal.elementa.000091

Sun, Y., Chen, G., Yan, B., Cheng, Z., & Ma, W. (2020). Behaviour of mercury during Co-incineration of sewage sludge and municipal solid waste. *Journal of Cleaner Production*, *253*, 119969, 1-10. https://doi.org/10.1016/j.jclepro.2020.119969

Sunderland, E. M., Cohen, M. D., Selin, N. E., & Chmura, G. L. (2008). Reconciling models and measurements to assess trends in atmospheric mercury deposition. *Environmental Pollution, 156 (2)*, 526–535. https://doi.org/10.1016/j.envpol.2008.01.021

Syversen, T., & Kaur, P. (2012). The toxicology of mercury and its compounds. *Journal of Trace Elements in Medicine and Biology, 26 (4)*, 215–226. https://doi.org/10.1016/j.jtemb.2012.02.004

Tabachnick, B. G., & Fidell, L. S. (2001). Using Multivariate Statistics, Fourth Edition. *Needham Heights*, MA: Allyn & Bacon. ISBN 0-321-05677-9.

Tartu, S., Goutte, A., Bustamante, P., Angelier, F., Moe, B., Clément-Chastel, C., Bech, C., Gabrielsen, G. W., Bustnes, J. O., & Chastel, O. (2013). To breed or not to breed: Endocrine response to mercury contamination by an Arctic seabird. *Biology Letters, 9 (4)*, 20130317, 1-4. https://doi.org/10.1098/rsbl.2013.0317

Teffer, A. K., Staudinger, M. D., Taylor, D. L., & Juanes, F. (2014). Trophic influences on mercury accumulation in top pelagic predators from offshore New England waters of the northwest atlantic ocean. *Marine Environmental Research, 101 (1)*, 124–134. https://doi.org/10.1016/j.marenvres.2014.09.008

Telisman, S., Cvitkovic, P., Jurasovit, J., Pizent, A., Gavella, M., & Rocic, B. (2000). Semen Quality and Reproductive Endocrine Function in Relation to Biomarkers of Lead, Cadmium, Zinc, and Copper in Men. *Environmental Health Perspectives*, *108 (1)*, 45-53.

Terrado, M., Barceló, D., & Tauler, R. (2006). Identification and distribution of contamination sources in the Ebro river basin by chemometrics modelling coupled to geographical information systems. *Talanta*, *70 (4)*, 691–704. https://doi.org/10.1016/j.talanta.2006.05.041

Thompson, S.A., Roellich, K.L., Grossmann, A., Gilbert, S.G., & Kavanagh, T.J. (1998). Alterations in immune parameters associated with low level methylmercury exposure in mice. *Immunopharmacology and Immunotoxicology*, *20 (2)*, 299–314.

Ting, Y., & Hsi, H. C. (2019). Iron sulfide minerals as potential active capping materials for mercury-contaminated sediment remediation: A minireview. *Sustainability (Switzerland)*, *11 (6)*, 1747, 1-13. https://doi.org/10.3390/su11061747

Toole-O'Neil, B., Tewalt, S. J., Finkelman, R. B., & Akers, D. J. (1999). Mercury concentration in coal - Unraveling the puzzle. *Fuel*, *78*, 47-54.

Torres, J., Kacem, H., Eira, C., Neifar, L., & Miquel, J. (2014). Total mercury and selenium concentrations in Sarpa salpa and Balistes capriscus and in their respective Digenean endoparasites Robphildollfusium fractum and Neoapocreadium chabaudi from Tunisia. *Acta Parasitologica*, *59 (4)*, 580–585. https://doi.org/10.2478/s11686-014-0293-4

Tsui, M. T. K., Uzun, H., Ruecker, A., Majidzadeh, H., Ulus, Y., Zhang, H., Bao, S., Blum, J. D., Karanfil, T., & Chow, A. T. (2020). Concentration and isotopic composition of mercury in a blackwater river affected by extreme flooding events. *Limnology and Oceanography*, *65 (9)*, 2158–2169. https://doi.org/10.1002/lno.11445

Tunsu, C., & Wickman, B. (2018). Effective removal of mercury from aqueous streams via electrochemical alloy formation on platinum. *Nature Communications*, *9 (1)*, 4876, 1-9. https://doi.org/10.1038/s41467-018-07300-z

Turner, R. R., Kopec, A. D., Charette, M. A., & Henderson, P. B. (2018). Current and historical rates of input of mercury to the Penobscot River, Maine, from a chlor-alkali plant. *Science of the Total Environment*, *637–638*, 1175–1186. https://doi.org/10.1016/j.scitotenv.2018.05.090

Ullrich, S. M., Ilyushchenko, M. A., Kamberov, I. M., & Tanton, T. W. (2007). Mercury contamination in the vicinity of a derelict chlor-alkali plant. Part I: Sediment and water contamination of Lake Balkyldak and the River Irtysh. *Science of the Total Environment*, *381 (1–3)*, 1–16. https://doi.org/10.1016/j.scitotenv.2007.02.033

UNEP – United Nations Environment Programme. (2018). Global Mercury Assessment, https://www.unenvironment.org/resources/publication/global-mercuryassessment-

UNEP – United Nations Environment Programme. (2002). Global mercury assessment. *December, 2002*, 1-270.

Unlu, E., & Gumgum, B. (1993). Concentrations of copper and zinc in fish and sediemnts from the Tigris River in Turkey. *Chemosphere, 26 (11)*, 2055-2061.

Uren, R. C., Bothma, F., van der Lingen, C. D., & Bouwman, H. (2020). Differences in Metal Compositions and Concentrations of Sympatric Predatory Fish and Squid from the South Atlantic Ocean. *African Zoology*, *55 (4)*, 278–291. https://doi.org/10.1080/15627020.2020.1810121

van der Lingen, C. D., Hutchings, L., & Field, J. G. (2006). Comparative trophodynamics of anchovy Engraulis encrasicolus and sardine Sardinops sagax in the southern Benguela: Are species alternations between small pelagic fish trophodynamically mediated? *African Journal of Marine Science*, *28 (3–4)*, 465–477. https://doi.org/10.2989/18142320609504199

Varekamp, J. C., & Buseck, P. R. (1986). Global mercury flux from volcanic and geothermal sources. *Applied Geochemistry*, *1*, 65-73.

Varma, M., & Jain, S. (2016). Immunotoxicity of cadmium in fishes: a review. *Advanced Pharmacology and Toxicology*, *17 (2)*, 1-8.

Velma, V., Vutukuru, S. S., & Tchounwou, P. B. (2009). Ecotoxicology of Hexavalent Chromium in Freshwater Fish: A Critical Review. *Reviews of Environmental Health*, *24 (2)*, 129-145.

Veltman, J. C., & Maines, M. D. (1986). Alterations of Heme, Cytochrome P-450, and Steroid Metabolism by Mercury in Rat Adrenal. *Archives of Biochemistry and Biochemistry*, *248 (2)*, 467-478.

Vericat, D., & Batalla, R. J. (2006). Sediment transport in a large impounded river: The lower Ebro, NE Iberian Peninsula. *Geomorphology*, *79 (1–2)*, 72–92. https://doi.org/10.1016/j.geomorph.2005.09.017

Vernet, J. P., & Thomas, R. L. (1972). Levels of mercury in the sediments of some Swiss lakes including Lake Geneva and the Rhone river. *Eclogae Geologicae Helvetiae*, *65 (2)*, 293-306. DOI : 10.5169/seals-164093

Vieira, H. C., Ramirez, M. M. B., Bordalo, M. D., Rodrigues, A. C. M., Soares, A. M. V. M., Abreu, S. N., Morgado, F., & Rendón-von Osten, J. (2021). Total and Organic Mercury in Fish from Different Geographical Areas in the North Atlantic Ocean and Health Risk Assessment. *Exposure and Health*, *13 (3)*, 361–373. https://doi.org/10.1007/s12403-021-00388-7

Walker, L. A., Lawlor, A. J., Chadwick, E. A., Potter, E., Pereira, M. G., & Shore, R. F. (2011). Inorganic elements in the livers of Eurasian otters, Lutra lutra, from

England and Wales in 2009-a Predatory Bird Monitoring Scheme (PBMS) report. *Predatory Bird Monitoring Scheme, Cardiff university – Inorganic elements in the livers of the Eurasian otter in 2009*, 1-12. www.ceh.ac.uk

Walser, J. W., Kristjánsdóttir, S., Gowland, R., & Desnica, N. (2019). Volcanoes, medicine, and monasticism: Investigating mercury exposure in medieval Iceland. *International Journal of Osteoarchaeology*, *29 (1)*, 48–61. https://doi.org/10.1002/oa.2712

Wang, D., He, L., Shi, X., Wei, S., & Feng, X. (2006). Release flux of mercury from different environmental surfaces in Chongqing, China. *Chemosphere*, *64 (11)*, 1845–1854. https://doi.org/10.1016/j.chemosphere.2006.01.054

Wang, L., Hou, D., Cao, Y., Ok, Y. S., Tack, F. M. G., Rinklebe, J., & O'Connor, D. (2020). Remediation of mercury contaminated soil, water, and air: A review of emerging materials and innovative technologies. *Environment International*, *134*, 105281, 1-19. https://doi.org/10.1016/j.envint.2019.105281

Wängberg, I., Mastromonaco, G. N., Munthe, J., & Gärdfeldt, K. (2016). Airborne mercury species at the Raö background monitoring site in Sweden: Distribution of mercury as an effect of long-range transport. *Atmospheric Chemistry and Physics*, *16 (21)*, 13379–13387. https://doi.org/10.5194/acp-16-13379-2016

Wei, H., Qiu, L., Divine, K. K., Ashbaugh, M. D., Mclntyre, L. C., Fernando, Q., & Gandolfi, A. J. (1999). Toxicity And Transport Of Three Synthesized Mercury-Thiol-Complexes In Isolated Rabbit Renal Proximal Tubule Suspensions, *Drug and Chemical Toxicology*, *22:2*, 323-341, DOI: 10.3109/01480549909017838

Weiyue, F., Meng, W., Ming, G., Yuan, H., Junwen, S., Bing, W., Motao, Z., Hong, O., Yuliang, Z., & Zhifang, C. (2011). Mercury speciation and mercury-binding protein study by HPLC-ICP-MS on the estimation of mercury toxicity between maternal and infant rats. *Journal of Analytical Atomic Spectrometry*, *26 (1)*, 156–164. https://doi.org/10.1039/c0ja00111b

WHO – World Health Organization. (1990). Methylmercury. *Environmental Health Criteria*, *101*, 1-148.

Wiatrowski, H. A., & Barkay, T. (2005). Monitoring of microbial metal transformations in the environment. *Current Opinion in Biotechnology*, *16 (3)*, 261–268. https://doi.org/10.1016/j.copbio.2005.04.011

Wiedinmyer, C., & Friedli, H. (2007). Mercury emission estimates from fires: An initial inventory for the United States. *Environmental Science and Technology*, *41 (23)*, 8092–8098. https://doi.org/10.1021/es071289o

Wiener, J. G., Knights, B. C., Sandheinrich, M. B., Jeremiason, J. D., Brigham, M. E., Engstrom, D. R., Woodruff, L. G., Cannon, W. F., & Balogh, S. J. (2006). Mercury in soils, lakes, and fish in Voyageurs National Park (Minnesota): Importance of

atmospheric deposition and ecosystem factors. *Environmental Science and Technology*, *40 (20)*, 6261–6268. https://doi.org/10.1021/es060822h

Wilhelm, S. M. (2001). Estimate of Mercury Emissions to the Atmosphere from Petroleum. *Environmental Science and Technology*, *35 (24)*, 4704-4710. DOI: 10.1021/es001804h

Witt, M. L. I., Mather, T. A., Pyle, D. M., Aiuppa, A., Bagnato, E., & Tsanev, V. I. (2008). Mercury and halogen emissions from Masaya and telica volcanoes, Nicaragua. *Journal of Geophysical Research: Solid Earth*, *113 (6)*, B06203, 1-15. https://doi.org/10.1029/2007JB005401

Wolfe, M. F., Schwarzbach, S., & Sulaiman, R. A. (1998). Effects of mercury on wildlife: A comprehensive review. *Environmental Toxicology and Chemistry*, *17 (2)*, 146–160. https://doi.org/10.1002/etc.5620170203

Wu, Q., Gao, W., Wang, S., & Hao, J. (2017). Updated atmospheric speciated mercury emissions from iron and steel production in China during 2000-2015. *Atmospheric Chemistry and Physics*, *17 (17)*, 10423–10433. https://doi.org/10.5194/acp-17-10423-2017

Xu, X., Zhang, Q., & Wang, W. X. (2016). Linking mercury, carbon, and nitrogen stable isotopes in Tibetan biota: Implications for using mercury stable isotopes as source tracers. *Scientific Reports*, *6*, 25394, 1-10. https://doi.org/10.1038/srep25394

Yabanli, M. (2013). Assessment of the heavy metal contents of Sardina pilchardus Sold in Izmir, Turkey. *Ekoloji*, *87*, 10–15. https://doi.org/10.5053/ekoloji.2013.872

Yang, Y., Liu, J., & Wang, Z. (2020). Reaction mechanisms and chemical kinetics of mercury transformation during coal combustion. *Progress in Energy and Combustion Science*, *79*, 100844, 1-41. https://doi.org/10.1016/j.pecs.2020.100844

Yilmaz, A. B. (2003). Levels of heavy metals (Fe, Cu, Ni, Cr, Pb, and Zn) in tissue of Mugil cephalus and Trachurus mediterraneus from Iskenderun Bay, Turkey. *Environmental Research*, *92 (3)*, 277–281. https://doi.org/10.1016/S0013-9351(02)00082-8

Yin, Z., Milatovic, D., Aschner, J. L., Syversen, T., Rocha, J. B. T., Souza, D. O., Sidoryk, M., Albrecht, J., & Aschner, M. (2007). Methylmercury induces oxidative injury, alterations in permeability and glutamine transport in cultured astrocytes. *Brain Research*, *1131 (1)*, 1–10. https://doi.org/10.1016/j.brainres.2006.10.070

Yin, R., Feng, X., & Shi, W. (2010). Application of the stable-isotope system to the study of sources and fate of Hg in the environment: A review. *Applied Geochemistry*, *25 (10)*, 1467–1477. https://doi.org/10.1016/j.apgeochem.2010.07.007

Yin, R., Feng, X., Li, X., Yu, B., & Du, B. (2014). Trends and advances in mercury stable isotopes as a geochemical tracer. *Trends in Environmental Analytical Chemistry*, *2*, 1–10. https://doi.org/10.1016/j.teac.2014.03.001

Ynalvez, R., Gutierrez, J.; & Gonzalez-Cantu, H. (2016). Mini-review: toxicity of mercury as a consequence of enzyme alteration. *Biometals*, *29(5)*, 781-788. DOI:10.1007/s10534-016-9967-8

Young, R.A. (1992). Toxicity summary for methylmercury. Oak Ridge Resevation Environmental Restoration Program (RAIS: Methyl Mercury (2269-92-6)). http://risk.lsd.ornl.gov/tox/profiles/methyl_mercury_f_V1.shtml

Yu, R. Q., Reinfelder, J. R., Hines, M. E., & Barkay, T. (2013). Mercury methylation by the methanogen Methanospirillum hungatei. *Applied and Environmental Microbiology*, *79 (20)*, 6325–6330. https://doi.org/10.1128/AEM.01556-13

Yu, X., Khan, S., Khan, A., Tang, Y., Nunes, L. M., Yan, J., Ye, X., & Li, G. (2020). Methyl mercury concentrations in seafood collected from Zhoushan Islands, Zhejiang, China, and their potential health risk for the fishing community: Capsule: Methyl mercury in seafood causes potential health risk. *Environment International*, *137*, 105420, 1-10. https://doi.org/10.1016/j.envint.2019.105420

Zhu, X., Kusaka, Y., Sato, K., & Zhang, Q. (2000). The endocrine disruptive effects of mercury. *Environmental Health and Preventive Medicine*, *4 (4)*, 174–183. https://doi.org/10.1007/BF02931255

Zhu, W., Li, Z., Li, P., Yu, B., Lin, C. J., Sommar, J., & Feng, X. (2018). Re-emission of legacy mercury from soil adjacent to closed point sources of Hg emission. *Environmental Pollution*, *242*, 718–727. https://doi.org/10.1016/j.envpol.2018.07.002

Zitko, V. (1975). Toxicity and pollution potential of thallium. *The Science of the Total Environment*, *4*, 185-192.

Zolnikov, T. R., & Ramirez Ortiz, D. (2018). A systematic review on the management and treatment of mercury in artisanal gold mining. *Science of the Total Environment*, *633*, 816–824. https://doi.org/10.1016/j.scitotenv.2018.03.241

# APPENDIX A

FISH SPECIES EQUIVALENCES

Table A1. Equivalence of the fish species between English, Catalan and Italian.

| English | Scientific | Catalan | Italian |
|---|---|---|---|
| Anchovie | *Engraulis encrasicolus* | Aladroc, Seitó | Acciuga |
| Angler, Monk fish | *Lophius piscatorius* | Rap | Rana pescatrice |
| Atlantic horse mackerel | *Trachurus trachurus* | Sorell | Suro, sugarello |
| Axillary seabream | *Pagellus acarne* | Besuc blanc, calet | Pagello bastardo |
| Black scorpionfish | *Scorpaena porcus* | Escórpora fosca o de fang | Scorfano nero |
| Black seabream | *Spondyliosoma cantharus* | Càntera | Cantaro |
| Blackspot seabream | *Pagellus bogaraveo* | Goràs, besuc de fonera | Pezzogna |
| Blue whiting | *Micromesistius poutassou* | Llúcera, maire, mare de lluç | Melù, potassolo |
| Brown meagre | *Sciaena umbra* | Corball de roca | Corvina |
| Comber | *Serranus cabrilla* | Serrà | Perchia |
| Common dentex | *Dentex dentex* | Déntol | Dentice |
| Common dolphinfish | *Coryphaena hippurus* | Llampuga | Lampuga |
| Common pandora | *Pagellus erythrinus* | Pagell | Fragolino |
| Common seabream | *Pagrus pagrus* | Pagre | Pagro |
| Common sole | *Solea solea* | Palaia, llenguado | Sogliola |
| Conger | *Conger conger* | Congre | Congro |
| Cuckoo wrasse | *Labrus bimaculatus* | Tord | Tordo fischietto |
| Cuttlefish | *Sepia officinalis* | Sípia | Seppia |
| Dusky grouper | *Epinephelus marginatus* | Anfós, nero | Cernia bruna |
| European barracuda | *Sphyraena sphyraena* | Espet | Barracuda |
| European eel | *Anguilla anguilla* | Anguila | Anguilla |
| European hake | *Merluccius merluccius* | Lluç europeu | Merluzzo |
| Flathead mullet | *Mugil cephalus* | Llissa llobarrera, llissa | Cefalo |
| Four-spot megrim | *Lepidorhombus boscii* | Palaia bruixa | Rombo quattrocchi |
| Gilthead seabream | *Sparus aurata* | Orada | Orata |
| Greater amberjack | *Seriola dumerili* | Círvia, letxa | Ricciola |
| Greater forkbeard | *Phycis blennoides* | Mòllera de fang | Musdea, mostella |
| Greater weever | *Trachinus draco* | Aranya blanca | Tracina |
| Gurnard | *Chelidonichthys lucerna* | Lluerna rossa | Gallinella |
| John dory | *Zeus faber* | Gall, gall de Sant Pere | Pesce di San Pietro |
| Mackerel | *Scomber scombrus* | Verat, cavalla | Sgombro |
| Mediterranean moray | *Muraena Helena* | Morena | Murena |
| Megrim sole | *Lepidorhombus whiffiagonis* | Bruixa sense taques, gall | Rombo giallo |

| English | Scientific | Catalan | Italian |
|---|---|---|---|
| Mussel | *Mytilus galloprovincialis* | Musclo | Cozza |
| Norway lobster | *Nephrops norvegicus* | Escamarlà, cigala | Scampo |
| Octopus | *Octopus vulgaris* | Pop | Polpo |
| Offshore rockfish | *Pontinus kuhlii* | Pontí | Scorfano corallino |
| Painted comber | *Serranus scriba* | Vaca, serrà | Boccaccia,sciarrano |
| Pearly razorfish | *Xyrichtys novacula* | Raor | Pesce pettine |
| Picarel | *Spicara smaris* | Gerret | Zerro |
| Poor cod | *Trisopterus minutus* | Capellà, mòllera | Cappellano |
| Porbeagle | *Lamna nasus* | Marraix, llúdria | Smeriglio |
| Red bandfish | *Cepola macrophthalma* | Codornera, veta | Cepola |
| Red mullet | *Mullus barbatus* | Moll de fang | Triglia di fango |
| Red scorpionfish | *Scorpaena scrofa* | Cap-roig, escórpora | Scorfano rosso |
| Round sardinella | *Sardinella aurita* | Alatxa | Alaccia |
| Salema | *Sarpa salpa* | Salpa | Salpa |
| Sand steenbras | *Lithognathus mormyrus* | Mabre | Marmora |
| Sardine | *Sardina pilchardus* | Sardina | Sardina |
| Shrimp | *Aristeus antennatus* | Gamba rosada | Gambero |
| Small-spotted catshark | *Scyliorhinus canicula* | Gat, gat ver | Gattuccio |
| Squid | *Loligo vulgaris* | Calamar comú | Calamaro |
| Surmullet | *Mullus surmuletus* | Moll de roca | Triglia di scoglio |
| Thornback ray | *Raja clavata* | Clavellada, rajada | Razza |
| Transparent goby | *Aphia minuta* | Xanguet, cabeçuda | Rossetto |
| White sardinella | *Sardinella albella* | Sardinella albella | Alaccia asiatica |
| White seabream | *Diplodus sargus sargus* | Sarg, sard | Sarago |
| Whiting | *Merlangius merlangus* | Merlà | Merlano, molo |

# APPENDIX B

FISH SPECIES INFORMATION AND TOTAL MERCURY CONCENTRATIONS

Table B1. Fish species, information (weight and length), mercury concentration (mg/kg ww), purchase site and trophic level.

