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Mercury in organisms from the Northwestern Mediterranean slope: Importance of food sources

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HIGHLIGHTS

- Hg and stable isotope ratios were assessed in 7 species from Mediterranean slope.
- Settling phytoplankton was the main OM and Hg source, as confirmed by $\delta^{13}C$ values.
- All species except one exceeded Hg consumption limits.
- Depth and diet were important factors explaining Hg content.
- Results confirmed the concern about Hg in the deep Mediterranean.

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ABSTRACT

Mercury (Hg) is a global threat for marine ecosystems, especially within the Mediterranean Sea. The concern is higher for deep-sea organisms, as the Hg concentration in their tissues is commonly high. To assess the influence of food supply at two trophic levels, total Hg concentrations and carbon and nitrogen stable isotope ratios were determined in 7 species (4 teleosts, 2 sharks, and 1 crustacean) sampled on the upper part of the continental slope of the Gulf of Lions (Northwestern Mediterranean Sea), at depths between 284 and 816 m. Mean Hg concentrations ranged from 1.30 ± 0.61 to 7.13 ± 7.09 μg g^{-1} dry mass, with maximum values observed for small-spotted catshark *Scyliorhinus canicula*. For all species except blue whiting *Micromesistius poutassou*, Hg concentrations were above the health safety limits for human consumption defined by the European Commission, with a variable proportion of the individuals exceeding limits (from 23% for the Norway lobster *Nephrops norvegicus* to 82% for the blackbelly rosefish *Helicolenus dactylopterus*). Measured concentrations increased with increasing trophic levels. Carbon isotopic ratios measured for these organisms demonstrated that settling phytoplanktonic organic matter is not only the main source fueling trophic webs but also the carrier of Hg to this habitat. Inter- and intraspecific variations of Hg concentrations revealed the importance of feeding patterns in Hg bioaccumulation. In addition, biological parameters, such as growth rate or bathymetric range explain the observed contamination trends.

1. Introduction

Anthropogenic inputs of chemicals are one of the main threats currently affecting the global ocean (Halpern et al., 2008). Among other contaminants, mercury (Hg) is assessed as being of great concern (UNEP, 2013), since it is a neurotoxic metal for mammals. It also affects fish and other animals at levels below acute toxicity (Fitzgerald and Clarkson, 1991; Scheuhammer et al., 2007; Tartu et al., 2013). The volatile metallic form of Hg (Hg^0) is emitted in the environment from both natural and anthropogenic sources, with an increase of anthropic releases in the last centuries (Streets et al., 2011; UNEP, 2013). Its ~1 year atmospheric residence time allows Hg to be transported over long distances (Streets et al., 2011; UNEP, 2013). However, Hg^0 is oxidized into its divalent form (Hg^{II}) in the atmosphere and dissolved in rain or dry deposited. Once deposited, Hg^{II} can be methylated by bacteria in

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soil, marine waters, and sediments (Fitzgerald and Lamborg, 2007). Methylation mainly occurs in low oxygen zones, and at the water/sediment interface, where heterotrophic bacterial activity is at a maximum (Blum et al., 2013; Cossa, 2013; Cossa et al., 2009, 2013; Heimbürger et al., 2010). Methylmercury (MeHg) is the chemical form of mercury of greatest concern, since it can easily be taken up by phytoplankton and magnified along trophic webs (Pickhardt and Fisher, 2007). Bioaccumulation (bioconcentration + biomagnification) is determined by complex processes, linked with metabolic specificities of each organism and physico-chemical properties of the contaminant (e.g. Harmelin-Vivien et al., 2012). Bioconcentration factors of 10^4 to 10^7 were observed for Hg concentrations between marine waters and organisms (Cossa et al., 2012). This process is responsible for the highest Hg concentrations observed in high trophic level species, and for risks for humans when consuming high trophic level marine organisms, like piscivorous fishes (Morel et al., 1998).

The fate of Hg is particularly important for the Mediterranean Sea. Anthropogenic pressure is thus high, as most of the countries around the Mediterranean are highly populated and industrialized (Durrieu de Madron et al., 2011). Since the 1970's, several authors have described high Hg concentrations in Mediterranean teleosts (ray-finned fishes), sharks, and marine mammals. In addition, higher concentrations were observed for Mediterranean organisms when compared with the same species in other oceans, despite rather similar concentrations in waters (Andre et al., 1991; Cossa et al., 2012; Harmelin-Vivien et al., 2009; Thibaud, 1971). On the basis of these observations, several early papers called this trend the "Mediterranean mercury anomaly" and research was undertaken to look for the cause (Aston and Fowler, 1985; Cossa and Coquery, 2005). One hypothesis is that it may be linked to the biogeochemistry of Hg and the methylation depth range. In the Mediterranean, the oxygen minimum zone is shallower than in other marine environments. The methylation zone occurs thus at a lower depth, increasing the potential for MeHg to be integrated in the trophic webs, which are shallow (Cossa et al., 2009, 2012). The second hypothesis focuses on a lower primary productivity in the oligotrophic Mediterranean, and thus a lower "biodilution" of contamination into less abundant biogenic particles at the base of trophic webs (Heimbürger et al., 2010). This lower productivity also reduces the growth rate of higher organisms compared to more eutrophic settings. This has been observed for hake (*Merluccius merluccius*), a predatory teleost (Mellon-Duval et al., 2010). At the same size, a Mediterranean individual is older and shows higher Hg levels than its Atlantic counterpart, due to a slower growth rate, and increased exposure time — hence, a higher potential for bioaccumulation (Cossa et al., 2012).

