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Methylmercury $\delta^{13}C$ Compound Specific Stable Isotopic Analysis: Application to Freshwater and Oceanic Fish Samples

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Mercury (Hg) is a global contaminant of major concern to human and ecosystem health. Mercury has a tendency to be naturally converted into the organometallic toxin methylmercury (MeHg, CH3Hg) which is characterized by elevated neurotoxicity, persistence, and biomagnification properties in food webs. Despite significant improvements in environmental Hg science, we still lack answers to some of the most fundamental questions about Hg cycling: where and how MeHg is produced in aquatic ecosystems. The investigation of the inorganic side of the Hg cycle through the use of Hg stable isotopes has allowed to improve our knowledge on Hg sources and dynamics in the environment. Less attention has been paid to the organic side of the Hg cycle, in particular the role and importance of the carbon sources that provide the methyl group ($-CH_3$), and how they contribute to MeHg formation in the environment. The carbon isotope ($\delta^{13}C$) signature of the carbon atom embedded in the methyl group of MeHg may hold key information about its source origin and fate. Here we present the analytical challenges for such type or measurement, and the methodology we developed to achieve MeHg $\delta^{13}C$ compound specific stable isotopic analysis (CSIA) in natural biological materials. We coupled a custom designed purge and trap system (PT) equipped with cryogenic adsorbents to a gas chromatography combustion isotope ratio mass spectrometry (GC-C-IRMS). Methylmercury $\delta^{13}C$ CSIA was performed on distilled biological samples followed by hydride generation (HG)-PT-GC-C-IRMS. Using this methodology, we obtained a MeHg $\delta^{13}C$ value of -22.2±0.5 ‰ on ERM CE-464 Tuna reference material. This value is not different from what we obtained before (-22.1±1.5‰) by the halogenation of MeHg (MeHgI), liquid extraction and injection GC-C-IRMS [1], and requesting higher MeHg concentrations for analysis. Application of this new method to selected freshwater fish samples (Pike) from the Carcans-Maubuisson lake (Southwestern, France) showed MeHg $\delta^{13}C$ values of -38 ‰. Atlantic Bluefin tuna samples collected in the Mediterranean Sea showed values ranging between -16.9 to -22.4‰. These differences in $\delta^{13}C$ values may relate to different carbon sources and/or methylation/demethylation processes.

[1] Masbou, Point et al. (2015), Anal. Chem. 87, 23, 11732–11738