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1 | SQUID | *Loligo vulgaris* | - | - | 0.36 | Balearic Islands | 4.2 |
| 2 | SQUID | *Loligo vulgaris* | - | - | 0.30 | Balearic Islands | 4.2 |
| 3 | SQUID | *Loligo vulgaris* | - | - | 0.30 | Balearic Islands | 4.2 |
| 4 | SQUID | *Loligo vulgaris* | - | - | 0.36 | Balearic Islands | 4.2 |
| 5 | SQUID | *Loligo vulgaris* | 309 | 55 | 0.03 | Alacant | 4.2 |
| 6 | SQUID | *Loligo vulgaris* | 235 | 53 | 0.03 | Alacant | 4.2 |
| 7 | SQUID | *Loligo vulgaris* | 270 | 57 | 0.02 | Alacant | 4.2 |
| 8 | SQUID | *Loligo vulgaris* | 282 | 57 | 0.03 | Alacant | 4.2 |
| 9 | SQUID | *Loligo vulgaris* | 145 | 37 | 0.14 | Alacant | 4.2 |
| 10 | SQUID | *Loligo vulgaris* | 185 | 43 | 0.13 | Alacant | 4.2 |
| 11 | SQUID | *Loligo vulgaris* | 157 | 37 | 0.17 | Alacant | 4.2 |
| 12 | SQUID | *Loligo vulgaris* | 175 | 36 | 0.21 | Alacant | 4.2 |
| 13 | SQUID | *Loligo vulgaris* | 91 | 37 | 0.11 | Marseille | 4.2 |
| 14 | SQUID | *Loligo vulgaris* | 74 | 41 | 0.19 | Marseille | 4.2 |
| 15 | TRANSPARENT GOBY | *Aphia minuta* | 60 | - | 0.09 | Balearic Islands | 3.1 ±0.28 |
| 16 | TRANSPARENT GOBY | *Aphia minuta* | 60 | - | 0.09 | Balearic Islands | 3.1 ±0.28 |
| 17 | TRANSPARENT GOBY | *Aphia minuta* | 60 | - | 0.09 | Balearic Islands | 3.1 ±0.28 |
| 18 | TRANSPARENT GOBY | *Aphia minuta* | 150 | - | 0.09 | Balearic Islands | 3.1 ±0.28 |
| 19 | SHRIMP | *Aristeus antennatus* | - | - | 2.35 | Balearic Islands | - |
| 20 | SHRIMP | *Aristeus antennatus* | - | - | 1.85 | Balearic Islands | - |
| 21 | SHRIMP | *Aristeus antennatus* | - | - | 1.85 | Balearic Islands | - |
| 22 | SHRIMP | *Aristeus antennatus* | - | - | 2.35 | Balearic Islands | - |
| 23 | SHRIMP | *Aristeus antennatus* | 12 | 12 | 0.66 | Alacant | - |
| 24 | SHRIMP | *Aristeus antennatus* | 13 | 12 | 0.19 | Alacant | - |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 25 | SHRIMP | *Aristeus antennatus* | 13 | 11 | 0.19 | Alacant | - |
| 26 | SHRIMP | *Aristeus antennatus* | 14 | 12 | 0.24 | Alacant | - |
| 27 | SHRIMP | *Aristeus antennatus* | 11 | 11 | 0.14 | Alacant | - |
| 28 | SHRIMP | *Aristeus antennatus* | 12 | 11 | 0.19 | Alacant | - |
| 29 | SHRIMP | *Aristeus antennatus* | 10 | 7 | 0.16 | Alacant | - |
| 30 | SHRIMP | *Aristeus antennatus* | 12 | 12 | 0.11 | Alacant | - |
| 31 | SHRIMP | *Aristeus antennatus* | 14 | 14 | 0.17 | Alacant | - |
| 32 | SHRIMP | *Aristeus antennatus* | 11 | 13 | 0.27 | L'Ampolla - Ebro Delta | - |
| 33 | SHRIMP | *Aristeus antennatus* | 12 | 13 | 0.38 | L'Ampolla - Ebro Delta | - |
| 34 | SHRIMP | *Aristeus antennatus* | 10 | 14 | 0.77 | L'Ampolla - Ebro Delta | - |
| 35 | SHRIMP | *Aristeus antennatus* | 9 | 10 | 0.22 | L'Ampolla - Ebro Delta | - |
| 36 | SHRIMP | *Aristeus antennatus* | 10 | 13 | 0.23 | L'Ampolla - Ebro Delta | - |
| 37 | SHRIMP | *Aristeus antennatus* | 10 | 13 | 0.21 | L'Ampolla - Ebro Delta | - |
| 38 | SHRIMP | *Aristeus antennatus* | 10 | 12 | 0.30 | L'Ampolla - Ebro Delta | - |
| 39 | SHRIMP | *Aristeus antennatus* | 10 | 12 | 0.19 | L'Ampolla - Ebro Delta | - |
| 40 | CONGER | *Conger conger* | 520 | - | 0.31 | Balearic Islands | 4.3 ± 0.4 |
| 41 | CONGER | *Conger conger* | 350 | 64 | 0.35 | Balearic Islands | 4.3 ± 0.4 |
| 42 | CONGER | *Conger conger* | 900 | - | 0.68 | Balearic Islands | 4.3 ± 0.4 |
| 43 | CONGER | *Conger conger* | 300 | 54 | 0.69 | Balearic Islands | 4.3 ± 0.4 |
| 44 | CONGER | *Conger conger* | 250 | 58 | 0.70 | Balearic Islands | 4.3 ± 0.4 |
| 45 | CONGER | *Conger conger* | 500 | 92 | 0.50 | Balearic Islands | 4.3 ± 0.4 |
| 46 | CONGER | *Conger conger* | 2500 | - | 0.21 | Balearic Islands | 4.3 ± 0.4 |
| 47 | CONGER | *Conger conger* | 840 | 80 | 0.65 | Balearic Islands | 4.3 ± 0.4 |
| 48 | CONGER | *Conger conger* | 2000 | - | 1.30 | Balearic Islands | 4.3 ± 0.4 |
| 49 | CONGER | *Conger conger* | 340 | - | 0.45 | Balearic Islands | 4.3 ± 0.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 50 | CONGER | *Conger conger* | 860 | 100 | 0.56 | Balearic Islands | 4.3 ± 0.4 |
| 51 | CONGER | *Conger conger* | 400 | 38 | 0.52 | Balearic Islands | 4.3 ± 0.4 |
| 52 | CONGER | *Conger conger* | 2160 | 100 | 0.38 | Balearic Islands | 4.3 ± 0.4 |
| 53 | CONGER | *Conger conger* | 1600 | 95 | 0.40 | Balearic Islands | 4.3 ± 0.4 |
| 54 | CONGER | *Conger conger* | 295 | 65 | 0.18 | Balearic Islands | 4.3 ± 0.4 |
| 55 | CONGER | *Conger conger* | - | 100 | 0.31 | Balearic Islands | 4.3 ± 0.4 |
| 56 | CONGER | *Conger conger* | 270 | 38 | 0.40 | Balearic Islands | 4.3 ± 0.4 |
| 57 | CONGER | *Conger conger* | 1560 | - | 0.38 | Balearic Islands | 4.3 ± 0.4 |
| 58 | CONGER | *Conger conger* | 1080 | 70 | 0.18 | Balearic Islands | 4.3 ± 0.4 |
| 59 | CONGER | *Conger conger* | 2020 | 100 | 0.25 | Balearic Islands | 4.3 ± 0.4 |
| 60 | CONGER | *Conger conger* | 350 | 64 | 0.36 | Balearic Islands | 4.3 ± 0.4 |
| 61 | CONGER | *Conger conger* | 2000 | - | 0.34 | Balearic Islands | 4.3 ± 0.4 |
| 62 | CONGER | *Conger conger* | - | - | 0.34 | Balearic Islands | 4.3 ± 0.4 |
| 63 | CONGER | *Conger conger* | 1000 | 120 | 0.17 | Balearic Islands | 4.3 ± 0.4 |
| 64 | CONGER | *Conger conger* | 760 | 80 | 0.28 | Balearic Islands | 4.3 ± 0.4 |
| 65 | CONGER | *Conger conger* | - | - | 0.37 | Balearic Islands | 4.3 ± 0.4 |
| 66 | CONGER | *Conger conger* | 1700 | - | 0.64 | Balearic Islands | 4.3 ± 0.4 |
| 67 | CONGER | *Conger conger* | 280 | - | 0.57 | Balearic Islands | 4.3 ± 0.4 |
| 68 | CONGER | *Conger conger* | 280 | 58 | 0.65 | Balearic Islands | 4.3 ± 0.4 |
| 69 | CONGER | *Conger conger* | - | - | 1.84 | Balearic Islands | 4.3 ± 0.4 |
| 70 | CONGER | *Conger conger* | - | - | 1.00 | Balearic Islands | 4.3 ± 0.4 |
| 71 | CONGER | *Conger conger* | - | - | 0.26 | Balearic Islands | 4.3 ± 0.4 |
| 72 | CONGER | *Conger conger* | - | - | 1.70 | Balearic Islands | 4.3 ± 0.4 |
| 73 | CONGER | *Conger conger* | - | - | 0.46 | Balearic Islands | 4.3 ± 0.4 |
| 74 | CONGER | *Conger conger* | - | 80 | 0.27 | Balearic Islands | 4.3 ± 0.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 75 | CONGER | *Conger conger* | 622 | 72 | 0.36 | Marseille | 4.3 ± 0.4 |
| 76 | CONGER | *Conger conger* | 576 | 70 | 0.29 | Marseille | 4.3 ± 0.4 |
| 77 | CONGER | *Conger conger* | 654 | 72 | 0.25 | Marseille | 4.3 ± 0.4 |
| 78 | CONGER | *Conger conger* | 576 | 70 | 0.36 | Marseille | 4.3 ± 0.4 |
| 79 | CONGER | *Conger conger* | 236 | 43 | 0.89 | Marseille | 4.3 ± 0.4 |
| 80 | CONGER | *Conger conger* | 451 | 57 | 0.48 | Marseille | 4.3 ± 0.4 |
| 81 | CONGER | *Conger conger* | 813 | 72 | 0.57 | Marseille | 4.3 ± 0.4 |
| 82 | CONGER | *Conger conger* | 794 | 69 | 0.41 | Marseille | 4.3 ± 0.4 |
| 83 | CONGER | *Conger conger* | 842 | 73 | 0.26 | Marseille | 4.3 ± 0.4 |
| 84 | CONGER | *Conger conger* | 1008 | 83 | 0.33 | Marseille | 4.3 ± 0.4 |
| 85 | COMMON DOLPHINFISH | *Coryphaena hippurus* | 1600 | 64 | 0.09 | Balearic Islands | 4.4 |
| 86 | COMMON DOLPHINFISH | *Coryphaena hippurus* | 1300 | 57 | 0.09 | Balearic Islands | 4.4 |
| 87 | COMMON DOLPHINFISH | *Coryphaena hippurus* | - | - | 0.05 | Balearic Islands | 4.4 |
| 88 | COMMON DOLPHINFISH | *Coryphaena hippurus* | - | - | 0.05 | Balearic Islands | 4.4 |
| 89 | COMMON DOLPHINFISH | *Coryphaena hippurus* | 280 | 50 | 0.09 | Balearic Islands | 4.4 |
| 90 | COMMON DENTEX | *Dentex dentex* | - | - | 0.15 | Balearic Islands | 4.5 ±0.4 |
| 91 | COMMON DENTEX | *Dentex dentex* | 1550 | 49 | 1.20 | Balearic Islands | 4.5 ±0.4 |
| 92 | COMMON DENTEX | *Dentex dentex* | 2750 | 59 | 1.10 | Balearic Islands | 4.5 ±0.4 |
| 93 | COMMON DENTEX | *Dentex dentex* | 2900 | 61 | 0.90 | Balearic Islands | 4.5 ±0.4 |
| 94 | COMMON DENTEX | *Dentex dentex* | - | - | 0.48 | Balearic Islands | 4.5 ±0.4 |
| 95 | COMMON DENTEX | *Dentex dentex* | - | - | 1.10 | Balearic Islands | 4.5 ±0.4 |
| 96 | COMMON DENTEX | *Dentex dentex* | - | - | 1.40 | Balearic Islands | 4.5 ±0.4 |
| 97 | COMMON DENTEX | *Dentex dentex* | 760 | 40 | 0.38 | Balearic Islands | 4.5 ±0.4 |
| 98 | COMMON DENTEX | *Dentex dentex* | 350 | 30 | 0.15 | Balearic Islands | 4.5 ±0.4 |
| 99 | COMMON DENTEX | *Dentex dentex* | - | - | 1.10 | Balearic Islands | 4.5 ±0.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 100 | COMMON DENTEX | *Dentex dentex* | - | - | 0.48 | Balearic Islands | 4.5 ±0.4 |
| 101 | COMMON DENTEX | *Dentex dentex* | 1400 | - | 1.40 | Balearic Islands | 4.5 ±0.4 |
| 102 | COMMON DENTEX | *Dentex dentex* | - | - | 0.38 | Balearic Islands | 4.5 ±0.4 |
| 103 | COMMON DENTEX | *Dentex dentex* | 1250 | 45 | 0.31 | Balearic Islands | 4.5 ±0.4 |
| 104 | COMMON DENTEX | *Dentex dentex* | 3590 | 60 | 1.50 | Balearic Islands | 4.5 ±0.4 |
| 105 | COMMON DENTEX | *Dentex dentex* | 1950 | 64 | 0.64 | Balearic Islands | 4.5 ±0.4 |
| 106 | COMMON DENTEX | *Dentex dentex* | 3175 | 51 | 0.42 | Balearic Islands | 4.5 ±0.4 |
| 107 | COMMON DENTEX | *Dentex dentex* | - | - | 1.40 | Balearic Islands | 4.5 ±0.4 |
| 108 | COMMON DENTEX | *Dentex dentex* | - | - | 2.03 | Balearic Islands | 4.5 ±0.4 |
| 109 | COMMON DENTEX | *Dentex dentex* | 7000 | - | 0.78 | Balearic Islands | 4.5 ±0.4 |
| 110 | COMMON DENTEX | *Dentex dentex* | 7000 | - | 1.30 | Balearic Islands | 4.5 ±0.4 |
| 111 | COMMON DENTEX | *Dentex dentex* | 7000 | - | 1.11 | Balearic Islands | 4.5 ±0.4 |
| 112 | COMMON DENTEX | *Dentex dentex* | - | - | 2.03 | Balearic Islands | 4.5 ±0.4 |
| 113 | COMMON DENTEX | *Dentex dentex* | - | - | 0.57 | Balearic Islands | 4.5 ±0.4 |
| 114 | COMMON DENTEX | *Dentex dentex* | 280 | 50 | 0.20 | Balearic Islands | 4.5 ±0.4 |
| 115 | COMMON DENTEX | *Dentex dentex* | - | - | 1.20 | Balearic Islands | 4.5 ±0.4 |
| 116 | COMMON DENTEX | *Dentex dentex* | - | - | 1.50 | Balearic Islands | 4.5 ±0.4 |
| 117 | COMMON DENTEX | *Dentex dentex* | - | - | 0.57 | Balearic Islands | 4.5 ±0.4 |
| 118 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.23 | Balearic Islands | 3.4 ± 0.1 |
| 119 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.31 | Balearic Islands | 3.4 ± 0.1 |
| 120 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.31 | Balearic Islands | 3.4 ± 0.1 |
| 121 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.32 | Balearic Islands | 3.4 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 122 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.48 | Balearic Islands | 3.4 ± 0.1 |
| 123 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.63 | Balearic Islands | 3.4 ± 0.1 |
| 124 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.27 | Balearic Islands | 3.4 ± 0.1 |
| 125 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.36 | Balearic Islands | 3.4 ± 0.1 |
| 126 | WHITE SEABREAM | *Diplodus sargus sargus* | - | - | 0.16 | Balearic Islands | 3.4 ± 0.1 |
| 127 | WHITE SEABREAM | *Diplodus sargus sargus* | 158 | 21 | 0.29 | Balearic Islands | 3.4 ± 0.1 |
| 128 | WHITE SEABREAM | *Diplodus sargus sargus* | 111 | 18 | 0.30 | Balearic Islands | 3.4 ± 0.1 |
| 129 | WHITE SEABREAM | *Diplodus sargus sargus* | 147 | 21 | 0.19 | Balearic Islands | 3.4 ± 0.1 |
| 130 | WHITE SEABREAM | *Diplodus sargus sargus* | 175 | 22 | 0.49 | Balearic Islands | 3.4 ± 0.1 |
| 131 | WHITE SEABREAM | *Diplodus sargus sargus* | 177 | 23 | 0.49 | Balearic Islands | 3.4 ± 0.1 |
| 132 | WHITE SEABREAM | *Diplodus sargus sargus* | 135 | 19 | 0.30 | Balearic Islands | 3.4 ± 0.1 |
| 133 | WHITE SEABREAM | *Diplodus sargus sargus* | 162 | 21 | 0.18 | Balearic Islands | 3.4 ± 0.1 |
| 134 | WHITE SEABREAM | *Diplodus sargus sargus* | 156 | 22 | 0.29 | Balearic Islands | 3.4 ± 0.1 |
| 135 | WHITE SEABREAM | *Diplodus sargus sargus* | 186 | 22 | 0.14 | Balearic Islands | 3.4 ± 0.1 |
| 136 | WHITE SEABREAM | *Diplodus sargus sargus* | 169 | 22 | 0.09 | Balearic Islands | 3.4 ± 0.1 |
| 137 | WHITE SEABREAM | *Diplodus sargus sargus* | 154 | 20.5 | 0.66 | Balearic Islands | 3.4 ± 0.1 |
| 138 | WHITE SEABREAM | *Diplodus sargus sargus* | 137 | 19 | 0.14 | Balearic Islands | 3.4 ± 0.1 |
| 139 | WHITE SEABREAM | *Diplodus sargus sargus* | 113 | 19 | 0.21 | Balearic Islands | 3.4 ± 0.1 |
| 140 | WHITE SEABREAM | *Diplodus sargus sargus* | 160 | 22 | 0.10 | Alghero | 3.4 ± 0.1 |
| 141 | WHITE SEABREAM | *Diplodus sargus sargus* | 112 | 19 | 0.28 | Alghero | 3.4 ± 0.1 |
| 142 | WHITE SEABREAM | *Diplodus sargus sargus* | 113 | 19 | 0.49 | Alghero | 3.4 ± 0.1 |
| 143 | WHITE SEABREAM | *Diplodus sargus sargus* | 122 | 19 | 0.48 | Alghero | 3.4 ± 0.1 |
| 144 | WHITE SEABREAM | *Diplodus sargus sargus* | 110 | 18 | 0.43 | Alghero | 3.4 ± 0.1 |
| 145 | WHITE SEABREAM | *Diplodus sargus sargus* | 155 | 21 | 0.70 | Alghero | 3.4 ± 0.1 |
| 146 | WHITE SEABREAM | *Diplodus sargus sargus* | 112 | 19 | 0.35 | Alghero | 3.4 ± 0.1 |
| 147 | WHITE SEABREAM | *Diplodus sargus sargus* | 121 | 19 | 0.43 | Alghero | 3.4 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 148 | WHITE SEABREAM | *Diplodus sargus sargus* | 151 | 21 | 0.16 | Alghero | 3.4 ± 0.1 |
| 149 | WHITE SEABREAM | *Diplodus sargus sargus* | 134 | 19 | 0.08 | Alghero | 3.4 ± 0.1 |
| 150 | WHITE SEABREAM | *Diplodus sargus sargus* | 114 | 20 | 0.28 | Alghero | 3.4 ± 0.1 |
| 151 | WHITE SEABREAM | *Diplodus sargus sargus* | 142 | 20 | 0.37 | Alghero | 3.4 ± 0.1 |
| 152 | WHITE SEABREAM | *Diplodus sargus sargus* | 185 | 21 | 0.48 | Alghero | 3.4 ± 0.1 |
| 153 | WHITE SEABREAM | *Diplodus sargus sargus* | 172 | 22 | 0.11 | Alacant | 3.4 ± 0.1 |
| 154 | WHITE SEABREAM | *Diplodus sargus sargus* | 78 | 18 | 0.09 | Alacant | 3.4 ± 0.1 |
| 155 | WHITE SEABREAM | *Diplodus sargus sargus* | 686 | 38 | 0.07 | Alacant | 3.4 ± 0.1 |
| 156 | WHITE SEABREAM | *Diplodus sargus sargus* | 54 | 18 | 0.12 | Alacant | 3.4 ± 0.1 |
| 157 | WHITE SEABREAM | *Diplodus sargus sargus* | 30 | 13 | 0.31 | Civitavecchia | 3.4 ± 0.1 |
| 158 | WHITE SEABREAM | *Diplodus sargus sargus* | 106 | 22 | 1.07 | Civitavecchia | 3.4 ± 0.1 |
| 159 | WHITE SEABREAM | *Diplodus sargus sargus* | 182 | 28 | 0.95 | Civitavecchia | 3.4 ± 0.1 |
| 160 | WHITE SEABREAM | *Diplodus sargus sargus* | 124 | 20 | 0.19 | Civitavecchia | 3.4 ± 0.1 |
| 161 | WHITE SEABREAM | *Diplodus sargus sargus* | 64 | 16 | 0.19 | Civitavecchia | 3.4 ± 0.1 |
| 162 | WHITE SEABREAM | *Diplodus sargus sargus* | 62 | 17 | 0.12 | Civitavecchia | 3.4 ± 0.1 |
| 163 | WHITE SEABREAM | *Diplodus sargus sargus* | 129 | 19 | 0.32 | Civitavecchia | 3.4 ± 0.1 |
| 164 | WHITE SEABREAM | *Diplodus sargus sargus* | 223 | 24 | 0.77 | Civitavecchia | 3.4 ± 0.1 |
| 165 | WHITE SEABREAM | *Diplodus sargus sargus* | 223 | 23 | 0.25 | Civitavecchia | 3.4 ± 0.1 |
| 166 | WHITE SEABREAM | *Diplodus sargus sargus* | 210 | 23 | 0.43 | Civitavecchia | 3.4 ± 0.1 |
| 167 | WHITE SEABREAM | *Diplodus sargus sargus* | 198 | 24 | 0.41 | Civitavecchia | 3.4 ± 0.1 |
| 168 | WHITE SEABREAM | *Diplodus sargus sargus* | 110 | 19 | 0.54 | Genoa | 3.4 ± 0.1 |
| 169 | WHITE SEABREAM | *Diplodus sargus sargus* | 167 | 24 | 0.13 | Genoa | 3.4 ± 0.1 |
| 170 | WHITE SEABREAM | *Diplodus sargus sargus* | 214 | 25 | 0.15 | Genoa | 3.4 ± 0.1 |
| 171 | WHITE SEABREAM | *Diplodus sargus sargus* | 181 | 21 | 0.14 | Genoa | 3.4 ± 0.1 |
| 172 | WHITE SEABREAM | *Diplodus sargus sargus* | 175 | 24 | 0.08 | Genoa | 3.4 ± 0.1 |
| 173 | WHITE SEABREAM | *Diplodus sargus sargus* | 196 | 25 | 0.11 | Genoa | 3.4 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 174 | WHITE SEABREAM | *Diplodus sargus sargus* | 194 | 24 | 0.18 | Genoa | 3.4 ± 0.1 |
| 175 | WHITE SEABREAM | *Diplodus sargus sargus* | 156 | 22 | 0.10 | Genoa | 3.4 ± 0.1 |
| 176 | WHITE SEABREAM | *Diplodus sargus sargus* | 170 | 22 | 0.15 | Genoa | 3.4 ± 0.1 |
| 177 | WHITE SEABREAM | *Diplodus sargus sargus* | 320 | 27 | 0.39 | Marseille | 3.4 ± 0.1 |
| 178 | WHITE SEABREAM | *Diplodus sargus sargus* | 178 | 23 | 0.26 | Marseille | 3.4 ± 0.1 |
| 179 | WHITE SEABREAM | *Diplodus sargus sargus* | 147 | 22 | 0.95 | Marseille | 3.4 ± 0.1 |
| 180 | WHITE SEABREAM | *Diplodus sargus sargus* | 144 | 21 | 0.15 | Marseille | 3.4 ± 0.1 |
| 181 | WHITE SEABREAM | *Diplodus sargus sargus* | 183 | 24 | 0.65 | Marseille | 3.4 ± 0.1 |
| 182 | WHITE SEABREAM | *Diplodus sargus sargus* | 168 | 21 | 0.18 | Marseille | 3.4 ± 0.1 |
| 183 | WHITE SEABREAM | *Diplodus sargus sargus* | 158 | 20 | 0.88 | Marseille | 3.4 ± 0.1 |
| 184 | WHITE SEABREAM | *Diplodus sargus sargus* | 174 | 21 | 0.13 | Marseille | 3.4 ± 0.1 |
| 185 | WHITE SEABREAM | *Diplodus sargus sargus* | 160 | 20 | 0.74 | Marseille | 3.4 ± 0.1 |
| 186 | WHITE SEABREAM | *Diplodus sargus sargus* | 126 | 19 | 0.91 | Marseille | 3.4 ± 0.1 |
| 187 | WHITE SEABREAM | *Diplodus sargus sargus* | 121 | 19 | 0.29 | Marseille | 3.4 ± 0.1 |
| 188 | WHITE SEABREAM | *Diplodus sargus sargus* | 77 | 18 | 0.65 | Marseille | 3.4 ± 0.1 |
| 189 | WHITE SEABREAM | *Diplodus sargus sargus* | 70 | 17 | 0.53 | Marseille | 3.4 ± 0.1 |
| 190 | WHITE SEABREAM | *Diplodus sargus sargus* | 197 | 23 | 0.77 | Marseille | 3.4 ± 0.1 |
| 191 | WHITE SEABREAM | *Diplodus sargus sargus* | 143 | 21 | 0.88 | Marseille | 3.4 ± 0.1 |
| 192 | WHITE SEABREAM | *Diplodus sargus sargus* | 274 | 25 | 0.93 | Marseille | 3.4 ± 0.1 |
| 193 | WHITE SEABREAM | *Diplodus sargus sargus* | 163 | 21.5 | 0.67 | Marseille | 3.4 ± 0.1 |
| 194 | ANCHOVIE | *Engraulis encrasicolus* | 12 | 13 | 0.06 | Balearic Islands | 3.1 ±0.36 |
| 195 | ANCHOVIE | *Engraulis encrasicolus* | 8 | 11 | 0.05 | Balearic Islands | 3.1 ±0.36 |
| 196 | ANCHOVIE | *Engraulis encrasicolus* | 27 | 17 | 0.07 | Balearic Islands | 3.1 ±0.36 |
| 197 | ANCHOVIE | *Engraulis encrasicolus* | 21 | 15 | 0.10 | Balearic Islands | 3.1 ±0.36 |
