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Mercury (Hg) transformation pathways: Evaluating Hg incubation experiments in seawaters using isotopic tracers

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Lately, a great effort has been made to develop novel coupled biogeochemical Hg models to evaluate the effectiveness of the Minamata Convention. Biogeochemical models rely on experimentally determined transformation rates. However, rates reported in the literature vary over orders of magnitude [1]. Large methodological differences among studies such as (i) the concentration of added spike, (ii) lack of replicate incubations, (iii) different incubation times, (iv) no detailed kinetics, (v) exclusion of likely present Hg species from quantification (e.g. Hg(0)) and (vi) differences in analytical methods or data processing render the comparison difficult.

Herein, we present a protocol for the simultaneous quantification of Hg species (Hg(II+), MMHg, MeHg, Hg(0)) which subsequently allows the calculation of potential (de-)methylation and net reduction rates. Seawater incubations are performed in sextuplicate at sub-ambient spiking concentrations ($MM^{201}Hg$ 0.2 ng L^{-1}, $^{199}Hg(II+)$ 2 ng L^{-1}) in PFA/FEP bottles under natural light/dark conditions. Discrete incubation reactors are sacrificed at chosen time points (triplicates) and immediately acidified. The remaining samples are purged on gold-coated sand traps to collect Hg(0) before acidification. The obtained solutions are analyzed by double-double isotope dilution analysis with GC-ICP-MS and gold traps by cryo-GC-ICP-MS.

Experiments were conducted in October 2020 on coastal (sub-)surface waters collected at the Endoume marine station and off-coast near Île-Riou (Marseille, Mediterranean Sea). Methylation was not observed during our experiments, contrasting to demethylation under natural light conditions for both waters (1.87 ± 0.85 to 2.9 ± 0.88 % h^{-1} (n=6)) and dark demethylation for shore waters (0.44 ± 0.49% h^{-1} (n=5)). Moreover, our results demonstrate, that after a short incubation time (light: 2h, dark: 8h), a significant fraction of Hg (around 25%) is present as Hg(0). Mass balance calculations indicate that Hg(0) is re-oxidized in non-purged samples. This clearly demonstrates that transformation rates determined from experiments where headspace has not been properly controlled or Hg(0) has not been determined, need to be revisited.

In conclusion, a consistent and systematic approach for incubation experiments is urgently needed, to produce robust and comparable transformation rates for global biogeochemical models and further understand Hg transformations.

[1] Batrakova et al. (2014), *Ocean Sci.* ***10****, 1047-1063.*