Assessment of mercury concentrations was mainly performed in shallow Mediterranean species of commercial or cultural interest. Numerous studies have investigated trophic position and Hg concentration in species such as hake, mullet, tuna or marine mammals (Andre et al., 1991; Cossa et al., 2012; Harmelin-Vivien et al., 2009; Storelli and Barone, 2013; Storelli et al., 2005; Thibaud, 1971). Fewer data are available regarding Hg concentrations for species living in deep Mediterranean waters (e.g. Cossa and Coquery, 2005; Hornhung et al., 1993; Koenig et al., 2013). However, there is considerable concern regarding Hg for these species. They are commonly considered as long-lived and slow-growing, which increases potential exposure time to contamination (Drazen and Haedrich, 2012; Koenig et al., 2013), given that deep-sea sediment with high concentrations of Hg have been observed in the Mediterranean Sea (Heimbürger et al., 2012).

Recently, Canals et al. (2013) reported that a strong research effort had been undertaken to assess the hydrological and climatic processes of the deep areas (mainly continental slopes and submarines canyons) of the Gulf of Lions (Northwestern Mediterranean Sea), but little has been done to connect physical trends to biological functioning. They stated that there is an urgent need to "identify major issues and threats affecting deep-sea ecosystems, and to evaluate their potential impacts". Among the missing knowledge that is required for this assessment are the origin of organic matter (OM) and the functioning of trophic webs in these zones. As diet is the main source of contamination for fishes (Hall et al., 1997), understanding what food source fuels deep-sea organisms is crucial to better understand their contamination patterns. Some early papers performed extensive studies of the diet of some teleosts and shark species (e.g. Carrassón et al., 1992; Macpherson, 1979, 1981; Morte et al., 2002) whereas more recent research characterized the OM reaching deep zones (Canals et al., 2006; Heussner et al., 2006; Pasqual et al., 2013; Sanchez-Vidal et al., 2013).

Although stable isotopes have been used to understand food webs in deep Mediterranean zones (Fanelli et al., 2011, 2013; Papiol et al., 2013; Polunin et al., 2001), the connection between OM and Hg at the base and consumers at the top of the trophic web has not been investigated. We hypothesized that stable isotope analyses would be useful for determining the source of OM and also Hg in deep Mediterranean trophic webs. Two OM and Hg sources should be considered, Rhône river OM inputs and sedimentation of phytoplanktonic OM. Stable isotope analyses are based on the link between the isotopic ratio of a consumer and the ratio of its diet, with a known fractionation factor between them. For carbon, this factor is rather low (theoretically + 1‰ at each trophic level), and allows the use of carbon as a tracer of the origin of OM supporting the consumer. Fractionation factor is more important for nitrogen (theoretically + 3.4‰ at each trophic level). Nitrogen is thus commonly used as a proxy of trophic level. Nevertheless, recently published studies demonstrated that these fractionation factors can vary with feeding behavior, trophic position, metabolism, body size or temperature (Cresson et al., 2014; Hussey et al., 2014; Mill et al., 2007; Wyatt et al., 2010). Using stable isotopes is appropriate for understanding trophic levels in deep-sea species in addition with the gut content analyses, as this technique suffers drawbacks (Drazen et al., 2008; Fanelli et al., 2011; Polunin et al., 2001). Due to rapid pressure change, deep-sea species commonly regurgitate their stomach content when caught. Feeding is also sporadic for carnivorous species. Extensive sampling is thus needed to avoid empty or regurgitated stomachs and to get reliable data. Finally, combining stable isotope and contaminant analyses is a powerful approach, as both techniques give integrated information on diet or contamination processes.

There is a paucity of data about the Hg concentration and trophic relationships of teleosts and sharks of the Mediterranean continental slope (Canals et al., 2013). Furthermore, the existing data have been collected in the Western part of the Gulf of Lions. It is generally considered that deep sea species exhibit high Hg concentration, suggesting that biological features of deep-dwelling species, trophic position, and nature of OM fueling deep zones can drive the high contamination pattern. Consequently, the aims of this study were (1) to characterize the trophic position of seven potentially high-trophic level species of sharks (blackmouth catshark *Galeus melastomus*, small-spotted catshark *Scyliorhinus canicula*) teleosts (blackbelly rosefish *Helicolenus dactylopterus*, four-spot megrim *Lepidorhombus boscii*, blue whiting *Micromesistius poutassou*, and Greater forkbeard *Phycis blennoides*), and crustacean (Norway lobster *Nephrops norvegicus*) in the upper continental slope of the French part of the Gulf of Lions (2) to gain knowledge on species Hg concentrations, and (3) to explain the Hg concentrations in the trophic web using stable isotopes.

2. Material and methods

2.1. Study area, organism sampling and dissection

Sampling was performed during the 2012 MEDITS bottom trawl campaign in the Gulf of Lions (R/V L'Europe) at 6 stations, located on the upper slope at depths between 284 and 816 m (Fig. 1). The MEDITS annual bottom trawl surveys have applied a common standardized protocol since 1994 to produce information on benthic and demersal species on the continental shelves and along the upper slopes in the whole Mediterranean Sea (Bertrand et al., 2002). Sites were selected