| 198 | ANCHOVIE | *Engraulis encrasicolus* | 17 | 15 | 0.06 | Balearic Islands | 3.1 ±0.36 |
| 199 | ANCHOVIE | *Engraulis encrasicolus* | 14 | 13 | 0.07 | Balearic Islands | 3.1 ±0.36 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 200 | ANCHOVIE | *Engraulis encrasicolus* | 15 | 13 | 0.08 | Balearic Islands | 3.1 ±0.36 |
| 201 | ANCHOVIE | *Engraulis encrasicolus* | 10 | 12 | 0.07 | Balearic Islands | 3.1 ±0.36 |
| 202 | ANCHOVIE | *Engraulis encrasicolus* | 18 | 15 | 0.08 | Balearic Islands | 3.1 ±0.36 |
| 203 | ANCHOVIE | *Engraulis encrasicolus* | 13 | 14 | 0.10 | Balearic Islands | 3.1 ±0.36 |
| 204 | ANCHOVIE | *Engraulis encrasicolus* | 19 | 15 | 0.06 | Balearic Islands | 3.1 ±0.36 |
| 205 | ANCHOVIE | *Engraulis encrasicolus* | 10 | 12 | 0.06 | Balearic Islands | 3.1 ±0.36 |
| 206 | ANCHOVIE | *Engraulis encrasicolus* | 14 | 14 | 0.05 | Balearic Islands | 3.1 ±0.36 |
| 207 | ANCHOVIE | *Engraulis encrasicolus* | 17 | 14 | 0.07 | Balearic Islands | 3.1 ±0.36 |
| 208 | ANCHOVIE | *Engraulis encrasicolus* | 12 | 13 | 0.04 | Alacant | 3.1 ±0.36 |
| 209 | ANCHOVIE | *Engraulis encrasicolus* | 8 | 11 | 0.03 | Alacant | 3.1 ±0.36 |
| 210 | ANCHOVIE | *Engraulis encrasicolus* | 16 | 13 | 0.03 | Alacant | 3.1 ±0.36 |
| 211 | ANCHOVIE | *Engraulis encrasicolus* | 10 | 12 | 0.04 | Alacant | 3.1 ±0.36 |
| 212 | ANCHOVIE | *Engraulis encrasicolus* | 11 | 12 | 0.04 | Alacant | 3.1 ±0.36 |
| 213 | ANCHOVIE | *Engraulis encrasicolus* | 7 | 11.5 | 0.04 | Alacant | 3.1 ±0.36 |
| 214 | ANCHOVIE | *Engraulis encrasicolus* | 11 | 12 | 0.05 | Alacant | 3.1 ±0.36 |
| 215 | ANCHOVIE | *Engraulis encrasicolus* | 12 | 13 | 0.03 | Alacant | 3.1 ±0.36 |
| 216 | ANCHOVIE | *Engraulis encrasicolus* | 18 | 14 | 0.13 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 217 | ANCHOVIE | *Engraulis encrasicolus* | 19 | 14 | 0.13 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 218 | ANCHOVIE | *Engraulis encrasicolus* | 24 | 16 | 0.24 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 219 | ANCHOVIE | *Engraulis encrasicolus* | 13 | 13 | 0.19 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 220 | ANCHOVIE | *Engraulis encrasicolus* | 18 | 15 | 0.12 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 221 | ANCHOVIE | *Engraulis encrasicolus* | 14 | 14 | 0.06 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 222 | ANCHOVIE | *Engraulis encrasicolus* | 16 | 14 | 0.10 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 223 | ANCHOVIE | *Engraulis encrasicolus* | 16 | 14 | 0.09 | L'Ampolla - Ebro Delta | 3.1 ±0.36 |
| 224 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 0.57 | Balearic Islands | 4.4 |
| 225 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 1.40 | Balearic Islands | 4.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 226 | DUSKY GROUPER | *Epinephelus marginatus* | 4150 | 64 | 0.92 | Balearic Islands | 4.4 |
| 227 | DUSKY GROUPER | *Epinephelus marginatus* | 4060 | 70 | 0.59 | Balearic Islands | 4.4 |
| 228 | DUSKY GROUPER | *Epinephelus marginatus* | 2800 | - | 0.73 | Balearic Islands | 4.4 |
| 229 | DUSKY GROUPER | *Epinephelus marginatus* | 2800 | - | 0.60 | Balearic Islands | 4.4 |
| 230 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 0.57 | Balearic Islands | 4.4 |
| 231 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 1.40 | Balearic Islands | 4.4 |
| 232 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 0.60 | Balearic Islands | 4.4 |
| 233 | DUSKY GROUPER | *Epinephelus marginatus* | 4800 | 70 | 0.73 | Balearic Islands | 4.4 |
| 234 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 0.59 | Balearic Islands | 4.4 |
| 235 | DUSKY GROUPER | *Epinephelus marginatus* | 4300 | 66 | 0.18 | Balearic Islands | 4.4 |
| 236 | DUSKY GROUPER | *Epinephelus marginatus* | 3400 | 60 | 0.27 | Balearic Islands | 4.4 |
| 237 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 0.47 | Balearic Islands | 4.4 |
| 238 | DUSKY GROUPER | *Epinephelus marginatus* | 4650 | 61 | 0.35 | Balearic Islands | 4.4 |
| 239 | DUSKY GROUPER | *Epinephelus marginatus* | 3600 | 67 | 0.24 | Balearic Islands | 4.4 |
| 240 | DUSKY GROUPER | *Epinephelus marginatus* | 3150 | 58 | 0.21 | Balearic Islands | 4.4 |
| 241 | DUSKY GROUPER | *Epinephelus marginatus* | 2950 | 59 | 0.20 | Balearic Islands | 4.4 |
| 242 | DUSKY GROUPER | *Epinephelus marginatus* | 3200 | 61 | 0.21 | Balearic Islands | 4.4 |
| 243 | DUSKY GROUPER | *Epinephelus marginatus* | 4350 | 65 | 0.30 | Balearic Islands | 4.4 |
| 244 | DUSKY GROUPER | *Epinephelus marginatus* | 5150 | 71 | 0.84 | Balearic Islands | 4.4 |
| 245 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 2.54 | Balearic Islands | 4.4 |
| 246 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 3.00 | Balearic Islands | 4.4 |
| 247 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 2.40 | Balearic Islands | 4.4 |
| 248 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 3.00 | Balearic Islands | 4.4 |
| 249 | DUSKY GROUPER | *Epinephelus marginatus* | 3200 | 60 | 1.80 | Balearic Islands | 4.4 |
| 250 | DUSKY GROUPER | *Epinephelus marginatus* | 3250 | 60 | 1.80 | Balearic Islands | 4.4 |
| 251 | DUSKY GROUPER | *Epinephelus marginatus* | 2600 | 120 | 2.40 | Balearic Islands | 4.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 252 | DUSKY GROUPER | *Epinephelus marginatus* | 2700 | 120 | 3.00 | Balearic Islands | 4.4 |
| 253 | DUSKY GROUPER | *Epinephelus marginatus* | 2600 | 120 | 3.00 | Balearic Islands | 4.4 |
| 254 | DUSKY GROUPER | *Epinephelus marginatus* | - | - | 2.54 | Balearic Islands | 4.4 |
| 255 | DUSKY GROUPER | *Epinephelus marginatus* | 5300 | 68 | 0.49 | Balearic Islands | 4.4 |
| 256 | DUSKY GROUPER | *Epinephelus marginatus* | 4100 | 65 | 0.34 | Balearic Islands | 4.4 |
| 257 | DUSKY GROUPER | *Epinephelus marginatus* | 3650 | 62 | 0.33 | Balearic Islands | 4.4 |
| 258 | DUSKY GROUPER | *Epinephelus marginatus* | 3250 | 58 | 0.30 | Balearic Islands | 4.4 |
| 259 | DUSKY GROUPER | *Epinephelus marginatus* | 3850 | 65 | 0.71 | Balearic Islands | 4.4 |
| 260 | DUSKY GROUPER | *Epinephelus marginatus* | 5550 | 72 | 1.00 | Balearic Islands | 4.4 |
| 261 | PORBEAGLE | *Lamna nasus* | 700 | - | 3.00 | Balearic Islands | 4.6 |
| 262 | FOUR-SPOT MEGRIM | *Lepidorhombus boscii* | - | - | 0.38 | Balearic Islands | 3.7 ±0.3 |
| 263 | ANGLER | *Lophius piscatorius* | 550 | - | 0.41 | Balearic Islands | 4.5 ± 0.1 |
| 264 | ANGLER | *Lophius piscatorius* | 250 | 36 | 0.57 | Balearic Islands | 4.5 ± 0.1 |
| 265 | ANGLER | *Lophius piscatorius* | 700 | 38 | 0.57 | Balearic Islands | 4.5 ± 0.1 |
| 266 | ANGLER | *Lophius piscatorius* | 500 | 48 | 0.56 | Balearic Islands | 4.5 ± 0.1 |
| 267 | ANGLER | *Lophius piscatorius* | 330 | 25 | 0.41 | Balearic Islands | 4.5 ± 0.1 |
| 268 | ANGLER | *Lophius piscatorius* | 450 | 34 | 0.32 | Balearic Islands | 4.5 ± 0.1 |
| 269 | ANGLER | *Lophius piscatorius* | 450 | 32 | 0.73 | Balearic Islands | 4.5 ± 0.1 |
| 270 | ANGLER | *Lophius piscatorius* | 450 | 31 | 0.12 | Balearic Islands | 4.5 ± 0.1 |
| 271 | ANGLER | *Lophius piscatorius* | 1300 | 44 | 0.36 | Balearic Islands | 4.5 ± 0.1 |
| 272 | ANGLER | *Lophius piscatorius* | 500 | 33 | 0.17 | Balearic Islands | 4.5 ± 0.1 |
| 273 | ANGLER | *Lophius piscatorius* | 1100 | 47 | 0.31 | Balearic Islands | 4.5 ± 0.1 |
| 274 | ANGLER | *Lophius piscatorius* | 1360 | 47 | 0.36 | Balearic Islands | 4.5 ± 0.1 |
| 275 | ANGLER | *Lophius piscatorius* | 920 | 41 | 0.37 | Balearic Islands | 4.5 ± 0.1 |
| 276 | ANGLER | *Lophius piscatorius* | 440 | - | 0.33 | Balearic Islands | 4.5 ± 0.1 |
| 277 | ANGLER | *Lophius piscatorius* | 520 | 33 | 0.73 | Balearic Islands | 4.5 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 278 | ANGLER | *Lophius piscatorius* | 540 | 32 | 0.98 | Balearic Islands | 4.5 ± 0.1 |
| 279 | ANGLER | *Lophius piscatorius* | - | - | 0.65 | Balearic Islands | 4.5 ± 0.1 |
| 280 | ANGLER | *Lophius piscatorius* | - | - | 3.10 | Balearic Islands | 4.5 ± 0.1 |
| 281 | ANGLER | *Lophius piscatorius* | 2300 | 50 | 0.65 | Balearic Islands | 4.5 ± 0.1 |
| 282 | ANGLER | *Lophius piscatorius* | 5500 | 70 | 3.10 | Balearic Islands | 4.5 ± 0.1 |
| 283 | ANGLER | *Lophius piscatorius* | 900 | - | 0.34 | Balearic Islands | 4.5 ± 0.1 |
| 284 | ANGLER | *Lophius piscatorius* | 1600 | - | 0.64 | Balearic Islands | 4.5 ± 0.1 |
| 285 | ANGLER | *Lophius piscatorius* | 700 | - | 0.43 | Balearic Islands | 4.5 ± 0.1 |
| 286 | ANGLER | *Lophius piscatorius* | - | - | 0.64 | Balearic Islands | 4.5 ± 0.1 |
| 287 | ANGLER | *Lophius piscatorius* | - | - | 0.67 | Balearic Islands | 4.5 ± 0.1 |
| 288 | ANGLER | *Lophius piscatorius* | 3000 | - | 0.94 | Balearic Islands | 4.5 ± 0.1 |
| 289 | ANGLER | *Lophius piscatorius* | - | - | 1.00 | Balearic Islands | 4.5 ± 0.1 |
| 290 | ANGLER | *Lophius piscatorius* | 3000 | - | 1.80 | Balearic Islands | 4.5 ± 0.1 |
| 291 | ANGLER | *Lophius piscatorius* | 3000 | - | 3.00 | Balearic Islands | 4.5 ± 0.1 |
| 292 | ANGLER | *Lophius piscatorius* | 3000 | - | 0.79 | Balearic Islands | 4.5 ± 0.1 |
| 293 | ANGLER | *Lophius piscatorius* | 385 | 24 | 0.75 | Balearic Islands | 4.5 ± 0.1 |
| 294 | ANGLER | *Lophius piscatorius* | 250 | 18 | 0.84 | Balearic Islands | 4.5 ± 0.1 |
| 295 | ANGLER | *Lophius piscatorius* | 420 | 30 | 0.36 | Balearic Islands | 4.5 ± 0.1 |
| 296 | ANGLER | *Lophius piscatorius* | 430 | 25 | 0.44 | Balearic Islands | 4.5 ± 0.1 |
| 297 | ANGLER | *Lophius piscatorius* | 1200 | 43 | 0.41 | Balearic Islands | 4.5 ± 0.1 |
| 298 | ANGLER | *Lophius piscatorius* | 540 | 30 | 1.10 | Balearic Islands | 4.5 ± 0.1 |
| 299 | ANGLER | *Lophius piscatorius* | - | - | 0.27 | Balearic Islands | 4.5 ± 0.1 |
| 300 | ANGLER | *Lophius piscatorius* | - | - | 0.62 | Balearic Islands | 4.5 ± 0.1 |
| 301 | ANGLER | *Lophius piscatorius* | - | - | 0.62 | Balearic Islands | 4.5 ± 0.1 |
| 302 | ANGLER | *Lophius piscatorius* | - | - | 0.27 | Balearic Islands | 4.5 ± 0.1 |
| 303 | ANGLER | *Lophius piscatorius* | 400 | 28 | 0.67 | Balearic Islands | 4.5 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 304 | ANGLER | *Lophius piscatorius* | 985 | 34 | 0.85 | Balearic Islands | 4.5 ± 0.1 |
| 305 | ANGLER | *Lophius piscatorius* | 615 | 33 | 0.55 | Balearic Islands | 4.5 ± 0.1 |
| 306 | ANGLER | *Lophius piscatorius* | 677 | 32 | 0.52 | Balearic Islands | 4.5 ± 0.1 |
| 307 | ANGLER | *Lophius piscatorius* | 750 | 35 | 0.46 | Balearic Islands | 4.5 ± 0.1 |
| 308 | ANGLER | *Lophius piscatorius* | 659 | 32 | 0.44 | Balearic Islands | 4.5 ± 0.1 |
| 309 | ANGLER | *Lophius piscatorius* | 875 | 40 | 0.38 | Balearic Islands | 4.5 ± 0.1 |
| 310 | ANGLER | *Lophius piscatorius* | 1427 | 41 | 0.61 | Balearic Islands | 4.5 ± 0.1 |
| 311 | ANGLER | *Lophius piscatorius* | 1152 | 38 | 0.78 | Balearic Islands | 4.5 ± 0.1 |
| 312 | ANGLER | *Lophius piscatorius* | 329 | 29 | 0.49 | Balearic Islands | 4.5 ± 0.1 |
| 313 | ANGLER | *Lophius piscatorius* | 341 | 30 | 0.56 | Alacant | 4.5 ± 0.1 |
| 314 | ANGLER | *Lophius piscatorius* | 378 | 32 | 0.58 | Alacant | 4.5 ± 0.1 |
| 315 | ANGLER | *Lophius piscatorius* | 345 | 31.5 | 0.41 | Alacant | 4.5 ± 0.1 |
| 316 | ANGLER | *Lophius piscatorius* | 412 | 32 | 0.82 | Alacant | 4.5 ± 0.1 |
| 317 | ANGLER | *Lophius piscatorius* | 312 | 30 | 0.67 | Alacant | 4.5 ± 0.1 |
| 318 | ANGLER | *Lophius piscatorius* | 724 | 42 | 0.50 | Alacant | 4.5 ± 0.1 |
| 319 | ANGLER | *Lophius piscatorius* | 864 | 41 | 0.55 | Alacant | 4.5 ± 0.1 |
| 320 | ANGLER | *Lophius piscatorius* | 552 | 44 | 1.04 | Alacant | 4.5 ± 0.1 |
| 321 | ANGLER | *Lophius piscatorius* | 181 | 26 | 1.10 | Civitavecchia | 4.5 ± 0.1 |
| 322 | ANGLER | *Lophius piscatorius* | 73 | 18 | 0.20 | Civitavecchia | 4.5 ± 0.1 |
| 323 | ANGLER | *Lophius piscatorius* | 1153 | 44 | 0.11 | Civitavecchia | 4.5 ± 0.1 |
| 324 | ANGLER | *Lophius piscatorius* | 815 | 40 | 1.85 | Civitavecchia | 4.5 ± 0.1 |
| 325 | ANGLER | *Lophius piscatorius* | 645 | 37 | 1.03 | Civitavecchia | 4.5 ± 0.1 |
| 326 | ANGLER | *Lophius piscatorius* | 548 | 34 | 1.95 | Civitavecchia | 4.5 ± 0.1 |
| 327 | ANGLER | *Lophius piscatorius* | 616 | 37 | 1.48 | Civitavecchia | 4.5 ± 0.1 |
| 328 | ANGLER | *Lophius piscatorius* | 499 | 34 | 1.23 | Civitavecchia | 4.5 ± 0.1 |
| 329 | ANGLER | *Lophius piscatorius* | 1085 | 43 | 1.11 | Civitavecchia | 4.5 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 330 | ANGLER | *Lophius piscatorius* | 1947 | 52 | 2.27 | Civitavecchia | 4.5 ± 0.1 |
| 331 | ANGLER | *Lophius piscatorius* | 1005 | 42 | 2.08 | Civitavecchia | 4.5 ± 0.1 |
| 332 | ANGLER | *Lophius piscatorius* | 597 | 36 | 1.64 | Civitavecchia | 4.5 ± 0.1 |
| 333 | ANGLER | *Lophius piscatorius* | 541 | 35 | 2.03 | Civitavecchia | 4.5 ± 0.1 |
| 334 | ANGLER | *Lophius piscatorius* | 514 | 34 | 2.98 | Civitavecchia | 4.5 ± 0.1 |
| 335 | ANGLER | *Lophius piscatorius* | 877 | 41 | 2.15 | Civitavecchia | 4.5 ± 0.1 |
| 336 | ANGLER | *Lophius piscatorius* | 1407 | 45 | 1.82 | Civitavecchia | 4.5 ± 0.1 |
| 337 | ANGLER | *Lophius piscatorius* | 1874 | 47 | 0.09 | Civitavecchia | 4.5 ± 0.1 |
| 338 | ANGLER | *Lophius piscatorius* | 1806 | 60 | 0.23 | Alghero | 4.5 ± 0.1 |
| 339 | ANGLER | *Lophius piscatorius* | 1642 | 53 | 4.49 | Alghero | 4.5 ± 0.1 |
| 340 | ANGLER | *Lophius piscatorius* | 1750 | 58 | 0.21 | Alghero | 4.5 ± 0.1 |
| 341 | ANGLER | *Lophius piscatorius* | 2486 | 65 | 0.11 | Alghero | 4.5 ± 0.1 |
| 342 | ANGLER | *Lophius piscatorius* | 419 | 34 | 0.52 | Genoa | 4.5 ± 0.1 |
| 343 | ANGLER | *Lophius piscatorius* | 543 | 39 | 0.83 | Genoa | 4.5 ± 0.1 |
| 344 | ANGLER | *Lophius piscatorius* | 524 | 39 | 1.05 | Genoa | 4.5 ± 0.1 |
| 345 | ANGLER | *Lophius piscatorius* | 712 | 42 | 0.82 | Genoa | 4.5 ± 0.1 |
| 346 | ANGLER | *Lophius piscatorius* | 583 | 37 | 1.00 | Genoa | 4.5 ± 0.1 |
| 347 | ANGLER | *Lophius piscatorius* | 734 | 41 | 1.67 | Genoa | 4.5 ± 0.1 |
| 348 | ANGLER | *Lophius piscatorius* | 697 | 40 | 0.81 | Genoa | 4.5 ± 0.1 |
| 349 | ANGLER | *Lophius piscatorius* | 456 | 36 | 0.60 | Genoa | 4.5 ± 0.1 |
| 350 | ANGLER | *Lophius piscatorius* | 190 | 22 | 0.44 | Marseille | 4.5 ± 0.1 |
| 351 | ANGLER | *Lophius piscatorius* | 161 | 25 | 1.36 | Marseille | 4.5 ± 0.1 |
| 352 | ANGLER | *Lophius piscatorius* | 543 | 35 | 1.15 | Marseille | 4.5 ± 0.1 |
| 353 | ANGLER | *Lophius piscatorius* | 199 | 26 | 1.05 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 354 | ANGLER | *Lophius piscatorius* | 274 | 27 | 1.05 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 355 | ANGLER | *Lophius piscatorius* | 169 | 25 | 0.81 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 356 | ANGLER | *Lophius piscatorius* | 222 | 24 | 0.38 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 357 | ANGLER | *Lophius piscatorius* | 258 | 24 | 0.60 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 358 | ANGLER | *Lophius piscatorius* | 134 | 24 | 1.40 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 359 | ANGLER | *Lophius piscatorius* | 159 | 24 | 0.48 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 360 | ANGLER | *Lophius piscatorius* | 236 | 26 | 1.12 | L'Ampolla - Ebro Delta | 4.5 ± 0.1 |
| 361 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 1.22 | Balearic Islands | 4.4 |
| 362 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 0.56 | Balearic Islands | 4.4 |
| 363 | EUROPEAN HAKE | *Merluccius merluccius* | 350 | 34 | 0.10 | Balearic Islands | 4.4 |
| 364 | EUROPEAN HAKE | *Merluccius merluccius* | 300 | 31.5 | 0.10 | Balearic Islands | 4.4 |
| 365 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | 35 | 0.17 | Balearic Islands | 4.4 |
| 366 | EUROPEAN HAKE | *Merluccius merluccius* | 300 | 37 | 0.44 | Balearic Islands | 4.4 |
| 367 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 0.09 | Balearic Islands | 4.4 |
| 368 | EUROPEAN HAKE | *Merluccius merluccius* | 300 | 32 | 0.13 | Balearic Islands | 4.4 |
| 369 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | 31 | 0.15 | Balearic Islands | 4.4 |
| 370 | EUROPEAN HAKE | *Merluccius merluccius* | 520 | 22 | 0.13 | Balearic Islands | 4.4 |
| 371 | EUROPEAN HAKE | *Merluccius merluccius* | 420 | 28 | 0.09 | Balearic Islands | 4.4 |
| 372 | EUROPEAN HAKE | *Merluccius merluccius* | 560 | 35 | 0.09 | Balearic Islands | 4.4 |
| 373 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | 33 | 0.50 | Balearic Islands | 4.4 |
| 374 | EUROPEAN HAKE | *Merluccius merluccius* | 280 | 31 | 0.23 | Balearic Islands | 4.4 |
| 375 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | 30 | 0.18 | Balearic Islands | 4.4 |
| 376 | EUROPEAN HAKE | *Merluccius merluccius* | 340 | 22 | 0.11 | Balearic Islands | 4.4 |
| 377 | EUROPEAN HAKE | *Merluccius merluccius* | 400 | 25 | 0.14 | Balearic Islands | 4.4 |
| 378 | EUROPEAN HAKE | *Merluccius merluccius* | 92 | 20 | 0.17 | Balearic Islands | 4.4 |
| 379 | EUROPEAN HAKE | *Merluccius merluccius* | 270 | 37 | 0.26 | Balearic Islands | 4.4 |
| 380 | EUROPEAN HAKE | *Merluccius merluccius* | 1300 | - | 0.99 | Balearic Islands | 4.4 |
| 381 | EUROPEAN HAKE | *Merluccius merluccius* | 600 | - | 0.51 | Balearic Islands | 4.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 382 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 0.36 | Balearic Islands | 4.4 |
| 383 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | - | 0.21 | Balearic Islands | 4.4 |
| 384 | EUROPEAN HAKE | *Merluccius merluccius* | 440 | - | 0.35 | Balearic Islands | 4.4 |
| 385 | EUROPEAN HAKE | *Merluccius merluccius* | 605 | - | 0.40 | Balearic Islands | 4.4 |
| 386 | EUROPEAN HAKE | *Merluccius merluccius* | 1500 | - | 0.52 | Balearic Islands | 4.4 |
| 387 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 0.56 | Balearic Islands | 4.4 |
| 388 | EUROPEAN HAKE | *Merluccius merluccius* | - | - | 1.22 | Balearic Islands | 4.4 |
| 389 | EUROPEAN HAKE | *Merluccius merluccius* | 550 | 35 | 0.56 | Balearic Islands | 4.4 |
| 390 | EUROPEAN HAKE | *Merluccius merluccius* | 300 | 25 | 0.12 | Balearic Islands | 4.4 |
| 391 | EUROPEAN HAKE | *Merluccius merluccius* | 250 | 25 | 0.13 | Balearic Islands | 4.4 |
| 392 | EUROPEAN HAKE | *Merluccius merluccius* | 620 | 35 | 0.58 | Balearic Islands | 4.4 |
| 393 | EUROPEAN HAKE | *Merluccius merluccius* | 280 | 26 | 0.83 | Balearic Islands | 4.4 |
| 394 | EUROPEAN HAKE | *Merluccius merluccius* | 270 | 25 | 0.61 | Balearic Islands | 4.4 |
| 395 | EUROPEAN HAKE | *Merluccius merluccius* | 300 | 26 | 0.15 | Balearic Islands | 4.4 |
| 396 | EUROPEAN HAKE | *Merluccius merluccius* | 296 | 45 | 0.16 | Alacant | 4.4 |
| 397 | EUROPEAN HAKE | *Merluccius merluccius* | 345 | 48.5 | 0.16 | Alacant | 4.4 |
| 398 | EUROPEAN HAKE | *Merluccius merluccius* | 321 | 47 | 0.07 | Alacant | 4.4 |
| 399 | EUROPEAN HAKE | *Merluccius merluccius* | 326 | 45 | 0.20 | Alacant | 4.4 |
| 400 | EUROPEAN HAKE | *Merluccius merluccius* | 308 | 45 | 0.11 | Alacant | 4.4 |
| 401 | EUROPEAN HAKE | *Merluccius merluccius* | 350 | 45.5 | 0.31 | Alacant | 4.4 |
| 402 | EUROPEAN HAKE | *Merluccius merluccius* | 302 | 46 | 0.16 | Alacant | 4.4 |
| 403 | EUROPEAN HAKE | *Merluccius merluccius* | 256 | 44 | 0.15 | Alacant | 4.4 |
| 404 | EUROPEAN HAKE | *Merluccius merluccius* | 340 | 35 | 0.06 | Marseille | 4.4 |
| 405 | EUROPEAN HAKE | *Merluccius merluccius* | 372 | 36 | 0.05 | Marseille | 4.4 |
| 406 | EUROPEAN HAKE | *Merluccius merluccius* | 284 | 37 | 0.09 | Marseille | 4.4 |
| 407 | EUROPEAN HAKE | *Merluccius merluccius* | 209 | 31 | 0.05 | Marseille | 4.4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 408 | EUROPEAN HAKE | *Merluccius merluccius* | 302 | 38 | 0.08 | Marseille | 4.4 |
| 409 | EUROPEAN HAKE | *Merluccius merluccius* | 361 | 39 | 0.05 | Marseille | 4.4 |
| 410 | EUROPEAN HAKE | *Merluccius merluccius* | 211 | 33 | 0.08 | Marseille | 4.4 |
| 411 | EUROPEAN HAKE | *Merluccius merluccius* | 173 | 30 | 0.06 | Marseille | 4.4 |
| 412 | EUROPEAN HAKE | *Merluccius merluccius* | 192 | 30.5 | 0.08 | Marseille | 4.4 |
| 413 | EUROPEAN HAKE | *Merluccius merluccius* | 275 | 37 | 0.13 | Marseille | 4.4 |
| 414 | EUROPEAN HAKE | *Merluccius merluccius* | 717 | 45 | 0.26 | Marseille | 4.4 |
| 415 | EUROPEAN HAKE | *Merluccius merluccius* | 406 | 41 | 0.57 | Marseille | 4.4 |
| 416 | EUROPEAN HAKE | *Merluccius merluccius* | 294 | 35 | 0.50 | L'Ampolla - Ebro Delta | 4.4 |
| 417 | EUROPEAN HAKE | *Merluccius merluccius* | 266 | 35 | 0.20 | L'Ampolla - Ebro Delta | 4.4 |
| 418 | EUROPEAN HAKE | *Merluccius merluccius* | 399 | 38 | 0.13 | L'Ampolla - Ebro Delta | 4.4 |
| 419 | EUROPEAN HAKE | *Merluccius merluccius* | 231 | 32 | 0.23 | L'Ampolla - Ebro Delta | 4.4 |
| 420 | EUROPEAN HAKE | *Merluccius merluccius* | 279 | 34 | 0.13 | L'Ampolla - Ebro Delta | 4.4 |
| 421 | EUROPEAN HAKE | *Merluccius merluccius* | 274 | 34 | 0.26 | L'Ampolla - Ebro Delta | 4.4 |
| 422 | EUROPEAN HAKE | *Merluccius merluccius* | 246 | 34 | 0.29 | L'Ampolla - Ebro Delta | 4.4 |
| 423 | EUROPEAN HAKE | *Merluccius merluccius* | 307 | 35 | 0.12 | L'Ampolla - Ebro Delta | 4.4 |
| 424 | RED MULLET | *Mullus barbatus* | 48 | 15 | 0.13 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 425 | RED MULLET | *Mullus barbatus* | 39 | 16 | 0.10 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 426 | RED MULLET | *Mullus barbatus* | 46 | 16 | 0.43 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 427 | RED MULLET | *Mullus barbatus* | 28 | 13 | 0.08 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 428 | RED MULLET | *Mullus barbatus* | 46 | 16 | 0.06 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 429 | RED MULLET | *Mullus barbatus* | 30 | 14 | 0.12 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 430 | RED MULLET | *Mullus barbatus* | 23 | 12 | 0.08 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 431 | RED MULLET | *Mullus barbatus* | 25 | 14 | 0.13 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 432 | RED MULLET | *Mullus barbatus* | 30 | 13 | 0.05 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 433 | RED MULLET | *Mullus barbatus* | 23 | 13 | 0.07 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 434 | RED MULLET | *Mullus barbatus* | - | - | 0.17 | Balearic Islands | 3.1 ± 0.1 |
| 435 | RED MULLET | *Mullus barbatus* | 58 | 17 | 0.23 | Balearic Islands | 3.1 ± 0.1 |
| 436 | RED MULLET | *Mullus barbatus* | 50 | 16 | 0.10 | Balearic Islands | 3.1 ± 0.1 |
| 437 | RED MULLET | *Mullus barbatus* | 63 | 18 | 0.19 | Balearic Islands | 3.1 ± 0.1 |
| 438 | RED MULLET | *Mullus barbatus* | 65 | 19 | 0.23 | Balearic Islands | 3.1 ± 0.1 |
| 439 | RED MULLET | *Mullus barbatus* | 47 | 13 | 0.10 | Balearic Islands | 3.1 ± 0.1 |
| 440 | RED MULLET | *Mullus barbatus* | 39 | 12 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 441 | RED MULLET | *Mullus barbatus* | 57 | 17 | 0.12 | Balearic Islands | 3.1 ± 0.1 |
| 442 | RED MULLET | *Mullus barbatus* | 61 | 17 | 0.15 | Balearic Islands | 3.1 ± 0.1 |
| 443 | RED MULLET | *Mullus barbatus* | 54 | 17 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 444 | RED MULLET | *Mullus barbatus* | 150 | 22.5 | 0.88 | Marseille | 3.1 ± 0.1 |
| 445 | RED MULLET | *Mullus barbatus* | 123 | 22 | 0.15 | Marseille | 3.1 ± 0.1 |
| 446 | RED MULLET | *Mullus barbatus* | 132 | 22 | 0.22 | Marseille | 3.1 ± 0.1 |
| 447 | RED MULLET | *Mullus barbatus* | 93 | 20 | 0.11 | Marseille | 3.1 ± 0.1 |
| 448 | RED MULLET | *Mullus barbatus* | 161 | 24 | 0.32 | Marseille | 3.1 ± 0.1 |
| 449 | RED MULLET | *Mullus barbatus* | 183 | 25 | 0.24 | Marseille | 3.1 ± 0.1 |
| 450 | RED MULLET | *Mullus barbatus* | 236 | 27 | 0.45 | Marseille | 3.1 ± 0.1 |
| 451 | RED MULLET | *Mullus barbatus* | 86 | 19 | 0.44 | Marseille | 3.1 ± 0.1 |
| 452 | RED MULLET | *Mullus barbatus* | 128 | 23 | 0.45 | Marseille | 3.1 ± 0.1 |
| 453 | RED MULLET | *Mullus barbatus* | 184 | 23 | 0.18 | Marseille | 3.1 ± 0.1 |
| 454 | RED MULLET | *Mullus barbatus* | 93 | 19 | 0.16 | Marseille | 3.1 ± 0.1 |
| 455 | RED MULLET | *Mullus barbatus* | 128 | 21 | 0.24 | Marseille | 3.1 ± 0.1 |
| 456 | RED MULLET | *Mullus barbatus* | 77 | 18 | 0.17 | Alghero | 3.1 ± 0.1 |
| 457 | RED MULLET | *Mullus barbatus* | 77 | 20 | 0.21 | Alghero | 3.1 ± 0.1 |
| 458 | RED MULLET | *Mullus barbatus* | 93 | 22 | 0.28 | Alghero | 3.1 ± 0.1 |
| 459 | RED MULLET | *Mullus barbatus* | 66 | 17 | 0.13 | Alghero | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 460 | RED MULLET | *Mullus barbatus* | 78 | 19 | 1.50 | Alghero | 3.1 ± 0.1 |
| 461 | RED MULLET | *Mullus barbatus* | 64 | 17 | 0.16 | Alghero | 3.1 ± 0.1 |
| 462 | RED MULLET | *Mullus barbatus* | 36 | 15 | 0.11 | Alghero | 3.1 ± 0.1 |
| 463 | RED MULLET | *Mullus barbatus* | 68 | 18 | 0.10 | Alghero | 3.1 ± 0.1 |
| 464 | RED MULLET | *Mullus barbatus* | 58 | 16 | 0.14 | Alghero | 3.1 ± 0.1 |
| 465 | RED MULLET | *Mullus barbatus* | 56 | 17 | 0.67 | Alghero | 3.1 ± 0.1 |
| 466 | RED MULLET | *Mullus barbatus* | 69 | 20 | 1.71 | Alghero | 3.1 ± 0.1 |
| 467 | RED MULLET | *Mullus barbatus* | 62 | 17 | 0.09 | Alghero | 3.1 ± 0.1 |
| 468 | RED MULLET | *Mullus barbatus* | 38 | 16 | 0.17 | Alghero | 3.1 ± 0.1 |
| 469 | RED MULLET | *Mullus barbatus* | 287 | 32 | 0.05 | Alacant | 3.1 ± 0.1 |
| 470 | RED MULLET | *Mullus barbatus* | 36 | 15 | 0.17 | Alacant | 3.1 ± 0.1 |
| 471 | RED MULLET | *Mullus barbatus* | 61 | 18 | 0.13 | Alacant | 3.1 ± 0.1 |
| 472 | RED MULLET | *Mullus barbatus* | 122 | 21 | 0.16 | Alacant | 3.1 ± 0.1 |
| 473 | RED MULLET | *Mullus barbatus* | 87 | 21 | 0.21 | Alacant | 3.1 ± 0.1 |
| 474 | RED MULLET | *Mullus barbatus* | 124 | 27 | 0.13 | Alacant | 3.1 ± 0.1 |
| 475 | RED MULLET | *Mullus barbatus* | 99 | 23 | 0.65 | Civitavecchia | 3.1 ± 0.1 |
| 476 | RED MULLET | *Mullus barbatus* | 115 | 22 | 1.24 | Civitavecchia | 3.1 ± 0.1 |
| 477 | RED MULLET | *Mullus barbatus* | 183 | 27 | 2.45 | Civitavecchia | 3.1 ± 0.1 |
| 478 | RED MULLET | *Mullus barbatus* | 97 | 21.5 | 1.35 | Civitavecchia | 3.1 ± 0.1 |
| 479 | RED MULLET | *Mullus barbatus* | 86 | 21 | 0.83 | Civitavecchia | 3.1 ± 0.1 |
| 480 | RED MULLET | *Mullus barbatus* | 137 | 24 | 1.09 | Civitavecchia | 3.1 ± 0.1 |
| 481 | RED MULLET | *Mullus barbatus* | 140 | 26 | 0.97 | Civitavecchia | 3.1 ± 0.1 |
| 482 | RED MULLET | *Mullus barbatus* | 81 | 22 | 0.61 | Civitavecchia | 3.1 ± 0.1 |
| 483 | RED MULLET | *Mullus barbatus* | 73 | 19 | 0.19 | Genoa | 3.1 ± 0.1 |
| 484 | RED MULLET | *Mullus barbatus* | 83 | 19 | 0.09 | Genoa | 3.1 ± 0.1 |
| 485 | RED MULLET | *Mullus barbatus* | 72 | 18 | 0.14 | Genoa | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 486 | RED MULLET | *Mullus barbatus* | 91 | 19 | 0.15 | Genoa | 3.1 ± 0.1 |
| 487 | RED MULLET | *Mullus barbatus* | 99 | 20 | 0.14 | Genoa | 3.1 ± 0.1 |
| 488 | RED MULLET | *Mullus barbatus* | 125 | 21 | 0.18 | Genoa | 3.1 ± 0.1 |
| 489 | RED MULLET | *Mullus barbatus* | 108 | 22 | 0.14 | Genoa | 3.1 ± 0.1 |
| 490 | RED MULLET | *Mullus barbatus* | 112 | 20 | 0.10 | Genoa | 3.1 ± 0.1 |
| 491 | SURMULLET | *Mullus surmuletus* | 250 | 21 | 0.15 | Balearic Islands | 3.5 ±0.3 |
| 492 | SURMULLET | *Mullus surmuletus* | 250 | 22 | 0.18 | Balearic Islands | 3.5 ±0.3 |
| 493 | SURMULLET | *Mullus surmuletus* | 300 | 19 | 0.26 | Balearic Islands | 3.5 ±0.3 |
| 494 | SURMULLET | *Mullus surmuletus* | 480 | 20 | 0.21 | Balearic Islands | 3.5 ±0.3 |
| 495 | SURMULLET | *Mullus surmuletus* | 400 | 16 | 0.09 | Balearic Islands | 3.5 ±0.3 |
| 496 | SURMULLET | *Mullus surmuletus* | 420 | 18 | 0.09 | Balearic Islands | 3.5 ±0.3 |
| 497 | SURMULLET | *Mullus surmuletus* | - | - | 0.17 | Balearic Islands | 3.5 ±0.3 |
| 498 | SURMULLET | *Mullus surmuletus* | - | - | 0.39 | Balearic Islands | 3.5 ±0.3 |
| 499 | SURMULLET | *Mullus surmuletus* | 280 | 30 | 0.49 | Balearic Islands | 3.5 ±0.3 |
| 500 | SURMULLET | *Mullus surmuletus* | 260 | 20 | 0.09 | Balearic Islands | 3.5 ±0.3 |
| 501 | SURMULLET | *Mullus surmuletus* | 270 | 15 | 0.15 | Balearic Islands | 3.5 ±0.3 |
| 502 | SURMULLET | *Mullus surmuletus* | - | - | 0.39 | Balearic Islands | 3.5 ±0.3 |
| 503 | SURMULLET | *Mullus surmuletus* | 84 | 20 | 0.20 | Alacant | 3.5 ±0.3 |
| 504 | SURMULLET | *Mullus surmuletus* | 20 | 11 | 0.06 | Alacant | 3.5 ±0.3 |
| 505 | SURMULLET | *Mullus surmuletus* | 27 | 12 | 0.08 | Alacant | 3.5 ±0.3 |
| 506 | SURMULLET | *Mullus surmuletus* | 124 | 22 | 0.13 | Alacant | 3.5 ±0.3 |
| 507 | SURMULLET | *Mullus surmuletus* | 100 | 23 | 0.10 | Alacant | 3.5 ±0.3 |
| 508 | SURMULLET | *Mullus surmuletus* | 236 | 31 | 0.09 | Alacant | 3.5 ±0.3 |
| 509 | MED. MORAY | *Muraena helena* | 332 | 75 | 0.30 | Balearic Islands | 4.2 ±0.61 |
| 510 | MED. MORAY | *Muraena helena* | 296 | 100 | 0.36 | Balearic Islands | 4.2 ±0.61 |
| 511 | MED. MORAY | *Muraena helena* | 421 | 100 | 0.52 | Balearic Islands | 4.2 ±0.61 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 512 | MED. MORAY | *Muraena helena* | 1600 | - | 0.30 | Balearic Islands | 4.2 ±0.61 |
| 513 | MED. MORAY | *Muraena helena* | 1800 | - | 0.39 | Balearic Islands | 4.2 ±0.61 |
| 514 | MED. MORAY | *Muraena helena* | 1400 | - | 0.50 | Balearic Islands | 4.2 ±0.61 |
| 515 | MED. MORAY | *Muraena helena* | 700 | 70 | 0.65 | Balearic Islands | 4.2 ±0.61 |
| 516 | MED. MORAY | *Muraena helena* | 360 | 60 | 0.47 | Balearic Islands | 4.2 ±0.61 |
| 517 | MED. MORAY | *Muraena helena* | 330 | 83 | 0.47 | Balearic Islands | 4.2 ±0.61 |
| 518 | MED. MORAY | *Muraena helena* | 1100 | 82 | 0.43 | Balearic Islands | 4.2 ±0.61 |
| 519 | MED. MORAY | *Muraena helena* | - | - | 0.43 | Balearic Islands | 4.2 ±0.61 |
| 520 | MED. MORAY | *Muraena helena* | - | - | 0.47 | Balearic Islands | 4.2 ±0.61 |
| 521 | MED. MORAY | *Muraena helena* | 700 | 75 | 0.45 | Balearic Islands | 4.2 ±0.61 |
| 522 | MED. MORAY | *Muraena helena* | - | - | 0.30 | Balearic Islands | 4.2 ±0.61 |
| 523 | MED. MORAY | *Muraena helena* | 1640 | 80 | 0.39 | Balearic Islands | 4.2 ±0.61 |
| 524 | MED. MORAY | *Muraena helena* | - | - | 0.50 | Balearic Islands | 4.2 ±0.61 |
| 525 | MED. MORAY | *Muraena helena* | 2000 | 100 | 0.29 | Balearic Islands | 4.2 ±0.61 |
| 526 | MED. MORAY | *Muraena helena* | 2200 | 100 | 0.42 | Balearic Islands | 4.2 ±0.61 |
| 527 | MED. MORAY | *Muraena helena* | 1640 | 80 | 0.43 | Balearic Islands | 4.2 ±0.61 |
| 528 | MED. MORAY | *Muraena helena* | 2000 | 90 | 0.24 | Balearic Islands | 4.2 ±0.61 |
| 529 | MED. MORAY | *Muraena helena* | 2280 | 101 | 0.35 | Balearic Islands | 4.2 ±0.61 |
| 530 | MED. MORAY | *Muraena helena* | 3000 | 115 | 0.38 | Balearic Islands | 4.2 ±0.61 |
| 531 | MED. MORAY | *Muraena helena* | 2925 | 104 | 0.57 | Balearic Islands | 4.2 ±0.61 |
| 532 | MED. MORAY | *Muraena helena* | 2500 | 102 | 0.37 | Balearic Islands | 4.2 ±0.61 |
| 533 | MED. MORAY | *Muraena helena* | 2100 | 85 | 0.42 | Balearic Islands | 4.2 ±0.61 |
| 534 | MED. MORAY | *Muraena helena* | 3185 | 94 | 0.53 | Balearic Islands | 4.2 ±0.61 |
| 535 | MED. MORAY | *Muraena helena* | 2940 | 95 | 0.39 | Balearic Islands | 4.2 ±0.61 |
| 536 | MED. MORAY | *Muraena helena* | 2660 | 100 | 0.51 | Balearic Islands | 4.2 ±0.61 |
| 537 | MED. MORAY | *Muraena helena* | 1000 | - | 0.31 | Balearic Islands | 4.2 ±0.61 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 538 | MED. MORAY | *Muraena helena* | 460 | - | 0.55 | Balearic Islands | 4.2 ±0.61 |
| 539 | MED. MORAY | *Muraena helena* | 2700 | - | 0.34 | Balearic Islands | 4.2 ±0.61 |
| 540 | MED. MORAY | *Muraena helena* | - | - | 0.37 | Balearic Islands | 4.2 ±0.61 |
| 541 | MED. MORAY | *Muraena helena* | 2000 | - | 0.37 | Balearic Islands | 4.2 ±0.61 |
| 542 | MED. MORAY | *Muraena helena* | - | - | 0.39 | Balearic Islands | 4.2 ±0.61 |
| 543 | MED. MORAY | *Muraena helena* | 3000 | - | 0.48 | Balearic Islands | 4.2 ±0.61 |
| 544 | MED. MORAY | *Muraena helena* | 2000 | - | 0.29 | Balearic Islands | 4.2 ±0.61 |
| 545 | MED. MORAY | *Muraena helena* | 1000 | - | 0.61 | Balearic Islands | 4.2 ±0.61 |
| 546 | MED. MORAY | *Muraena helena* | - | - | 1.11 | Balearic Islands | 4.2 ±0.61 |
| 547 | MED. MORAY | *Muraena helena* | 285 | 60 | 0.28 | Balearic Islands | 4.2 ±0.61 |
| 548 | MED. MORAY | *Muraena helena* | 535 | 40 | 0.68 | Balearic Islands | 4.2 ±0.61 |
| 549 | MED. MORAY | *Muraena helena* | 380 | 70 | 0.32 | Balearic Islands | 4.2 ±0.61 |
| 550 | MED. MORAY | *Muraena helena* | 225 | 100 | 0.33 | Balearic Islands | 4.2 ±0.61 |
| 551 | MED. MORAY | *Muraena helena* | 380 | 70 | 0.41 | Balearic Islands | 4.2 ±0.61 |
| 552 | MED. MORAY | *Muraena helena* | - | - | 1.11 | Balearic Islands | 4.2 ±0.61 |
| 553 | MED. MORAY | *Muraena helena* | 762 | 79 | 0.35 | Alghero | 4.2 ±0.61 |
| 554 | MED. MORAY | *Muraena helena* | 694 | 61 | 0.51 | Alghero | 4.2 ±0.61 |
| 555 | MED. MORAY | *Muraena helena* | 393 | 58 | 0.28 | Alghero | 4.2 ±0.61 |
| 556 | MED. MORAY | *Muraena helena* | 648 | 70 | 0.36 | Alghero | 4.2 ±0.61 |
| 557 | MED. MORAY | *Muraena helena* | 789 | 79 | 0.55 | Alghero | 4.2 ±0.61 |
| 558 | MED. MORAY | *Muraena helena* | 935 | 83 | 0.58 | Alghero | 4.2 ±0.61 |
| 559 | MED. MORAY | *Muraena helena* | 781 | 79 | 0.45 | Alghero | 4.2 ±0.61 |
| 560 | MUSSEL | *Mytilus galloprovincialis* | - | - | 0.07 | Balearic Islands | - |
| 561 | MUSSEL | *Mytilus galloprovincialis* | - | - | 0.07 | Balearic Islands | - |
| 562 | NORWAY LOBSTER | *Nephrops norvegicus* | - | - | 0.77 | Balearic Islands | - |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 563 | NORWAY LOBSTER | *Nephrops norvegicus* | - | - | 0.74 | Balearic Islands | - |
| 564 | NORWAY LOBSTER | *Nephrops norvegicus* | - | - | 0.74 | Balearic Islands | - |
| 565 | NORWAY LOBSTER | *Nephrops norvegicus* | - | - | 0.77 | Balearic Islands | - |
| 566 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | - | - | 0.21 | Balearic Islands | 4.2 ± 0.6 |
| 567 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | - | - | 0.21 | Balearic Islands | 4.2 ± 0.6 |
| 568 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | 260 | 16 | 0.11 | Balearic Islands | 4.2 ± 0.6 |
| 569 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | 300 | 18 | 0.30 | Balearic Islands | 4.2 ± 0.6 |
| 570 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | 103 | 18 | 0.18 | Marseille | 4.2 ± 0.6 |
| 571 | RED SEABREAM/BLACKSPOT SEABREAM | *Pagellus bogaroevo* | 101 | 18 | 0.30 | Marseille | 4.2 ± 0.6 |
| 572 | COMMON PANDORA | *Pagellus erythrinus* | 450 | 29 | 0.86 | Balearic Islands | 3.5 ± 0.1 |
| 573 | COMMON PANDORA | *Pagellus erythrinus* | 370 | 20 | 0.22 | Balearic Islands | 3.5 ± 0.1 |
| 574 | COMMON PANDORA | *Pagellus erythrinus* | 350 | 21 | 0.31 | Balearic Islands | 3.5 ± 0.1 |
| 575 | COMMON PANDORA | *Pagellus erythrinus* | 400 | 23 | 0.42 | Balearic Islands | 3.5 ± 0.1 |
| 576 | COMMON PANDORA | *Pagellus erythrinus* | 250 | 14 | 0.24 | Balearic Islands | 3.5 ± 0.1 |
| 577 | COMMON PANDORA | *Pagellus erythrinus* | 400 | 16 | 0.22 | Balearic Islands | 3.5 ± 0.1 |
| 578 | COMMON PANDORA | *Pagellus erythrinus* | 300 | 30 | 0.52 | Balearic Islands | 3.5 ± 0.1 |
| 579 | COMMON PANDORA | *Pagellus erythrinus* | 325 | 20 | 0.23 | Balearic Islands | 3.5 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 580 | COMMON PANDORA | *Pagellus erythrinus* | - | - | 0.16 | Balearic Islands | 3.5 ± 0.1 |
| 581 | COMMON PANDORA | *Pagellus erythrinus* | 350 | 22 | 0.22 | Balearic Islands | 3.5 ± 0.1 |
| 582 | COMMON PANDORA | *Pagellus erythrinus* | - | - | 0.23 | Balearic Islands | 3.5 ± 0.1 |
| 583 | COMMON PANDORA | *Pagellus erythrinus* | - | - | 0.16 | Balearic Islands | 3.5 ± 0.1 |
| 584 | COMMON PANDORA | *Pagellus erythrinus* | 87 | 19 | 0.36 | Alacant | 3.5 ± 0.1 |
| 585 | COMMON PANDORA | *Pagellus erythrinus* | 111 | 21 | 0.29 | Alacant | 3.5 ± 0.1 |
| 586 | COMMON PANDORA | *Pagellus erythrinus* | 105 | 21 | 0.40 | Alacant | 3.5 ± 0.1 |
| 587 | COMMON PANDORA | *Pagellus erythrinus* | 92 | 20.5 | 0.24 | Alacant | 3.5 ± 0.1 |
| 588 | COMMON PANDORA | *Pagellus erythrinus* | 91 | 20 | 0.17 | Alacant | 3.5 ± 0.1 |
| 589 | COMMON PANDORA | *Pagellus erythrinus* | 93 | 20 | 0.21 | Alacant | 3.5 ± 0.1 |
| 590 | COMMON PANDORA | *Pagellus erythrinus* | 121 | 21 | 0.10 | Alacant | 3.5 ± 0.1 |
| 591 | COMMON PANDORA | *Pagellus erythrinus* | 81 | 19 | 0.18 | Alacant | 3.5 ± 0.1 |
| 592 | COMMON PANDORA | *Pagellus erythrinus* | 114 | 20 | 0.44 | Alacant | 3.5 ± 0.1 |
| 593 | COMMON PANDORA | *Pagellus erythrinus* | 86 | 18 | 0.28 | Alacant | 3.5 ± 0.1 |
| 594 | COMMON PANDORA | *Pagellus erythrinus* | 96 | 19.5 | 0.14 | Alacant | 3.5 ± 0.1 |
| 595 | COMMON PANDORA | *Pagellus erythrinus* | 71 | 18 | 0.30 | Marseille | 3.5 ± 0.1 |
| 596 | AXILLARY SEABREAM | *Pagellus acarne* | - | - | 0.33 | Balearic Islands | 3.8 |
| 597 | AXILLARY SEABREAM | *Pagellus acarne* | 550 | 37 | 0.61 | Balearic Islands | 3.8 |
| 598 | AXILLARY SEABREAM | *Pagellus acarne* | 150 | 22 | 0.10 | Balearic Islands | 3.8 |
| 599 | AXILLARY SEABREAM | *Pagellus acarne* | 180 | 25.5 | 0.10 | Balearic Islands | 3.8 |
| 600 | AXILLARY SEABREAM | *Pagellus acarne* | 220 | - | 0.52 | Balearic Islands | 3.8 |
| 601 | AXILLARY SEABREAM | *Pagellus acarne* | 205 | - | 0.09 | Balearic Islands | 3.8 |
| 602 | AXILLARY SEABREAM | *Pagellus acarne* | 395 | 20 | 1.00 | Balearic Islands | 3.8 |
| 603 | AXILLARY SEABREAM | *Pagellus acarne* | 330 | 20 | 0.17 | Balearic Islands | 3.8 |
| 604 | AXILLARY SEABREAM | *Pagellus acarne* | 260 | 19 | 0.17 | Balearic Islands | 3.8 |
| 605 | AXILLARY SEABREAM | *Pagellus acarne* | 260 | 19 | 0.19 | Balearic Islands | 3.8 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 606 | AXILLARY SEABREAM | *Pagellus acarne* | 285 | 27 | 0.14 | Balearic Islands | 3.8 |
| 607 | AXILLARY SEABREAM | *Pagellus acarne* | - | - | 0.69 | Balearic Islands | 3.8 |
| 608 | AXILLARY SEABREAM | *Pagellus acarne* | - | - | 0.69 | Balearic Islands | 3.8 |
| 609 | AXILLARY SEABREAM | *Pagellus acarne* | 600 | - | 0.30 | Balearic Islands | 3.8 |
| 610 | AXILLARY SEABREAM | *Pagellus acarne* | 679 | 44 | 0.19 | Alacant | 3.8 |
| 611 | AXILLARY SEABREAM | *Pagellus acarne* | 176 | 24 | 0.71 | Marseille | 3.8 |
| 612 | GREATER FORKBEARD | *Phycis blennoides* | - | - | 1.36 | Balearic Islands | 3.7 ±0.66 |
| 613 | GREATER FORKBEARD | *Phycis blennoides* | - | - | 1.02 | Balearic Islands | 3.7 ±0.66 |
| 614 | GREATER FORKBEARD | *Phycis blennoides* | - | - | 1.02 | Balearic Islands | 3.7 ±0.66 |
| 615 | GREATER FORKBEARD | *Phycis blennoides* | - | - | 1.36 | Balearic Islands | 3.7 ±0.66 |
| 616 | GREATER FORKBEARD | *Phycis blennoides* | 80 | 22 | 0.14 | Alacant | 3.7 ±0.66 |
| 617 | GREATER FORKBEARD | *Phycis blennoides* | 125 | 23 | 0.10 | Alacant | 3.7 ±0.66 |
| 618 | GREATER FORKBEARD | *Phycis blennoides* | 128 | 28 | 0.07 | Alacant | 3.7 ±0.66 |
| 619 | GREATER FORKBEARD | *Phycis blennoides* | 89 | 22 | 0.18 | Alacant | 3.7 ±0.66 |
| 620 | THORNBACK RAY | *Raja clavata* | - | - | 2.12 | Balearic Islands | 3.8 ±0.2 |
| 621 | THORNBACK RAY | *Raja clavata* | - | - | 1.68 | Balearic Islands | 3.8 ±0.2 |
| 622 | THORNBACK RAY | *Raja clavata* | - | - | 1.68 | Balearic Islands | 3.8 ±0.2 |
| 623 | THORNBACK RAY | *Raja clavata* | - | - | 2.12 | Balearic Islands | 3.8 ±0.2 |
| 624 | THORNBACK RAY | *Raja clavata* | 766 | 52 | 0.70 | Ametlla de Mar - Ebro Delta | 3.8 ±0.2 |
| 625 | THORNBACK RAY | *Raja clavata* | 1291 | 61 | 1.76 | Ametlla de Mar - Ebro Delta | 3.8 ±0.2 |
| 626 | THORNBACK RAY | *Raja clavata* | 336 | 42 | 0.18 | L'Ampolla - Ebro Delta | 3.8 ±0.2 |
| 627 | SARDINE | *Sardina pilchardus* | 65 | 21 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 628 | SARDINE | *Sardina pilchardus* | 66 | 21 | 0.13 | Balearic Islands | 3.1 ± 0.1 |
| 629 | SARDINE | *Sardina pilchardus* | 84 | 22 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 630 | SARDINE | *Sardina pilchardus* | 110 | 23 | 0.15 | Balearic Islands | 3.1 ± 0.1 |
| 631 | SARDINE | *Sardina pilchardus* | 67 | 21 | 0.12 | Balearic Islands | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 632 | SARDINE | *Sardina pilchardus* | 55 | 20 | 0.13 | Balearic Islands | 3.1 ± 0.1 |
| 633 | SARDINE | *Sardina pilchardus* | 78 | 20 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 634 | SARDINE | *Sardina pilchardus* | 90 | 23 | 0.13 | Balearic Islands | 3.1 ± 0.1 |
| 635 | SARDINE | *Sardina pilchardus* | 88 | 22 | 0.13 | Balearic Islands | 3.1 ± 0.1 |
| 636 | SARDINE | *Sardina pilchardus* | 77 | 22 | 0.09 | Balearic Islands | 3.1 ± 0.1 |
| 637 | SARDINE | *Sardina pilchardus* | 82 | 21 | 0.12 | Balearic Islands | 3.1 ± 0.1 |
| 638 | SARDINE | *Sardina pilchardus* | 61 | 21 | 0.11 | Balearic Islands | 3.1 ± 0.1 |
| 639 | SARDINE | *Sardina pilchardus* | 67 | 21 | 0.13 | Balearic Islands | 3.1 ± 0.1 |
| 640 | SARDINE | *Sardina pilchardus* | 24 | 15 | 0.06 | Alghero | 3.1 ± 0.1 |
| 641 | SARDINE | *Sardina pilchardus* | 37 | 17 | 0.12 | Alghero | 3.1 ± 0.1 |
| 642 | SARDINE | *Sardina pilchardus* | 23 | 15 | 0.12 | Alghero | 3.1 ± 0.1 |
| 643 | SARDINE | *Sardina pilchardus* | 39 | 17 | 0.15 | Alghero | 3.1 ± 0.1 |
| 644 | SARDINE | *Sardina pilchardus* | 42 | 17 | 0.19 | Alghero | 3.1 ± 0.1 |
| 645 | SARDINE | *Sardina pilchardus* | 39 | 17 | 0.09 | Alghero | 3.1 ± 0.1 |
| 646 | SARDINE | *Sardina pilchardus* | 37 | 17 | 0.14 | Alghero | 3.1 ± 0.1 |
| 647 | SARDINE | *Sardina pilchardus* | 26 | 15 | 0.08 | Alghero | 3.1 ± 0.1 |
| 648 | SARDINE | *Sardina pilchardus* | 38 | 17 | 0.13 | Alghero | 3.1 ± 0.1 |
| 649 | SARDINE | *Sardina pilchardus* | 43 | 17 | 0.11 | Alghero | 3.1 ± 0.1 |
| 650 | SARDINE | *Sardina pilchardus* | 37 | 17 | 0.16 | Alacant | 3.1 ± 0.1 |
| 651 | SARDINE | *Sardina pilchardus* | 28 | 17 | 0.05 | Alacant | 3.1 ± 0.1 |
| 652 | SARDINE | *Sardina pilchardus* | 34 | 16 | 0.06 | Alacant | 3.1 ± 0.1 |
| 653 | SARDINE | *Sardina pilchardus* | 26 | 15 | 0.09 | Alacant | 3.1 ± 0.1 |
| 654 | SARDINE | *Sardina pilchardus* | 22 | 15 | 0.09 | Alacant | 3.1 ± 0.1 |
| 655 | SARDINE | *Sardina pilchardus* | 28 | 16 | 0.07 | Alacant | 3.1 ± 0.1 |
| 656 | SARDINE | *Sardina pilchardus* | 30 | 16 | 0.07 | Alacant | 3.1 ± 0.1 |
| 657 | SARDINE | *Sardina pilchardus* | 30 | 15.5 | 0.05 | Alacant | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 658 | SARDINE | *Sardina pilchardus* | 28 | 15.5 | 0.06 | Alacant | 3.1 ± 0.1 |
| 659 | SARDINE | *Sardina pilchardus* | 29 | 15.5 | 0.05 | Alacant | 3.1 ± 0.1 |
| 660 | SARDINE | *Sardina pilchardus* | 15 | 13 | 0.09 | Civitavecchia | 3.1 ± 0.1 |
| 661 | SARDINE | *Sardina pilchardus* | 15 | 14 | 0.07 | Civitavecchia | 3.1 ± 0.1 |
| 662 | SARDINE | *Sardina pilchardus* | 14 | 13 | 0.07 | Civitavecchia | 3.1 ± 0.1 |
| 663 | SARDINE | *Sardina pilchardus* | 15 | 12 | 0.08 | Civitavecchia | 3.1 ± 0.1 |
| 664 | SARDINE | *Sardina pilchardus* | 17 | 14 | 0.09 | Civitavecchia | 3.1 ± 0.1 |
| 665 | SARDINE | *Sardina pilchardus* | 15 | 15 | 0.07 | Civitavecchia | 3.1 ± 0.1 |
| 666 | SARDINE | *Sardina pilchardus* | 13 | 14 | 0.09 | Civitavecchia | 3.1 ± 0.1 |
| 667 | SARDINE | *Sardina pilchardus* | 16 | 14 | 0.06 | Civitavecchia | 3.1 ± 0.1 |
| 668 | SARDINE | *Sardina pilchardus* | 16 | 13 | 0.08 | Civitavecchia | 3.1 ± 0.1 |
| 669 | SARDINE | *Sardina pilchardus* | 15 | 13 | 0.07 | Civitavecchia | 3.1 ± 0.1 |
| 670 | SARDINE | *Sardina pilchardus* | 20 | 14 | 0.11 | Marseille | 3.1 ± 0.1 |
| 671 | SARDINE | *Sardina pilchardus* | 18 | 14 | 0.12 | Marseille | 3.1 ± 0.1 |
| 672 | SARDINE | *Sardina pilchardus* | 21 | 15 | 0.07 | Marseille | 3.1 ± 0.1 |
| 673 | SARDINE | *Sardina pilchardus* | 17 | 13 | 0.11 | Marseille | 3.1 ± 0.1 |
| 674 | SARDINE | *Sardina pilchardus* | 19 | 16 | 0.06 | Marseille | 3.1 ± 0.1 |
| 675 | SARDINE | *Sardina pilchardus* | 18 | 18 | 0.11 | Marseille | 3.1 ± 0.1 |
| 676 | SARDINE | *Sardina pilchardus* | 17 | 16 | 0.07 | Marseille | 3.1 ± 0.1 |
| 677 | SARDINE | *Sardina pilchardus* | 18 | 16 | 0.09 | Marseille | 3.1 ± 0.1 |
| 678 | SARDINE | *Sardina pilchardus* | 19 | 17 | 0.08 | Marseille | 3.1 ± 0.1 |
| 679 | SARDINE | *Sardina pilchardus* | 20 | 22 | 0.09 | Marseille | 3.1 ± 0.1 |
| 680 | SARDINE | *Sardina pilchardus* | 17 | 16 | 0.08 | Marseille | 3.1 ± 0.1 |
| 681 | SARDINE | *Sardina pilchardus* | 15 | 14 | 0.07 | Marseille | 3.1 ± 0.1 |
| 682 | SARDINE | *Sardina pilchardus* | 16 | 15 | 0.05 | Marseille | 3.1 ± 0.1 |
| 683 | SARDINE | *Sardina pilchardus* | 21 | 19 | 0.12 | Marseille | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 684 | SARDINE | *Sardina pilchardus* | 16 | 16 | 0.05 | Marseille | 3.1 ± 0.1 |
| 685 | SARDINE | *Sardina pilchardus* | 17 | 18 | 0.09 | Marseille | 3.1 ± 0.1 |
| 686 | SARDINE | *Sardina pilchardus* | 18 | 16 | 0.08 | Marseille | 3.1 ± 0.1 |
| 687 | SARDINE | *Sardina pilchardus* | 18 | 17 | 0.05 | Marseille | 3.1 ± 0.1 |
| 688 | SARDINE | *Sardina pilchardus* | 19 | 18 | 0.11 | Marseille | 3.1 ± 0.1 |
| 689 | SARDINE | *Sardina pilchardus* | 18 | 17 | 0.05 | Marseille | 3.1 ± 0.1 |
| 690 | SARDINE | *Sardina pilchardus* | 20 | 17 | 0.08 | Marseille | 3.1 ± 0.1 |
| 691 | SARDINE | *Sardina pilchardus* | 17 | 18 | 0.07 | Marseille | 3.1 ± 0.1 |
| 692 | SARDINE | *Sardina pilchardus* | 18 | 16 | 0.06 | Marseille | 3.1 ± 0.1 |
| 693 | SARDINE | *Sardina pilchardus* | 17 | 15 | 0.06 | Marseille | 3.1 ± 0.1 |
| 694 | SARDINE | *Sardina pilchardus* | 16 | 14 | 0.09 | Marseille | 3.1 ± 0.1 |
| 695 | SARDINE | *Sardina pilchardus* | 16 | 15 | 0.08 | Marseille | 3.1 ± 0.1 |
| 696 | SARDINE | *Sardina pilchardus* | 18 | 16 | 0.07 | Marseille | 3.1 ± 0.1 |
| 697 | SARDINE | *Sardina pilchardus* | 19 | 16 | 0.06 | Marseille | 3.1 ± 0.1 |
| 698 | SARDINE | *Sardina pilchardus* | 19 | 17 | 0.06 | Marseille | 3.1 ± 0.1 |
| 699 | SARDINE | *Sardina pilchardus* | 19 | 16 | 0.08 | Marseille | 3.1 ± 0.1 |
| 700 | SARDINE | *Sardina pilchardus* | 20 | 18 | 0.08 | Marseille | 3.1 ± 0.1 |
| 701 | SARDINE | *Sardina pilchardus* | 21 | 19 | 0.10 | Marseille | 3.1 ± 0.1 |
| 702 | SARDINE | *Sardina pilchardus* | 18 | 16 | 0.07 | Marseille | 3.1 ± 0.1 |
| 703 | SARDINE | *Sardina pilchardus* | 31 | 15 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 704 | SARDINE | *Sardina pilchardus* | 29 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 705 | SARDINE | *Sardina pilchardus* | 30 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 706 | SARDINE | *Sardina pilchardus* | 31 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 707 | SARDINE | *Sardina pilchardus* | 30 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 708 | SARDINE | *Sardina pilchardus* | 32 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 709 | SARDINE | *Sardina pilchardus* | 33 | 17 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 710 | SARDINE | *Sardina pilchardus* | 31 | 16 | 0.02 | L'Ampolla - Ebro Delta | 3.1 ± 0.1 |
| 711 | ROUND SARDINELLA | *Sardinella aurita* | 250 | 17 | 0.11 | Balearic Islands | 3.4 ±0.5 |
| 712 | BROWN MEAGRE | *Sciaena umbra* | 394 | 29 | 0.12 | Balearic Islands | 3.8 ±0.5 |
| 713 | BROWN MEAGRE | *Sciaena umbra* | 369 | 27 | 0.15 | Balearic Islands | 3.8 ±0.5 |
| 714 | BROWN MEAGRE | *Sciaena umbra* | 340 | 30 | 0.09 | Balearic Islands | 3.8 ±0.5 |
| 715 | BROWN MEAGRE | *Sciaena umbra* | 250 | 25 | 0.09 | Balearic Islands | 3.8 ±0.5 |
| 716 | BROWN MEAGRE | *Sciaena umbra* | 390 | 27 | 0.09 | Balearic Islands | 3.8 ±0.5 |
| 717 | BROWN MEAGRE | *Sciaena umbra* | - | - | 0.23 | Balearic Islands | 3.8 ±0.5 |
| 718 | BROWN MEAGRE | *Sciaena umbra* | 364 | 26 | 0.09 | Balearic Islands | 3.8 ±0.5 |
| 719 | BROWN MEAGRE | *Sciaena umbra* | 340 | 26 | 0.09 | Balearic Islands | 3.8 ±0.5 |
| 720 | BLACK SCORPIONFISH | *Scorpaena porcus* | - | - | 0.15 | Balearic Islands | 3.9 ±0.2 |
| 721 | BLACK SCORPIONFISH | *Scorpaena porcus* | - | - | 0.16 | Balearic Islands | 3.9 ±0.2 |
| 722 | BLACK SCORPIONFISH | *Scorpaena porcus* | 380 | 27 | 0.54 | Ametlla de Mar - Ebro Delta | 3.9 ±0.2 |
| 723 | BLACK SCORPIONFISH | *Scorpaena porcus* | 249 | 24 | 0.41 | Ametlla de Mar - Ebro Delta | 3.9 ±0.2 |
| 724 | BLACK SCORPIONFISH | *Scorpaena porcus* | 359 | 26 | 0.51 | Ametlla de Mar - Ebro Delta | 3.9 ±0.2 |
| 725 | SCORPIONFISH | *Scorpaena scrofa* | 250 | 25 | 0.12 | Balearic Islands | 4.3 ±0.5 |
| 726 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.15 | Balearic Islands | 4.3 ±0.5 |
| 727 | SCORPIONFISH | *Scorpaena scrofa* | 367 | 26 | 0.17 | Balearic Islands | 4.3 ±0.5 |
| 728 | SCORPIONFISH | *Scorpaena scrofa* | 286 | 21 | 0.09 | Balearic Islands | 4.3 ±0.5 |
| 729 | SCORPIONFISH | *Scorpaena scrofa* | 521 | 32 | 0.09 | Balearic Islands | 4.3 ±0.5 |
| 730 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.09 | Balearic Islands | 4.3 ±0.5 |
| 731 | SCORPIONFISH | *Scorpaena scrofa* | 500 | 29 | 0.14 | Balearic Islands | 4.3 ±0.5 |
| 732 | SCORPIONFISH | *Scorpaena scrofa* | 300 | 21 | 0.18 | Balearic Islands | 4.3 ±0.5 |
| 733 | SCORPIONFISH | *Scorpaena scrofa* | 250 | 21 | 0.18 | Balearic Islands | 4.3 ±0.5 |
| 734 | SCORPIONFISH | *Scorpaena scrofa* | 300 | 28 | 0.26 | Balearic Islands | 4.3 ±0.5 |
| 735 | SCORPIONFISH | *Scorpaena scrofa* | 300 | 25 | 0.21 | Balearic Islands | 4.3 ±0.5 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 736 | SCORPIONFISH | *Scorpaena scrofa* | 446 | 32 | 0.12 | Balearic Islands | 4.3 ±0.5 |
| 737 | SCORPIONFISH | *Scorpaena scrofa* | 450 | 28 | 0.10 | Balearic Islands | 4.3 ±0.5 |
| 738 | SCORPIONFISH | *Scorpaena scrofa* | 380 | 30 | 0.14 | Balearic Islands | 4.3 ±0.5 |
| 739 | SCORPIONFISH | *Scorpaena scrofa* | 400 | 27 | 0.29 | Balearic Islands | 4.3 ±0.5 |
| 740 | SCORPIONFISH | *Scorpaena scrofa* | 450 | 28 | 0.40 | Balearic Islands | 4.3 ±0.5 |
| 741 | SCORPIONFISH | *Scorpaena scrofa* | 420 | 29 | 0.14 | Balearic Islands | 4.3 ±0.5 |
| 742 | SCORPIONFISH | *Scorpaena scrofa* | 635 | - | 0.14 | Balearic Islands | 4.3 ±0.5 |
| 743 | SCORPIONFISH | *Scorpaena scrofa* | 340 | 23 | 0.48 | Balearic Islands | 4.3 ±0.5 |
| 744 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.05 | Balearic Islands | 4.3 ±0.5 |
| 745 | SCORPIONFISH | *Scorpaena scrofa* | 791 | 40 | 0.15 | Balearic Islands | 4.3 ±0.5 |
| 746 | SCORPIONFISH | *Scorpaena scrofa* | 500 | - | 0.33 | Balearic Islands | 4.3 ±0.5 |
| 747 | SCORPIONFISH | *Scorpaena scrofa* | 600 | - | 0.23 | Balearic Islands | 4.3 ±0.5 |
| 748 | SCORPIONFISH | *Scorpaena scrofa* | 340 | - | 0.33 | Balearic Islands | 4.3 ±0.5 |
| 749 | SCORPIONFISH | *Scorpaena scrofa* | 1000 | - | 0.42 | Balearic Islands | 4.3 ±0.5 |
| 750 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.42 | Balearic Islands | 4.3 ±0.5 |
| 751 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 1.39 | Balearic Islands | 4.3 ±0.5 |
| 752 | SCORPIONFISH | *Scorpaena scrofa* | 250 | 28 | 0.14 | Balearic Islands | 4.3 ±0.5 |
| 753 | SCORPIONFISH | *Scorpaena scrofa* | 250 | - | 0.15 | Balearic Islands | 4.3 ±0.5 |
| 754 | SCORPIONFISH | *Scorpaena scrofa* | 455 | 20 | 0.16 | Balearic Islands | 4.3 ±0.5 |
| 755 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.21 | Balearic Islands | 4.3 ±0.5 |
| 756 | SCORPIONFISH | *Scorpaena scrofa* | 1405 | - | 0.45 | Balearic Islands | 4.3 ±0.5 |
| 757 | SCORPIONFISH | *Scorpaena scrofa* | 610 | - | 0.58 | Balearic Islands | 4.3 ±0.5 |
| 758 | SCORPIONFISH | *Scorpaena scrofa* | 260 | - | 0.16 | Balearic Islands | 4.3 ±0.5 |
| 759 | SCORPIONFISH | *Scorpaena scrofa* | 410 | 25 | 0.23 | Balearic Islands | 4.3 ±0.5 |
| 760 | SCORPIONFISH | *Scorpaena scrofa* | 492 | 28 | 0.13 | Balearic Islands | 4.3 ±0.5 |
| 761 | SCORPIONFISH | *Scorpaena scrofa* | 440 | 26 | 0.22 | Balearic Islands | 4.3 ±0.5 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 762 | SCORPIONFISH | *Scorpaena scrofa* | 250 | 17 | 0.18 | Balearic Islands | 4.3 ±0.5 |
| 763 | SCORPIONFISH | *Scorpaena scrofa* | 330 | 20 | 0.27 | Balearic Islands | 4.3 ±0.5 |
| 764 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 1.39 | Balearic Islands | 4.3 ±0.5 |
| 765 | SCORPIONFISH | *Scorpaena scrofa* | - | - | 0.42 | Balearic Islands | 4.3 ±0.5 |
| 766 | SCORPIONFISH | *Scorpaena scrofa* | 70 | 15 | 0.61 | Alghero | 4.3 ±0.5 |
| 767 | SCORPIONFISH | *Scorpaena scrofa* | 175 | 21 | 0.10 | Alghero | 4.3 ±0.5 |
| 768 | SCORPIONFISH | *Scorpaena scrofa* | 149 | 19 | 0.27 | Alghero | 4.3 ±0.5 |
| 769 | SCORPIONFISH | *Scorpaena scrofa* | 115 | 18 | 0.24 | Alghero | 4.3 ±0.5 |
| 770 | SCORPIONFISH | *Scorpaena scrofa* | 225 | 22 | 0.28 | Alghero | 4.3 ±0.5 |
| 771 | SCORPIONFISH | *Scorpaena scrofa* | 166 | 21 | 0.25 | Alghero | 4.3 ±0.5 |
| 772 | SCORPIONFISH | *Scorpaena scrofa* | 187 | 21 | 0.29 | Alghero | 4.3 ±0.5 |
| 773 | SCORPIONFISH | *Scorpaena scrofa* | 206 | 21 | 0.16 | Alghero | 4.3 ±0.5 |
| 774 | SCORPIONFISH | *Scorpaena scrofa* | 216 | 22 | 0.41 | Alghero | 4.3 ±0.5 |
| 775 | SCORPIONFISH | *Scorpaena scrofa* | 219 | 22 | 0.24 | Alghero | 4.3 ±0.5 |
| 776 | SCORPIONFISH | *Scorpaena scrofa* | 745 | 34 | 0.13 | Alacant | 4.3 ±0.5 |
| 777 | SCORPIONFISH | *Scorpaena scrofa* | 1031 | 31 | 0.22 | Alacant | 4.3 ±0.5 |
| 778 | SCORPIONFISH | *Scorpaena scrofa* | 836 | 38 | 0.12 | Alacant | 4.3 ±0.5 |
| 779 | SCORPIONFISH | *Scorpaena scrofa* | 97 | 17 | 0.47 | Alacant | 4.3 ±0.5 |
| 780 | SCORPIONFISH | *Scorpaena scrofa* | 61 | 14 | 0.26 | Alacant | 4.3 ±0.5 |
| 781 | SCORPIONFISH | *Scorpaena scrofa* | 63 | 15 | 0.13 | Alacant | 4.3 ±0.5 |
| 782 | SCORPIONFISH | *Scorpaena scrofa* | 1096 | 31 | 0.25 | Alacant | 4.3 ±0.5 |
| 783 | SCORPIONFISH | *Scorpaena scrofa* | 930 | 31 | 0.23 | Alacant | 4.3 ±0.5 |
| 784 | SCORPIONFISH | *Scorpaena scrofa* | 927 | 30 | 0.12 | Alacant | 4.3 ±0.5 |
| 785 | SCORPIONFISH | *Scorpaena scrofa* | 671 | 26 | 0.39 | Alacant | 4.3 ±0.5 |
| 786 | SCORPIONFISH | *Scorpaena scrofa* | 62 | 14 | 0.65 | Alacant | 4.3 ±0.5 |
| 787 | SCORPIONFISH | *Scorpaena scrofa* | 63 | 14 | 0.36 | Alacant | 4.3 ±0.5 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 788 | SCORPIONFISH | *Scorpaena scrofa* | 50 | 13 | 0.87 | Alacant | 4.3 ±0.5 |
| 789 | SCORPIONFISH | *Scorpaena scrofa* | 51 | 13 | 0.07 | Alacant | 4.3 ±0.5 |
| 790 | SCORPIONFISH | *Scorpaena scrofa* | 75 | 17 | 0.50 | Civitavecchia | 4.3 ±0.5 |
| 791 | SCORPIONFISH | *Scorpaena scrofa* | 228 | 24 | 0.46 | Civitavecchia | 4.3 ±0.5 |
| 792 | SCORPIONFISH | *Scorpaena scrofa* | 147 | 22 | 0.88 | Civitavecchia | 4.3 ±0.5 |
| 793 | SCORPIONFISH | *Scorpaena scrofa* | 73 | 17 | 0.39 | Civitavecchia | 4.3 ±0.5 |
| 794 | SCORPIONFISH | *Scorpaena scrofa* | 172 | 22 | 0.84 | Civitavecchia | 4.3 ±0.5 |
| 795 | SCORPIONFISH | *Scorpaena scrofa* | 107 | 19 | 0.71 | Civitavecchia | 4.3 ±0.5 |
| 796 | SCORPIONFISH | *Scorpaena scrofa* | 111 | 20 | 0.91 | Civitavecchia | 4.3 ±0.5 |
| 797 | SCORPIONFISH | *Scorpaena scrofa* | 102 | 19 | 0.62 | Civitavecchia | 4.3 ±0.5 |
| 798 | SCORPIONFISH | *Scorpaena scrofa* | 88 | 21 | 0.48 | Civitavecchia | 4.3 ±0.5 |
| 799 | SCORPIONFISH | *Scorpaena scrofa* | 93 | 18 | 0.39 | Civitavecchia | 4.3 ±0.5 |
| 800 | SCORPIONFISH | *Scorpaena scrofa* | 85 | 20 | 0.80 | Civitavecchia | 4.3 ±0.5 |
| 801 | SCORPIONFISH | *Scorpaena scrofa* | 71 | 17 | 0.65 | Civitavecchia | 4.3 ±0.5 |
| 802 | SCORPIONFISH | *Scorpaena scrofa* | 95 | 18 | 0.22 | Civitavecchia | 4.3 ±0.5 |
| 803 | SCORPIONFISH | *Scorpaena scrofa* | 84 | 17 | 0.53 | Civitavecchia | 4.3 ±0.5 |
| 804 | SCORPIONFISH | *Scorpaena scrofa* | 45 | 13 | 0.33 | Civitavecchia | 4.3 ±0.5 |
| 805 | SCORPIONFISH | *Scorpaena scrofa* | 306 | 35 | 0.21 | Genoa | 4.3 ±0.5 |
| 806 | SCORPIONFISH | *Scorpaena scrofa* | 365 | 36 | 0.30 | Genoa | 4.3 ±0.5 |
| 807 | SCORPIONFISH | *Scorpaena scrofa* | 180 | 21 | 0.32 | Genoa | 4.3 ±0.5 |
| 808 | SCORPIONFISH | *Scorpaena scrofa* | 109 | 20 | 0.38 | Genoa | 4.3 ±0.5 |
| 809 | SCORPIONFISH | *Scorpaena scrofa* | 105 | 18 | 0.34 | Genoa | 4.3 ±0.5 |
| 810 | SCORPIONFISH | *Scorpaena scrofa* | 126 | 20 | 0.31 | Genoa | 4.3 ±0.5 |
| 811 | SCORPIONFISH | *Scorpaena scrofa* | 103 | 19 | 0.28 | Genoa | 4.3 ±0.5 |
| 812 | SCORPIONFISH | *Scorpaena scrofa* | 126 | 19 | 0.51 | Genoa | 4.3 ±0.5 |
| 813 | SCORPIONFISH | *Scorpaena scrofa* | 180 | 22 | 0.42 | Genoa | 4.3 ±0.5 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 814 | SCORPIONFISH | *Scorpaena scrofa* | 134 | 19 | 0.48 | Genoa | 4.3 ±0.5 |
| 815 | SCORPIONFISH | *Scorpaena scrofa* | 213 | 22 | 0.41 | Genoa | 4.3 ±0.5 |
| 816 | SCORPIONFISH | *Scorpaena scrofa* | 197 | 23 | 0.20 | Genoa | 4.3 ±0.5 |
| 817 | SCORPIONFISH | *Scorpaena scrofa* | 47 | 14 | 0.32 | Marseille | 4.3 ±0.5 |
| 818 | SCORPIONFISH | *Scorpaena scrofa* | 52 | 14 | 0.20 | Marseille | 4.3 ±0.5 |
| 819 | SCORPIONFISH | *Scorpaena scrofa* | 48 | 14 | 0.34 | Marseille | 4.3 ±0.5 |
| 820 | SCORPIONFISH | *Scorpaena scrofa* | 50 | 15 | 0.37 | Marseille | 4.3 ±0.5 |
| 821 | SCORPIONFISH | *Scorpaena scrofa* | 50 | 15 | 0.30 | Marseille | 4.3 ±0.5 |
| 822 | SCORPIONFISH | *Scorpaena scrofa* | 53 | 15 | 0.26 | Marseille | 4.3 ±0.5 |
| 823 | SCORPIONFISH | *Scorpaena scrofa* | 52 | 16 | 0.51 | Marseille | 4.3 ±0.5 |
| 824 | SCORPIONFISH | *Scorpaena scrofa* | 47 | 15 | 0.36 | Marseille | 4.3 ±0.5 |
| 825 | SCORPIONFISH | *Scorpaena scrofa* | 46 | 15 | 0.35 | Marseille | 4.3 ±0.5 |
| 826 | SCORPIONFISH | *Scorpaena scrofa* | 266 | 24 | 0.29 | Marseille | 4.3 ±0.5 |
| 827 | SCORPIONFISH | *Scorpaena scrofa* | 326 | 28 | 0.25 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 828 | SCORPIONFISH | *Scorpaena scrofa* | 56 | 14 | 0.85 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 829 | SCORPIONFISH | *Scorpaena scrofa* | 25 | 11 | 0.27 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 830 | SCORPIONFISH | *Scorpaena scrofa* | 37 | 13 | 0.44 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 831 | SCORPIONFISH | *Scorpaena scrofa* | 21 | 11 | 0.32 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 832 | SCORPIONFISH | *Scorpaena scrofa* | 16 | 9 | 0.21 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 833 | SCORPIONFISH | *Scorpaena scrofa* | 15 | 9 | 0.21 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 834 | SCORPIONFISH | *Scorpaena scrofa* | 14 | 9 | 0.48 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 835 | SCORPIONFISH | *Scorpaena scrofa* | 17 | 10 | 0.15 | Ametlla de Mar - Ebro Delta | 4.3 ±0.5 |
| 836 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 300 | 43.5 | 0.51 | Balearic Islands | 3.8 ±0.3 |
| 837 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 300 | 43 | 0.87 | Balearic Islands | 3.8 ±0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 838 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 300 | 45 | 1.10 | Balearic Islands | 3.8 ±0.3 |
| 839 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 860 | 42 | 0.39 | Balearic Islands | 3.8 ±0.3 |
| 840 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 920 | 43 | 0.48 | Balearic Islands | 3.8 ±0.3 |
| 841 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 1400 | 40 | 1.50 | Balearic Islands | 3.8 ±0.3 |
| 842 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | - | - | 1.07 | Balearic Islands | 3.8 ±0.3 |
| 843 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | - | - | 3.77 | Balearic Islands | 3.8 ±0.3 |
| 844 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 265 | 29 | 0.58 | Balearic Islands | 3.8 ±0.3 |
| 845 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 350 | 30 | 0.78 | Balearic Islands | 3.8 ±0.3 |
| 846 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | - | - | 3.77 | Balearic Islands | 3.8 ±0.3 |
| 847 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | - | - | 1.07 | Balearic Islands | 3.8 ±0.3 |
| 848 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 54 | 24 | 0.06 | Alacant | 3.8 ±0.3 |
| 849 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 143 | 31 | 0.04 | Alacant | 3.8 ±0.3 |
| 850 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 58 | 25 | 0.04 | Alacant | 3.8 ±0.3 |
| 851 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 100 | 25 | 0.05 | Alacant | 3.8 ±0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 852 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 93 | 28 | 0.03 | Alacant | 3.8 ±0.3 |
| 853 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 148 | 29 | 0.07 | Alacant | 3.8 ±0.3 |
| 854 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 81 | 25 | 0.08 | Alacant | 3.8 ±0.3 |
| 855 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 133 | 30 | 0.04 | Alacant | 3.8 ±0.3 |
| 856 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 79 | 25 | 0.06 | Alacant | 3.8 ±0.3 |
| 857 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 105 | 27 | 0.04 | Alacant | 3.8 ±0.3 |
| 858 | SMALL-SPOTTED CATSHARK | *Scyliorhinus canicula* | 67 | 24.5 | 0.05 | Alacant | 3.8 ±0.3 |
| 859 | GREATER AMBERJACK | *Seriola dumerili* | 710 | 39 | 0.20 | Balearic Islands | 4.5 |
| 860 | GREATER AMBERJACK | *Seriola dumerili* | 630 | 40 | 0.13 | Balearic Islands | 4.5 |
| 861 | GREATER AMBERJACK | *Seriola dumerili* | 330 | 20 | 0.09 | Balearic Islands | 4.5 |
| 862 | GREATER AMBERJACK | *Seriola dumerili* | 1700 | - | 0.54 | Balearic Islands | 4.5 |
| 863 | GREATER AMBERJACK | *Seriola dumerili* | 25 | 20 | 0.09 | Balearic Islands | 4.5 |
| 864 | GREATER AMBERJACK | *Seriola dumerili* | 300 | 36 | 0.09 | Balearic Islands | 4.5 |
| 865 | GREATER AMBERJACK | *Seriola dumerili* | 2840 | - | 1.90 | Balearic Islands | 4.5 |
| 866 | GREATER AMBERJACK | *Seriola dumerili* | 265 | 40 | 0.09 | Balearic Islands | 4.5 |
| 867 | GREATER AMBERJACK | *Seriola dumerili* | 450 | 33 | 0.05 | Balearic Islands | 4.5 |
| 868 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 1.90 | Balearic Islands | 4.5 |
| 869 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.05 | Balearic Islands | 4.5 |
| 870 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.05 | Balearic Islands | 4.5 |
| 871 | GREATER AMBERJACK | *Seriola dumerili* | 1200 | 31 | 0.10 | Balearic Islands | 4.5 |
| 872 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.13 | Balearic Islands | 4.5 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 873 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.10 | Balearic Islands | 4.5 |
| 874 | GREATER AMBERJACK | *Seriola dumerili* | 700 | - | 0.01 | Balearic Islands | 4.5 |
| 875 | GREATER AMBERJACK | *Seriola dumerili* | 700 | - | 0.10 | Balearic Islands | 4.5 |
| 876 | GREATER AMBERJACK | *Seriola dumerili* | 775 | - | 0.01 | Balearic Islands | 4.5 |
| 877 | GREATER AMBERJACK | *Seriola dumerili* | 410 | 45 | 0.16 | Balearic Islands | 4.5 |
| 878 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.11 | Balearic Islands | 4.5 |
| 879 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.09 | Balearic Islands | 4.5 |
| 880 | GREATER AMBERJACK | *Seriola dumerili* | - | - | 0.09 | Balearic Islands | 4.5 |
| 881 | COMBER | *Serranus cabrilla* | 300 | 22 | 0.46 | Balearic Islands | 3.4 ±0.3 |
| 882 | COMBER | *Serranus cabrilla* | 300 | 22 | 0.50 | Balearic Islands | 3.4 ±0.3 |
| 883 | COMBER | *Serranus cabrilla* | 300 | 22 | 0.53 | Balearic Islands | 3.4 ±0.3 |
| 884 | COMBER | *Serranus cabrilla* | 360 | 15 | 0.16 | Balearic Islands | 3.4 ±0.3 |
| 885 | COMBER | *Serranus cabrilla* | 386 | 16 | 0.10 | Balearic Islands | 3.4 ±0.3 |
| 886 | COMBER | *Serranus cabrilla* | - | - | 0.16 | Balearic Islands | 3.4 ±0.3 |
| 887 | COMBER | *Serranus cabrilla* | - | 15 | 0.48 | Balearic Islands | 3.4 ±0.3 |
| 888 | COMBER | *Serranus cabrilla* | 220 | 13.5 | 0.09 | Balearic Islands | 3.4 ±0.3 |
| 889 | COMBER | *Serranus cabrilla* | 315 | 15 | 0.18 | Balearic Islands | 3.4 ±0.3 |
| 890 | COMBER | *Serranus cabrilla* | 310 | 18 | 0.50 | Balearic Islands | 3.4 ±0.3 |
| 891 | COMBER | *Serranus cabrilla* | 270 | 17 | 0.18 | Balearic Islands | 3.4 ±0.3 |
| 892 | COMBER | *Serranus cabrilla* | 51 | 17 | 0.18 | Alacant | 3.4 ±0.3 |
| 893 | COMBER | *Serranus cabrilla* | 58 | 18 | 0.12 | Alacant | 3.4 ±0.3 |
| 894 | COMBER | *Serranus cabrilla* | 55 | 16.5 | 0.09 | Alacant | 3.4 ±0.3 |
| 895 | COMBER | *Serranus cabrilla* | 65 | 18.5 | 0.15 | Alacant | 3.4 ±0.3 |
| 896 | COMBER | *Serranus cabrilla* | 84 | 20 | 2.02 | Civitavecchia | 3.4 ±0.3 |
| 897 | COMBER | *Serranus cabrilla* | 77 | 20 | 0.65 | Civitavecchia | 3.4 ±0.3 |
| 898 | COMBER | *Serranus cabrilla* | 65 | 17 | 0.41 | Civitavecchia | 3.4 ±0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 899 | PAINTED COMBER | *Serranus scriba* | - | - | 0.20 | Balearic Islands | 3.8 ±0.3 |
| 900 | PAINTED COMBER | *Serranus scriba* | 440 | 13 | 0.24 | Balearic Islands | 3.8 ±0.3 |
| 901 | PAINTED COMBER | *Serranus scriba* | 274 | 15 | 0.26 | Balearic Islands | 3.8 ±0.3 |
| 902 | PAINTED COMBER | *Serranus scriba* | - | 10 | 0.30 | Balearic Islands | 3.8 ±0.3 |
| 903 | PAINTED COMBER | *Serranus scriba* | 460 | - | 0.46 | Balearic Islands | 3.8 ±0.3 |
| 904 | PAINTED COMBER | *Serranus scriba* | - | - | 0.27 | Balearic Islands | 3.8 ±0.3 |
| 905 | PAINTED COMBER | *Serranus scriba* | 250 | 14 | 0.25 | Balearic Islands | 3.8 ±0.3 |
| 906 | PAINTED COMBER | *Serranus scriba* | 300 | 16 | 0.27 | Balearic Islands | 3.8 ±0.3 |
| 907 | PAINTED COMBER | *Serranus scriba* | 81 | 20 | 0.44 | Alacant | 3.8 ±0.3 |
| 908 | PAINTED COMBER | *Serranus scriba* | 117 | 21 | 0.12 | Alacant | 3.8 ±0.3 |
| 909 | PAINTED COMBER | *Serranus scriba* | 81 | 20 | 0.33 | Alacant | 3.8 ±0.3 |
| 910 | SOLE | *Solea solea* | 450 | 29 | 0.14 | Balearic Islands | 3.2 ± 0.1 |
| 911 | SOLE | *Solea solea* | 225 | 24 | 0.46 | Balearic Islands | 3.2 ± 0.1 |
| 912 | SOLE | *Solea solea* | 350 | 24 | 0.69 | Balearic Islands | 3.2 ± 0.1 |
| 913 | SOLE | *Solea solea* | 440 | 32 | 0.34 | Balearic Islands | 3.2 ± 0.1 |
| 914 | SOLE | *Solea solea* | 360 | 26 | 0.09 | Balearic Islands | 3.2 ± 0.1 |
| 915 | SOLE | *Solea solea* | 380 | 28 | 0.09 | Balearic Islands | 3.2 ± 0.1 |
| 916 | SOLE | *Solea solea* | 300 | 23 | 0.66 | Balearic Islands | 3.2 ± 0.1 |
| 917 | SOLE | *Solea solea* | 305 | 23 | 1.20 | Balearic Islands | 3.2 ± 0.1 |
| 918 | SOLE | *Solea solea* | 251 | 31 | 0.21 | Balearic Islands | 3.2 ± 0.1 |
| 919 | SOLE | *Solea solea* | 130 | 22 | 0.12 | Balearic Islands | 3.2 ± 0.1 |
| 920 | SOLE | *Solea solea* | 308 | 33 | 0.07 | Balearic Islands | 3.2 ± 0.1 |
| 921 | SOLE | *Solea solea* | 354 | 34 | 0.97 | Balearic Islands | 3.2 ± 0.1 |
| 922 | SOLE | *Solea solea* | 244 | 30 | 0.28 | Balearic Islands | 3.2 ± 0.1 |
| 923 | SOLE | *Solea solea* | 296 | 32 | 1.52 | Balearic Islands | 3.2 ± 0.1 |
| 924 | SOLE | *Solea solea* | 301 | 32 | 0.66 | Balearic Islands | 3.2 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 925 | SOLE | *Solea solea* | 279 | 31 | 0.82 | Balearic Islands | 3.2 ± 0.1 |
| 926 | SOLE | *Solea solea* | 327 | 34 | 0.76 | Balearic Islands | 3.2 ± 0.1 |
| 927 | SOLE | *Solea solea* | 300 | 32 | 0.13 | Balearic Islands | 3.2 ± 0.1 |
| 928 | SOLE | *Solea solea* | 260 | 32 | 0.05 | Civitavecchia | 3.2 ± 0.1 |
| 929 | SOLE | *Solea solea* | 268 | 31 | 0.04 | Civitavecchia | 3.2 ± 0.1 |
| 930 | SOLE | *Solea solea* | 259 | 30 | 0.07 | Civitavecchia | 3.2 ± 0.1 |
| 931 | SOLE | *Solea solea* | 313 | 32 | 0.06 | Civitavecchia | 3.2 ± 0.1 |
| 932 | SOLE | *Solea solea* | 309 | 31 | 0.05 | Civitavecchia | 3.2 ± 0.1 |
| 933 | SOLE | *Solea solea* | 297 | 31 | 0.04 | Civitavecchia | 3.2 ± 0.1 |
| 934 | SOLE | *Solea solea* | 319 | 33 | 0.05 | Civitavecchia | 3.2 ± 0.1 |
| 935 | SOLE | *Solea solea* | 294 | 34 | 0.06 | Civitavecchia | 3.2 ± 0.1 |
| 936 | SOLE | *Solea solea* | 264 | 31 | 0.06 | Civitavecchia | 3.2 ± 0.1 |
| 937 | SOLE | *Solea solea* | 363 | 35 | 1.81 | Civitavecchia | 3.2 ± 0.1 |
| 938 | SOLE | *Solea solea* | 210 | 30 | 0.21 | Civitavecchia | 3.2 ± 0.1 |
| 939 | SOLE | *Solea solea* | 204 | 29 | 0.67 | Civitavecchia | 3.2 ± 0.1 |
| 940 | SOLE | *Solea solea* | 204 | 31 | 0.18 | Civitavecchia | 3.2 ± 0.1 |
| 941 | SOLE | *Solea solea* | 318 | 32 | 0.20 | Civitavecchia | 3.2 ± 0.1 |
| 942 | SOLE | *Solea solea* | 206 | 30 | 0.46 | Civitavecchia | 3.2 ± 0.1 |
| 943 | SOLE | *Solea solea* | 248 | 31 | 1.08 | Civitavecchia | 3.2 ± 0.1 |
| 944 | SOLE | *Solea solea* | 357 | 35 | 1.18 | Civitavecchia | 3.2 ± 0.1 |
| 945 | SOLE | *Solea solea* | 191 | 28 | 0.31 | Civitavecchia | 3.2 ± 0.1 |
| 946 | SOLE | *Solea solea* | 247 | 33 | 0.11 | Genoa | 3.2 ± 0.1 |
| 947 | SOLE | *Solea solea* | 223 | 31 | 0.05 | Genoa | 3.2 ± 0.1 |
| 948 | SOLE | *Solea solea* | 145 | 27 | 0.06 | Genoa | 3.2 ± 0.1 |
| 949 | SOLE | *Solea solea* | 172 | 30 | 0.15 | Genoa | 3.2 ± 0.1 |
| 950 | SOLE | *Solea solea* | 152 | 29 | 0.04 | Genoa | 3.2 ± 0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 951 | SOLE | *Solea solea* | 186 | 30 | 0.03 | Genoa | 3.2 ± 0.1 |
| 952 | SOLE | *Solea solea* | 224 | 33 | 0.07 | Genoa | 3.2 ± 0.1 |
| 953 | SOLE | *Solea solea* | 192 | 30 | 0.05 | Genoa | 3.2 ± 0.1 |
| 954 | SOLE | *Solea solea* | 197 | 30 | 0.04 | Marseille | 3.2 ± 0.1 |
| 955 | SOLE | *Solea solea* | 178 | 28 | 0.25 | Marseille | 3.2 ± 0.1 |
| 956 | SOLE | *Solea solea* | 184 | 30 | 0.07 | Marseille | 3.2 ± 0.1 |
| 957 | SOLE | *Solea solea* | 167 | 30 | 0.75 | Marseille | 3.2 ± 0.1 |
| 958 | SOLE | *Solea solea* | 194 | 30 | 0.07 | Marseille | 3.2 ± 0.1 |
| 959 | SOLE | *Solea solea* | 167 | 28 | 0.07 | Marseille | 3.2 ± 0.1 |
| 960 | SOLE | *Solea solea* | 177 | 30 | 0.23 | Marseille | 3.2 ± 0.1 |
| 961 | SOLE | *Solea solea* | 180 | 29 | 0.12 | Marseille | 3.2 ± 0.1 |
| 962 | SOLE | *Solea solea* | 138 | 27 | 0.02 | Marseille | 3.2 ± 0.1 |
| 963 | SOLE | *Solea solea* | 138 | 26 | 0.02 | Marseille | 3.2 ± 0.1 |
| 964 | SOLE | *Solea solea* | 244 | 30 | 0.01 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 965 | SOLE | *Solea solea* | 229 | 31 | 0.04 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 966 | SOLE | *Solea solea* | 178 | 29 | 0.08 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 967 | SOLE | *Solea solea* | 197 | 30 | 0.10 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 968 | SOLE | *Solea solea* | 228 | 30 | 0.06 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 969 | SOLE | *Solea solea* | 188 | 20 | 0.05 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 970 | SOLE | *Solea solea* | 202 | 29 | 0.05 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 971 | SOLE | *Solea solea* | 250 | 30 | 0.12 | L'Ampolla - Ebro Delta | 3.2 ± 0.1 |
| 972 | EUROPEAN BARRACUDA | *Sphyraena sphyraena* | 385 | 100 | 1.00 | Balearic Islands | 4.0 ±0.51 |
| 973 | PICAREL | *Spicara smaris* | 620 | 14 | 0.09 | Balearic Islands | 3.0 |
| 974 | PICAREL | *Spicara smaris* | 900 | 14 | 0.09 | Balearic Islands | 3.0 |
| 975 | PICAREL | *Spicara smaris* | 880 | - | 0.09 | Balearic Islands | 3.0 |
| 976 | PICAREL | *Spicara smaris* | 250 | 15 | 0.09 | Balearic Islands | 3.0 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 977 | PICAREL | *Spicara smaris* | 300 | 15 | 0.09 | Balearic Islands | 3.0 |
| 978 | PICAREL | *Spicara smaris* | 280 | 16 | 0.15 | Balearic Islands | 3.0 |
| 979 | PICAREL | *Spicara smaris* | 280 | 16 | 0.09 | Balearic Islands | 3.0 |
| 980 | BLACK SEABREAM | *Spondyliosoma cantharus* | 454 | 30 | 0.15 | Balearic Islands | 3.3 ±0.2 |
| 981 | BLACK SEABREAM | *Spondyliosoma cantharus* | 448 | 31 | 0.16 | Balearic Islands | 3.3 ±0.2 |
| 982 | BLACK SEABREAM | *Spondyliosoma cantharus* | 568 | 30 | 0.19 | Balearic Islands | 3.3 ±0.2 |
| 983 | BLACK SEABREAM | *Spondyliosoma cantharus* | 709 | 32 | 0.79 | Balearic Islands | 3.3 ±0.2 |
| 984 | BLACK SEABREAM | *Spondyliosoma cantharus* | 650 | 35 | 0.25 | Balearic Islands | 3.3 ±0.2 |
| 985 | BLACK SEABREAM | *Spondyliosoma cantharus* | 250 | 24.5 | 0.12 | Balearic Islands | 3.3 ±0.2 |
| 986 | BLACK SEABREAM | *Spondyliosoma cantharus* | 350 | 22 | 0.22 | Balearic Islands | 3.3 ±0.2 |
| 987 | BLACK SEABREAM | *Spondyliosoma cantharus* | 238 | 20 | 0.10 | Balearic Islands | 3.3 ±0.2 |
| 988 | BLACK SEABREAM | *Spondyliosoma cantharus* | 300 | 20 | 0.19 | Balearic Islands | 3.3 ±0.2 |
| 989 | BLACK SEABREAM | *Spondyliosoma cantharus* | 145 | 19 | 0.09 | Balearic Islands | 3.3 ±0.2 |
| 990 | BLACK SEABREAM | *Spondyliosoma cantharus* | 120 | 20 | 0.09 | Balearic Islands | 3.3 ±0.2 |
| 991 | BLACK SEABREAM | *Spondyliosoma cantharus* | 165 | 20 | 0.09 | Balearic Islands | 3.3 ±0.2 |
| 992 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.98 | Balearic Islands | 3.3 ±0.2 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 993 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.76 | Balearic Islands | 3.3 ±0.2 |
| 994 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.11 | Balearic Islands | 3.3 ±0.2 |
| 995 | BLACK SEABREAM | *Spondyliosoma cantharus* | 375 | 40 | 0.13 | Balearic Islands | 3.3 ±0.2 |
| 996 | BLACK SEABREAM | *Spondyliosoma cantharus* | 400 | 32 | 0.15 | Balearic Islands | 3.3 ±0.2 |
| 997 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.11 | Balearic Islands | 3.3 ±0.2 |
| 998 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.76 | Balearic Islands | 3.3 ±0.2 |
| 999 | BLACK SEABREAM | *Spondyliosoma cantharus* | - | - | 0.98 | Balearic Islands | 3.3 ±0.2 |
| 1000 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 300 | 22 | 0.19 | Balearic Islands | 3.8 ± 0.3 |
| 1001 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 300 | 26 | 0.35 | Balearic Islands | 3.8 ± 0.3 |
| 1002 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 300 | 23 | 0.40 | Balearic Islands | 3.8 ± 0.3 |
| 1003 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 285 | 18 | 0.10 | Balearic Islands | 3.8 ± 0.3 |
| 1004 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 350 | 11 | 0.09 | Balearic Islands | 3.8 ± 0.3 |
| 1005 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 340 | - | 0.09 | Balearic Islands | 3.8 ± 0.3 |
| 1006 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 380 | - | 0.10 | Balearic Islands | 3.8 ± 0.3 |
| 1007 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 90 | 21 | 0.22 | Balearic Islands | 3.8 ± 0.3 |
| 1008 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 120 | 26 | 0.30 | Balearic Islands | 3.8 ± 0.3 |
| 1009 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 71 | 20 | 0.13 | Balearic Islands | 3.8 ± 0.3 |
| 1010 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 82 | 22 | 0.19 | Balearic Islands | 3.8 ± 0.3 |
| 1011 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 83 | 22 | 0.23 | Balearic Islands | 3.8 ± 0.3 |
| 1012 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 92 | 23 | 0.21 | Balearic Islands | 3.8 ± 0.3 |
| 1013 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 58 | 19 | 0.13 | Balearic Islands | 3.8 ± 0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1014 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 54 | 20 | 0.14 | Balearic Islands | 3.8 ± 0.3 |
| 1015 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 78 | 21 | 0.26 | Balearic Islands | 3.8 ± 0.3 |
| 1016 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 117 | 26 | 0.34 | Balearic Islands | 3.8 ± 0.3 |
| 1017 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 94 | 24 | 0.45 | Alghero | 3.8 ± 0.3 |
| 1018 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 75 | 22 | 0.34 | Alghero | 3.8 ± 0.3 |
| 1019 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 154 | 27 | 0.63 | Alghero | 3.8 ± 0.3 |
| 1020 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 113 | 25 | 0.44 | Alghero | 3.8 ± 0.3 |
| 1021 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 207 | 29 | 0.60 | Alghero | 3.8 ± 0.3 |
| 1022 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 165 | 27 | 0.59 | Alghero | 3.8 ± 0.3 |
| 1023 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 101 | 24 | 0.40 | Alghero | 3.8 ± 0.3 |
| 1024 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 105 | 24 | 0.67 | Alghero | 3.8 ± 0.3 |
| 1025 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 130 | 24 | 0.36 | Alghero | 3.8 ± 0.3 |
| 1026 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 65 | 20 | 0.13 | Alghero | 3.8 ± 0.3 |
| 1027 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 70 | 22 | 0.42 | Civitavecchia | 3.8 ± 0.3 |
| 1028 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 75 | 23 | 0.38 | Civitavecchia | 3.8 ± 0.3 |
| 1029 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 169 | 29 | 2.15 | Civitavecchia | 3.8 ± 0.3 |
| 1030 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 105 | 27 | 0.50 | Civitavecchia | 3.8 ± 0.3 |
| 1031 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 132 | 27 | 0.56 | Civitavecchia | 3.8 ± 0.3 |
| 1032 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 173 | 30 | 0.51 | Civitavecchia | 3.8 ± 0.3 |
| 1033 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 341 | 35 | 0.56 | Civitavecchia | 3.8 ± 0.3 |
| 1034 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 368 | 39 | 0.40 | Civitavecchia | 3.8 ± 0.3 |
| 1035 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 404 | 38 | 1.49 | Civitavecchia | 3.8 ± 0.3 |
| 1036 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 264 | 32 | 0.21 | Civitavecchia | 3.8 ± 0.3 |
| 1037 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 81 | 23 | 0.38 | Civitavecchia | 3.8 ± 0.3 |
| 1038 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 287 | 31 | 0.24 | Genoa | 3.8 ± 0.3 |
| 1039 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 300 | 32 | 0.16 | Genoa | 3.8 ± 0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1040 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 306 | 32 | 0.16 | Genoa | 3.8 ± 0.3 |
| 1041 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 253 | 30 | 0.46 | Genoa | 3.8 ± 0.3 |
| 1042 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 230 | 28 | 0.20 | Genoa | 3.8 ± 0.3 |
| 1043 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 328 | 33 | 0.34 | Genoa | 3.8 ± 0.3 |
| 1044 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 284 | 30 | 0.22 | Genoa | 3.8 ± 0.3 |
| 1045 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 287 | 31 | 0.41 | Genoa | 3.8 ± 0.3 |
| 1046 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 351 | 34 | 0.28 | Genoa | 3.8 ± 0.3 |
| 1047 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 440 | 35 | 0.28 | Genoa | 3.8 ± 0.3 |
| 1048 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 393 | 34 | 0.28 | Genoa | 3.8 ± 0.3 |
| 1049 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 330 | 32 | 0.26 | Genoa | 3.8 ± 0.3 |
| 1050 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 102 | 23.5 | 0.35 | Marseille | 3.8 ± 0.3 |
| 1051 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 86 | 21 | 0.30 | Marseille | 3.8 ± 0.3 |
| 1052 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 111 | 24 | 0.41 | Marseille | 3.8 ± 0.3 |
| 1053 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 118 | 24 | 0.65 | Marseille | 3.8 ± 0.3 |
| 1054 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 112 | 23.5 | 0.41 | Marseille | 3.8 ± 0.3 |
| 1055 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 128 | 27 | 0.92 | Marseille | 3.8 ± 0.3 |
| 1056 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 127 | 24 | 0.68 | Marseille | 3.8 ± 0.3 |
| 1057 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 203 | 30 | 0.81 | Marseille | 3.8 ± 0.3 |
| 1058 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 166 | 27 | 0.78 | Marseille | 3.8 ± 0.3 |
| 1059 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 128 | 25 | 0.73 | Marseille | 3.8 ± 0.3 |
| 1060 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 168 | 26 | 0.31 | Marseille | 3.8 ± 0.3 |
| 1061 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 157 | 27 | 0.28 | Marseille | 3.8 ± 0.3 |
| 1062 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 85 | 23 | 0.05 | Marseille | 3.8 ± 0.3 |
| 1063 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 342 | 34 | 0.91 | Marseille | 3.8 ± 0.3 |
| 1064 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 200 | 32 | 2.49 | Marseille | 3.8 ± 0.3 |
| 1065 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 114 | 26 | 0.31 | Marseille | 3.8 ± 0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1066 | ATL. HORSE MACKEREL | *Trachurus trachurus* | 245 | 34 | 1.43 | Marseille | 3.8 ± 0.3 |
| 1067 | PEARLY RAZORFISH | *Xyrichtys novacula* | - | - | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1068 | PEARLY RAZORFISH | *Xyrichtys novacula* | - | - | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1069 | PEARLY RAZORFISH | *Xyrichtys novacula* | - | - | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1070 | PEARLY RAZORFISH | *Xyrichtys novacula* | - | - | 0.13 | Balearic Islands | 3.5 ±0.1 |
| 1071 | PEARLY RAZORFISH | *Xyrichtys novacula* | 310 | 12 | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1072 | PEARLY RAZORFISH | *Xyrichtys novacula* | 280 | 17 | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1073 | PEARLY RAZORFISH | *Xyrichtys novacula* | 270 | 16 | 0.09 | Balearic Islands | 3.5 ±0.1 |
| 1074 | JOHN DORY | *Zeus faber* | 450 | 42 | 0.17 | Balearic Islands | 4.5 ±0.8 |
| 1075 | JOHN DORY | *Zeus faber* | 350 | 40 | 0.24 | Balearic Islands | 4.5 ±0.8 |
| 1076 | JOHN DORY | *Zeus faber* | 450 | 42 | 0.36 | Balearic Islands | 4.5 ±0.8 |
| 1077 | JOHN DORY | *Zeus faber* | 400 | 35 | 0.12 | Balearic Islands | 4.5 ±0.8 |
| 1078 | JOHN DORY | *Zeus faber* | 300 | 31 | 0.12 | Balearic Islands | 4.5 ±0.8 |
| 1079 | JOHN DORY | *Zeus faber* | 340 | 40 | 0.09 | Balearic Islands | 4.5 ±0.8 |
| 1080 | JOHN DORY | *Zeus faber* | 300 | 20 | 0.18 | Balearic Islands | 4.5 ±0.8 |
| 1081 | JOHN DORY | *Zeus faber* | 250 | 20 | 0.09 | Balearic Islands | 4.5 ±0.8 |
| 1082 | JOHN DORY | *Zeus faber* | 1900 | 54 | 0.38 | Balearic Islands | 4.5 ±0.8 |
| 1083 | JOHN DORY | *Zeus faber* | 1620 | 50 | 1.30 | Balearic Islands | 4.5 ±0.8 |
| 1084 | JOHN DORY | *Zeus faber* | 780 | 39 | 0.26 | Balearic Islands | 4.5 ±0.8 |
| 1085 | JOHN DORY | *Zeus faber* | - | - | 1.30 | Balearic Islands | 4.5 ±0.8 |
| 1086 | JOHN DORY | *Zeus faber* | - | - | 0.26 | Balearic Islands | 4.5 ±0.8 |
| 1087 | JOHN DORY | *Zeus faber* | 500 | 23.5 | 0.13 | Balearic Islands | 4.5 ±0.8 |
| 1088 | JOHN DORY | *Zeus faber* | 760 | 28 | 0.13 | Balearic Islands | 4.5 ±0.8 |
| 1089 | JOHN DORY | *Zeus faber* | 1100 | 38 | 0.93 | Balearic Islands | 4.5 ±0.8 |
| 1090 | JOHN DORY | *Zeus faber* | 1100 | 30 | 0.57 | Balearic Islands | 4.5 ±0.8 |
| 1091 | JOHN DORY | *Zeus faber* | 830 | 30 | 0.29 | Balearic Islands | 4.5 ±0.8 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1092 | JOHN DORY | *Zeus faber* | 115 | 21 | 0.07 | Alacant | 4.5 ±0.8 |
| 1093 | JOHN DORY | *Zeus faber* | 185 | 24 | 0.17 | Alacant | 4.5 ±0.8 |
| 1094 | JOHN DORY | *Zeus faber* | 397 | 32 | 0.11 | Alacant | 4.5 ±0.8 |
| 1095 | JOHN DORY | *Zeus faber* | 473 | 34 | 0.09 | Alacant | 4.5 ±0.8 |
| 1096 | JOHN DORY | *Zeus faber* | 349 | 30.5 | 0.09 | Alacant | 4.5 ±0.8 |
| 1097 | COMMON SEABREAM | *Pagrus pagrus* | 192 | 26 | 0.92 | Alghero | 3.9 ±0.2 |
| 1098 | COMMON SEABREAM | *Pagrus pagrus* | 151 | 22 | 0.43 | Alghero | 3.9 ±0.2 |
| 1099 | COMMON SEABREAM | *Pagrus pagrus* | 152 | 20 | 0.27 | Alghero | 3.9 ±0.2 |
| 1100 | COMMON SEABREAM | *Pagrus pagrus* | 215 | 25 | 0.53 | Alghero | 3.9 ±0.2 |
| 1101 | COMMON SEABREAM | *Pagrus pagrus* | 976 | 38 | 0.12 | Alacant | 3.9 ±0.2 |
| 1102 | COMMON SEABREAM | *Pagrus pagrus* | 847 | 38 | 0.16 | Alacant | 3.9 ±0.2 |
| 1103 | COMMON SEABREAM | *Pagrus pagrus* | 655 | 39 | 0.14 | Alacant | 3.9 ±0.2 |
| 1104 | COMMON SEABREAM | *Pagrus pagrus* | 711 | 34 | 0.13 | Alacant | 3.9 ±0.2 |
| 1105 | COMMON SEABREAM | *Pagrus pagrus* | 63 | 17 | 0.59 | Civitavecchia | 3.9 ±0.2 |
| 1106 | COMMON SEABREAM | *Pagrus pagrus* | 38 | 16 | 0.63 | Civitavecchia | 3.9 ±0.2 |
| 1107 | COMMON SEABREAM | *Pagrus pagrus* | 37 | 14 | 0.67 | Civitavecchia | 3.9 ±0.2 |
| 1108 | COMMON SEABREAM | *Pagrus pagrus* | 32 | 13 | 0.52 | Civitavecchia | 3.9 ±0.2 |
| 1109 | COMMON SEABREAM | *Pagrus pagrus* | 39 | 15 | 0.37 | Civitavecchia | 3.9 ±0.2 |
| 1110 | COMMON SEABREAM | *Pagrus pagrus* | 31 | 14 | 0.60 | Civitavecchia | 3.9 ±0.2 |
| 1111 | COMMON SEABREAM | *Pagrus pagrus* | 51 | 17 | 0.44 | Civitavecchia | 3.9 ±0.2 |
| 1112 | COMMON SEABREAM | *Pagrus pagrus* | 43 | 16 | 0.49 | Civitavecchia | 3.9 ±0.2 |
| 1113 | COMMON SEABREAM | *Pagrus pagrus* | 37 | 14 | 0.12 | Civitavecchia | 3.9 ±0.2 |
| 1114 | COMMON SEABREAM | *Pagrus pagrus* | 51 | 16 | 0.46 | Civitavecchia | 3.9 ±0.2 |
| 1115 | COMMON SEABREAM | *Pagrus pagrus* | 43 | 15 | 0.72 | Civitavecchia | 3.9 ±0.2 |
| 1116 | COMMON SEABREAM | *Pagrus pagrus* | 60 | 18 | 0.62 | Civitavecchia | 3.9 ±0.2 |
| 1117 | COMMON SEABREAM | *Pagrus pagrus* | 62 | 18 | 0.73 | Civitavecchia | 3.9 ±0.2 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1118 | COMMON SEABREAM | *Pagrus pagrus* | 35 | 15 | 0.34 | Civitavecchia | 3.9 ±0.2 |
| 1119 | COMMON SEABREAM | *Pagrus pagrus* | 41 | 15 | 0.24 | Civitavecchia | 3.9 ±0.2 |
| 1120 | COMMON SEABREAM | *Pagrus pagrus* | 102 | 21 | 0.25 | Civitavecchia | 3.9 ±0.2 |
| 1121 | COMMON SEABREAM | *Pagrus pagrus* | 103 | 21 | 0.38 | Civitavecchia | 3.9 ±0.2 |
| 1122 | COMMON SEABREAM | *Pagrus pagrus* | 390 | 29 | 0.52 | Genoa | 3.9 ±0.2 |
| 1123 | COMMON SEABREAM | *Pagrus pagrus* | 310 | 30 | 0.46 | Genoa | 3.9 ±0.2 |
| 1124 | COMMON SEABREAM | *Pagrus pagrus* | 312 | 31 | 1.01 | Genoa | 3.9 ±0.2 |
| 1125 | COMMON SEABREAM | *Pagrus pagrus* | 377 | 31 | 0.99 | Genoa | 3.9 ±0.2 |
| 1126 | COMMON SEABREAM | *Pagrus pagrus* | 337 | 31 | 0.15 | Genoa | 3.9 ±0.2 |
| 1127 | COMMON SEABREAM | *Pagrus pagrus* | 401 | 33 | 0.77 | Genoa | 3.9 ±0.2 |
| 1128 | COMMON SEABREAM | *Pagrus pagrus* | 332 | 30 | 0.98 | Genoa | 3.9 ±0.2 |
| 1129 | COMMON SEABREAM | *Pagrus pagrus* | 311 | 29 | 0.43 | Genoa | 3.9 ±0.2 |
| 1130 | COMMON SEABREAM | *Pagrus pagrus* | 324 | 32 | 0.79 | Genoa | 3.9 ±0.2 |
| 1131 | COMMON SEABREAM | *Pagrus pagrus* | 287 | 28 | 0.73 | Genoa | 3.9 ±0.2 |
| 1132 | COMMON SEABREAM | *Pagrus pagrus* | 448 | 33 | 0.87 | Genoa | 3.9 ±0.2 |
| 1133 | COMMON SEABREAM | *Pagrus pagrus* | 411 | 32 | 1.14 | Genoa | 3.9 ±0.2 |
| 1134 | COMMON SEABREAM | *Pagrus pagrus* | 208 | 26 | 0.72 | Marseille | 3.9 ±0.2 |
| 1135 | COMMON SEABREAM | *Pagrus pagrus* | 200 | 23 | 0.93 | Marseille | 3.9 ±0.2 |
| 1136 | COMMON SEABREAM | *Pagrus pagrus* | 212 | 27 | 0.87 | Marseille | 3.9 ±0.2 |
| 1137 | COMMON SEABREAM | *Pagrus pagrus* | 180 | 26 | 0.72 | Marseille | 3.9 ±0.2 |
| 1138 | COMMON SEABREAM | *Pagrus pagrus* | 257 | 28 | 1.07 | Marseille | 3.9 ±0.2 |
| 1139 | COMMON SEABREAM | *Pagrus pagrus* | 240 | 27 | 0.80 | Marseille | 3.9 ±0.2 |
| 1140 | COMMON SEABREAM | *Pagrus pagrus* | 201 | 26 | 0.96 | Marseille | 3.9 ±0.2 |
| 1141 | COMMON SEABREAM | *Pagrus pagrus* | 84 | 19 | 0.74 | Marseille | 3.9 ±0.2 |
| 1142 | COMMON SEABREAM | *Pagrus pagrus* | 154 | 22 | 0.75 | Marseille | 3.9 ±0.2 |
| 1143 | COMMON SEABREAM | *Pagrus pagrus* | 207 | 26 | 0.27 | Marseille | 3.9 ±0.2 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1144 | COMMON SEABREAM | *Pagrus pagrus* | 258 | 28 | 1.31 | Marseille | 3.9 ±0.2 |
| 1145 | COMMON SEABREAM | *Pagrus pagrus* | 268 | 30 | 1.68 | Marseille | 3.9 ±0.2 |
| 1146 | COMMON SEABREAM | *Pagrus pagrus* | 405 | 32 | 0.73 | Marseille | 3.9 ±0.2 |
| 1147 | COMMON SEABREAM | *Pagrus pagrus* | 176 | 24.5 | 0.31 | Marseille | 3.9 ±0.2 |
| 1148 | COMMON SEABREAM | *Pagrus pagrus* | 448 | 31 | 0.22 | Balearic Islands | 3.9 ±0.2 |
| 1149 | COMMON SEABREAM | *Pagrus pagrus* | 562 | 32 | 0.18 | Balearic Islands | 3.9 ±0.2 |
| 1150 | COMMON SEABREAM | *Pagrus pagrus* | 833 | 39 | 1.27 | Balearic Islands | 3.9 ±0.2 |
| 1151 | COMMON SEABREAM | *Pagrus pagrus* | 785 | 35 | 0.48 | Balearic Islands | 3.9 ±0.2 |
| 1152 | COMMON SEABREAM | *Pagrus pagrus* | 254 | 24 | 0.19 | Balearic Islands | 3.9 ±0.2 |
| 1153 | COMMON SEABREAM | *Pagrus pagrus* | 288 | 26 | 0.22 | Balearic Islands | 3.9 ±0.2 |
| 1154 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 49 | 17 | 0.09 | Alghero | 4.3 ±0.1 |
| 1155 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 33 | 14 | 0.09 | Alghero | 4.3 ±0.1 |
| 1156 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 21 | 13 | 0.05 | Alghero | 4.3 ±0.1 |
| 1157 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 34 | 16 | 0.07 | Alghero | 4.3 ±0.1 |
| 1158 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 40 | 16 | 0.18 | Alghero | 4.3 ±0.1 |
| 1159 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 25 | 14 | 0.06 | Alghero | 4.3 ±0.1 |
| 1160 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 42 | 16 | 0.08 | Alghero | 4.3 ±0.1 |
| 1161 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 41 | 17 | 0.16 | Alghero | 4.3 ±0.1 |
| 1162 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 60 | 19 | 0.17 | Alghero | 4.3 ±0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1163 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 27 | 15 | 0.09 | Alghero | 4.3 ±0.1 |
| 1164 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 25 | 15 | 0.09 | Alacant | 4.3 ±0.1 |
| 1165 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 24 | 16 | 0.17 | Alacant | 4.3 ±0.1 |
| 1166 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 23 | 17 | 0.08 | Alacant | 4.3 ±0.1 |
| 1167 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 35 | 16 | 0.63 | Alacant | 4.3 ±0.1 |
| 1168 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 49 | 18 | 0.08 | Alacant | 4.3 ±0.1 |
| 1169 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 39 | 16.5 | 0.06 | Alacant | 4.3 ±0.1 |
| 1170 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 29 | 16 | 0.09 | Alacant | 4.3 ±0.1 |
| 1171 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 33 | 17 | 0.25 | Alacant | 4.3 ±0.1 |
| 1172 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 21 | 14 | 0.10 | Alacant | 4.3 ±0.1 |
| 1173 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 14 | 12 | 0.08 | Alacant | 4.3 ±0.1 |
| 1174 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 16 | 13 | 0.09 | Alacant | 4.3 ±0.1 |
| 1175 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 23 | 16 | 0.11 | Alacant | 4.3 ±0.1 |
| 1176 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 31 | 17 | 0.10 | Alacant | 4.3 ±0.1 |
| 1177 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 13 | 12.5 | 0.12 | Alacant | 4.3 ±0.1 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1178 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 33 | 18 | 0.10 | Alacant | 4.3 ±0.1 |
| 1179 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 18 | 14.5 | 0.11 | Alacant | 4.3 ±0.1 |
| 1180 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 15 | 12 | 0.10 | Alacant | 4.3 ±0.1 |
| 1181 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 15 | 14 | 0.13 | Alacant | 4.3 ±0.1 |
| 1182 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 41 | 19 | 1.08 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1183 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 33 | 17 | 1.07 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1184 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 37 | 17 | 0.71 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1185 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 42 | 19 | 0.55 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1186 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 39 | 17 | 0.86 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1187 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 36 | 18 | 0.60 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1188 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 43 | 18 | 0.86 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1189 | MEGRIM SOLE | *Lepidorhombus whiffiagonis* | 38 | 18 | 1.23 | L'Ampolla - Ebro Delta | 4.3 ±0.1 |
| 1190 | GURNARD | *Chelidonichthys lucerna* | 251 | 30 | 0.12 | Alacant | 4 |
| 1191 | GURNARD | *Chelidonichthys lucerna* | 244 | 30 | 0.15 | Alacant | 4 |
| 1192 | GURNARD | *Chelidonichthys lucerna* | 135 | 25 | 0.75 | Alacant | 4 |
| 1193 | GURNARD | *Chelidonichthys lucerna* | 98 | 21 | 0.37 | Alacant | 4 |
| 1194 | GURNARD | *Chelidonichthys lucerna* | 154 | 25 | 0.91 | Marseille | 4 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1195 | GURNARD | *Chelidonichthys lucerna* | 159 | 25 | 0.38 | Marseille | 4 |
| 1196 | GURNARD | *Chelidonichthys lucerna* | 141 | 25 | 0.18 | Marseille | 4 |
| 1197 | GILTHEAD SEABREAM | *Sparus aurata* | 620 | 36 | 0.20 | Alacant | 3.7 |
| 1198 | GILTHEAD SEABREAM | *Sparus aurata* | 605 | 35 | 0.11 | Alacant | 3.7 |
| 1199 | GILTHEAD SEABREAM | *Sparus aurata* | 548 | 47 | 0.01 | Alacant | 3.7 |
| 1200 | GILTHEAD SEABREAM | *Sparus aurata* | 650 | 47 | 0.36 | Alacant | 3.7 |
| 1201 | GILTHEAD SEABREAM | *Sparus aurata* | 536 | 33 | 0.19 | Marseille | 3.7 |
| 1202 | GREATER WEEVER | *Trachinus draco* | 116 | 25.5 | 0.09 | Alacant | 4.2 ±0.71 |
| 1203 | GREATER WEEVER | *Trachinus draco* | 76 | 23.5 | 0.16 | Alacant | 4.2 ±0.71 |
| 1204 | GREATER WEEVER | *Trachinus draco* | 125 | 27.5 | 0.19 | Alacant | 4.2 ±0.71 |
| 1205 | GREATER WEEVER | *Trachinus draco* | 76 | 23.5 | 0.19 | Alacant | 4.2 ±0.71 |
| 1206 | GREATER WEEVER | *Trachinus draco* | 105 | 26 | 0.04 | Alacant | 4.2 ±0.71 |
| 1207 | GREATER WEEVER | *Trachinus draco* | 101 | 20 | 0.35 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1208 | GREATER WEEVER | *Trachinus draco* | 57 | 21 | 0.18 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1209 | GREATER WEEVER | *Trachinus draco* | 96 | 24 | 1.34 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1210 | GREATER WEEVER | *Trachinus draco* | 127 | 27 | 0.88 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1211 | GREATER WEEVER | *Trachinus draco* | 90 | 24 | 0.81 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1212 | GREATER WEEVER | *Trachinus draco* | 133 | 27 | 1.34 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1213 | GREATER WEEVER | *Trachinus draco* | 55 | 19 | 0.32 | L'Ampolla - Ebro Delta | 4.2 ±0.71 |
| 1214 | POOR COD | *Trisopterus minutus* | 23 | 14 | 0.15 | Alacant | 3.7 ±0.2 |
| 1215 | POOR COD | *Trisopterus minutus* | 107 | 21 | 0.51 | Alacant | 3.7 ±0.2 |
| 1216 | POOR COD | *Trisopterus minutus* | 54 | 18 | 0.19 | Alacant | 3.7 ±0.2 |
| 1217 | POOR COD | *Trisopterus minutus* | 62 | 19.5 | 0.24 | Alacant | 3.7 ±0.2 |
| 1218 | POOR COD | *Trisopterus minutus* | 58 | 18 | 0.17 | Alacant | 3.7 ±0.2 |
| 1219 | POOR COD | *Trisopterus minutus* | 106 | 21 | 0.41 | Alacant | 3.7 ±0.2 |
| 1220 | POOR COD | *Trisopterus minutus* | 58 | 19 | 0.36 | Alacant | 3.7 ±0.2 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1221 | POOR COD | *Trisopterus minutus* | 73 | 20 | 0.25 | Alacant | 3.7 ±0.2 |
| 1222 | POOR COD | *Trisopterus minutus* | 57 | 18.5 | 0.18 | Alacant | 3.7 ±0.2 |
| 1223 | POOR COD | *Trisopterus minutus* | 56 | 18.5 | 0.22 | Alacant | 3.7 ±0.2 |
| 1224 | POOR COD | *Trisopterus minutus* | 46 | 17 | 0.22 | Alacant | 3.7 ±0.2 |
| 1225 | POOR COD | *Trisopterus minutus* | 61 | 19 | 0.27 | Alacant | 3.7 ±0.2 |
| 1226 | POOR COD | *Trisopterus minutus* | 38 | 16 | 0.17 | Alacant | 3.7 ±0.2 |
| 1227 | POOR COD | *Trisopterus minutus* | 44 | 17.5 | 0.51 | Alacant | 3.7 ±0.2 |
| 1228 | POOR COD | *Trisopterus minutus* | 40 | 11 | 0.12 | Alacant | 3.7 ±0.2 |
| 1229 | RED BANDFISH | *Cepola macrophthalma* | 173 | 72 | 0.02 | Alacant | 3.1 ±0.23 |
| 1230 | RED BANDFISH | *Cepola macrophthalma* | 184 | 75 | 0.03 | Alacant | 3.1 ±0.23 |
| 1231 | WHITE SARDINELLA | *Sardinella albella* | 46 | 18.5 | 0.09 | Alacant | 2.6 ±0.14 |
| 1232 | WHITE SARDINELLA | *Sardinella albella* | 65 | 25 | 0.31 | Alacant | 2.6 ±0.14 |
| 1233 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 601 | 34 | 0.11 | Alacant | 4.1 ±0.70 |
| 1234 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 113 | 20 | 0.41 | Alacant | 4.1 ±0.70 |
| 1235 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 276 | 27 | 0.05 | Alacant | 4.1 ±0.70 |
| 1236 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 42 | 16 | 0.09 | Alacant | 4.1 ±0.70 |
| 1237 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 67 | 16 | 0.10 | Alacant | 4.1 ±0.70 |
| 1238 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 86 | 19.5 | 0.09 | Alacant | 4.1 ±0.70 |
| 1239 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 104 | 20 | 1.02 | Ametlla de Mar - Ebro Delta | 4.1 ±0.70 |
| 1240 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 113 | 20 | 0.52 | Ametlla de Mar - Ebro Delta | 4.1 ±0.70 |
| 1241 | OFFSHORE ROCKFISH | *Pontinus kuhlii* | 159 | 22 | 0.60 | Ametlla de Mar - Ebro Delta | 4.1 ±0.70 |
| 1242 | CUCKOO WRASSE | *Labrus bimaculatus* | 376 | 32 | 0.29 | Alacant | 3.9 ±0.62 |
| 1243 | BLUE WHITING | *Micromesistius poutassou* | 47 | 20 | 0.05 | Alacant | 4.1 ±0.3 |
| 1244 | BLUE WHITING | *Micromesistius poutassou* | 54 | 21 | 0.19 | Alacant | 4.1 ±0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1245 | BLUE WHITING | *Micromesistius poutassou* | 55 | 21 | 0.08 | Alacant | 4.1 ±0.3 |
| 1246 | BLUE WHITING | *Micromesistius poutassou* | 56 | 20 | 0.16 | Alacant | 4.1 ±0.3 |
| 1247 | BLUE WHITING | *Micromesistius poutassou* | 48 | 20 | 0.09 | Alacant | 4.1 ±0.3 |
| 1248 | BLUE WHITING | *Micromesistius poutassou* | 64 | 22 | 0.16 | Alacant | 4.1 ±0.3 |
| 1249 | BLUE WHITING | *Micromesistius poutassou* | 46 | 24 | 0.09 | Alacant | 4.1 ±0.3 |
| 1250 | BLUE WHITING | *Micromesistius poutassou* | 50 | 20 | 0.08 | Alacant | 4.1 ±0.3 |
| 1251 | CUTTLEFISH | *Sepia officinalis* | 686 | 48 | 0.02 | Alacant | 3.6 |
| 1252 | CUTTLEFISH | *Sepia officinalis* | 664 | 43 | 0.02 | Alacant | 3.6 |
| 1253 | CUTTLEFISH | *Sepia officinalis* | 465 | 43 | 0.02 | Alacant | 3.6 |
| 1254 | CUTTLEFISH | *Sepia officinalis* | 647 | 44 | 0.03 | Alacant | 3.6 |
| 1255 | CUTTLEFISH | *Sepia officinalis* | 270 | 42 | 0.11 | Alacant | 3.6 |
| 1256 | CUTTLEFISH | *Sepia officinalis* | 204 | 41 | 0.12 | Alacant | 3.6 |
| 1257 | CUTTLEFISH | *Sepia officinalis* | 245 | 44 | 0.09 | Alacant | 3.6 |
| 1258 | CUTTLEFISH | *Sepia officinalis* | 296 | 40 | 0.08 | Alacant | 3.6 |
| 1259 | CUTTLEFISH | *Sepia officinalis* | 235 | 40 | 0.02 | Civitavecchia | 3.6 |
| 1260 | CUTTLEFISH | *Sepia officinalis* | 233 | 37 | 0.03 | Civitavecchia | 3.6 |
| 1261 | CUTTLEFISH | *Sepia officinalis* | 291 | 45 | 0.02 | Civitavecchia | 3.6 |
| 1262 | CUTTLEFISH | *Sepia officinalis* | 306 | 48 | 0.03 | Civitavecchia | 3.6 |
| 1263 | CUTTLEFISH | *Sepia officinalis* | 158 | 45 | 0.26 | Civitavecchia | 3.6 |
| 1264 | CUTTLEFISH | *Sepia officinalis* | 214 | 41 | 0.26 | Civitavecchia | 3.6 |
| 1265 | CUTTLEFISH | *Sepia officinalis* | 204 | 38 | 0.23 | Civitavecchia | 3.6 |
| 1266 | CUTTLEFISH | *Sepia officinalis* | 176 | 24 | 0.11 | Civitavecchia | 3.6 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1267 | CUTTLEFISH | *Sepia officinalis* | 102 | 32 | 0.04 | Genoa | 3.6 |
| 1268 | CUTTLEFISH | *Sepia officinalis* | 128 | 38 | 0.05 | Genoa | 3.6 |
| 1269 | CUTTLEFISH | *Sepia officinalis* | 95 | 30 | 0.05 | Genoa | 3.6 |
| 1270 | CUTTLEFISH | *Sepia officinalis* | 130 | 35 | 0.07 | Genoa | 3.6 |
| 1271 | CUTTLEFISH | *Sepia officinalis* | 85 | 29 | 0.06 | Genoa | 3.6 |
| 1272 | CUTTLEFISH | *Sepia officinalis* | 126 | 34 | 0.09 | Genoa | 3.6 |
| 1273 | CUTTLEFISH | *Sepia officinalis* | 120 | 37 | 0.14 | Genoa | 3.6 |
| 1274 | CUTTLEFISH | *Sepia officinalis* | 81 | 29 | 0.05 | Genoa | 3.6 |
| 1275 | CUTTLEFISH | *Sepia officinalis* | 367 | 50 | 0.40 | Marseille | 3.6 |
| 1276 | CUTTLEFISH | *Sepia officinalis* | 864 | 58 | 0.71 | Marseille | 3.6 |
| 1277 | CUTTLEFISH | *Sepia officinalis* | 275 | 43 | 0.47 | Marseille | 3.6 |
| 1278 | CUTTLEFISH | *Sepia officinalis* | 221 | 47 | 0.12 | Marseille | 3.6 |
| 1279 | CUTTLEFISH | *Sepia officinalis* | 260 | 47 | 0.05 | Marseille | 3.6 |
| 1280 | CUTTLEFISH | *Sepia officinalis* | 176 | 30 | 0.05 | Balearic Islands | 3.6 |
| 1281 | CUTTLEFISH | *Sepia officinalis* | 143 | 32 | 0.03 | Balearic Islands | 3.6 |
| 1282 | CUTTLEFISH | *Sepia officinalis* | 126 | 32 | 0.06 | Balearic Islands | 3.6 |
| 1283 | CUTTLEFISH | *Sepia officinalis* | 146 | 26 | 0.06 | Balearic Islands | 3.6 |
| 1284 | CUTTLEFISH | *Sepia officinalis* | 169 | 41 | 0.06 | Balearic Islands | 3.6 |
| 1285 | CUTTLEFISH | *Sepia officinalis* | 151 | 37 | 0.05 | Balearic Islands | 3.6 |
| 1286 | CUTTLEFISH | *Sepia officinalis* | 125 | 34 | 0.05 | Balearic Islands | 3.6 |
| 1287 | CUTTLEFISH | *Sepia officinalis* | 157 | 23 | 0.05 | Balearic Islands | 3.6 |
| 1288 | CUTTLEFISH | *Sepia officinalis* | 160 | 24 | 0.06 | Balearic Islands | 3.6 |
| 1289 | CUTTLEFISH | *Sepia officinalis* | 185 | 36 | 0.04 | Balearic Islands | 3.6 |
| 1290 | CUTTLEFISH | *Sepia officinalis* | 133 | 39 | 0.08 | L'Ampolla - Ebro Delta | 3.6 |
| 1291 | CUTTLEFISH | *Sepia officinalis* | 110 | 33 | 0.08 | L'Ampolla - Ebro Delta | 3.6 |
| 1292 | CUTTLEFISH | *Sepia officinalis* | 174 | 40 | 0.11 | L'Ampolla - Ebro Delta | 3.6 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1293 | CUTTLEFISH | *Sepia officinalis* | 122 | 39 | 0.10 | L'Ampolla - Ebro Delta | 3.6 |
| 1294 | CUTTLEFISH | *Sepia officinalis* | 167 | 22 | 0.17 | L'Ampolla - Ebro Delta | 3.6 |
| 1295 | CUTTLEFISH | *Sepia officinalis* | 134 | 40 | 0.10 | L'Ampolla - Ebro Delta | 3.6 |
| 1296 | CUTTLEFISH | *Sepia officinalis* | 100 | 27 | 0.09 | L'Ampolla - Ebro Delta | 3.6 |
| 1297 | CUTTLEFISH | *Sepia officinalis* | 135 | 37 | 0.14 | L'Ampolla - Ebro Delta | 3.6 |
| 1298 | WHITING | *Merlangius merlangus* | 114 | 28 | 0.23 | Civitavecchia | 4.4 ±0.2 |
| 1299 | WHITING | *Merlangius merlangus* | 123 | 26 | 0.24 | Civitavecchia | 4.4 ±0.2 |
| 1300 | WHITING | *Merlangius merlangus* | 182 | 30 | 0.17 | Civitavecchia | 4.4 ±0.2 |
| 1301 | WHITING | *Merlangius merlangus* | 176 | 31 | 0.44 | Civitavecchia | 4.4 ±0.2 |
| 1302 | WHITING | *Merlangius merlangus* | 176 | 29 | 0.33 | Civitavecchia | 4.4 ±0.2 |
| 1303 | WHITING | *Merlangius merlangus* | 173 | 30 | 0.81 | Civitavecchia | 4.4 ±0.2 |
| 1304 | WHITING | *Merlangius merlangus* | 107 | 26 | 0.13 | Civitavecchia | 4.4 ±0.2 |
| 1305 | WHITING | *Merlangius merlangus* | 85 | 23 | 0.14 | Civitavecchia | 4.4 ±0.2 |
| 1306 | WHITING | *Merlangius merlangus* | 140 | 28 | 0.15 | Marseille | 4.4 ±0.2 |
| 1307 | WHITING | *Merlangius merlangus* | 126 | 25 | 0.13 | Marseille | 4.4 ±0.2 |
| 1308 | WHITING | *Merlangius merlangus* | 112 | 24 | 0.19 | Marseille | 4.4 ±0.2 |
| 1309 | WHITING | *Merlangius merlangus* | 136 | 25 | 0.22 | Marseille | 4.4 ±0.2 |
| 1310 | WHITING | *Merlangius merlangus* | 114 | 23 | 0.15 | Marseille | 4.4 ±0.2 |
| 1311 | WHITING | *Merlangius merlangus* | 109 | 24 | 0.13 | Marseille | 4.4 ±0.2 |
| 1312 | WHITING | *Merlangius merlangus* | 120 | 25 | 0.25 | Marseille | 4.4 ±0.2 |
| 1313 | WHITING | *Merlangius merlangus* | 114 | 25 | 0.28 | Marseille | 4.4 ±0.2 |
| 1314 | WHITING | *Merlangius merlangus* | 123 | 25 | 0.13 | Marseille | 4.4 ±0.2 |
| 1315 | WHITING | *Merlangius merlangus* | 115 | 25 | 0.42 | Marseille | 4.4 ±0.2 |
| 1316 | WHITING | *Merlangius merlangus* | 123 | 26 | 0.10 | Marseille | 4.4 ±0.2 |
| 1317 | WHITING | *Merlangius merlangus* | 121 | 28 | 0.07 | Marseille | 4.4 ±0.2 |
| 1318 | SAND STEENBRAS | *Lithognathus mormyrus* | 207 | 35 | 0.01 | Genoa | 3.4 ±0.50 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1319 | SALEMA | *Sarpa salpa* | 547 | 34 | 0.01 | Marseille | 2 |
| 1320 | SALEMA | *Sarpa salpa* | 501 | 35 | 0.00 | Marseille | 2 |
| 1321 | SALEMA | *Sarpa salpa* | 406 | 33 | 0.01 | Marseille | 2 |
| 1322 | SALEMA | *Sarpa salpa* | 630 | 37 | 0.01 | Marseille | 2 |
| 1323 | SALEMA | *Sarpa salpa* | 562 | 33 | 0.01 | Marseille | 2 |
| 1324 | SALEMA | *Sarpa salpa* | 475 | 32 | 0.01 | Marseille | 2 |
| 1325 | SALEMA | *Sarpa salpa* | 287 | 29 | 0.01 | Marseille | 2 |
| 1326 | SALEMA | *Sarpa salpa* | 351 | 30 | 0.01 | Marseille | 2 |
| 1327 | MACKEREL | *Scomber scombrus* | 505 | 38 | 0.29 | Marseille | 3.6 ±0.2 |
| 1328 | MACKEREL | *Scomber scombrus* | 591 | 39 | 0.76 | Marseille | 3.6 ±0.2 |
| 1329 | MACKEREL | *Scomber scombrus* | 494 | 36 | 0.45 | Marseille | 3.6 ±0.2 |
| 1330 | MACKEREL | *Scomber scombrus* | 431 | 36 | 0.43 | Marseille | 3.6 ±0.2 |
| 1331 | MACKEREL | *Scomber scombrus* | 248 | 30 | 0.34 | Marseille | 3.6 ±0.2 |
| 1332 | MACKEREL | *Scomber scombrus* | 263 | 31 | 0.40 | Marseille | 3.6 ±0.2 |
| 1333 | MACKEREL | *Scomber scombrus* | 293 | 33 | 0.39 | Marseille | 3.6 ±0.2 |
| 1334 | MACKEREL | *Scomber scombrus* | 332 | 34 | 0.79 | Marseille | 3.6 ±0.2 |
| 1335 | MACKEREL | *Scomber scombrus* | 269 | 33 | 0.33 | Marseille | 3.6 ±0.2 |
| 1336 | MACKEREL | *Scomber scombrus* | 305 | 32 | 0.47 | Marseille | 3.6 ±0.2 |
| 1337 | MACKEREL | *Scomber scombrus* | 281 | 34 | 0.32 | Marseille | 3.6 ±0.2 |
| 1338 | MACKEREL | *Scomber scombrus* | 272 | 32 | 0.44 | Marseille | 3.6 ±0.2 |
| 1339 | MACKEREL | *Scomber scombrus* | 275 | 31.5 | 0.45 | Marseille | 3.6 ±0.2 |
| 1340 | MACKEREL | *Scomber scombrus* | 273 | 31 | 0.36 | Marseille | 3.6 ±0.2 |
| 1341 | FLATHEAD MULLET | *Mugil Cephalus* | 675 | 41 | 0.05 | Marseille | 2.5 ±0.17 |
| 1342 | FLATHEAD MULLET | *Mugil Cephalus* | 710 | 40 | 0.03 | Marseille | 2.5 ±0.17 |
| 1343 | EUROPEAN EEL | *Anguilla anguilla* | 387 | 62 | 0.46 | Marseille | 3.6 ± 0.3 |
| 1344 | EUROPEAN EEL | *Anguilla anguilla* | 525 | 66 | 0.78 | Marseille | 3.6 ± 0.3 |

| Nº SAMPLE | SPECIES | | WEIGHT (g) | LENGTH (cm) | HG CONC (mg/kg), ww | SITE | TROPHIC LEVEL |
|---|---|---|---|---|---|---|---|
| 1345 | OCTOPUS | *Octopus vulgaris* | 428 | 43 | 0.03 | Marseille | 3.74 ± 0.15 |