MOLECULAR ORGANIC MATTER COMPOSITION CONTROLS MERCURY METHYLATION IN BOREAL LAKE SEDIMENTS

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Introduction

One major challenge in contemporary environmental science is to identify factors controlling the formation of methylmercury (MeHg). The methylation of inorganic-Hg to MeHg is biotically mediated[1] in aquatic systems and occurs mainly in oxygen-deficient sediments or water columns[2,3]. Organic matter (OM) interacts very strongly with Hg, affecting its speciation, solubility, mobility, and toxicity in the aquatic environment. In aquatic ecosystems, OM is an extremely heterogeneous mixture derived from a combination of terrigenous and planktonic derived OM. Terrigenous OM, resistant to microbial degradation, might be an important vector for Hg and MeHg transport from catchment soils to surface waters[4]. In contrast, labile planktonic-derived OM fuels microbial growth and activity in both water columns and sediments. In this study we test whether planktonic derived OM compounds control Hg methylation rates in boreal lake sediments.

Methods

We collected sediments from 10 boreal lakes with different trophic states and loadings of terrigenous OM. Potential Hg methylation rate constants (k_m) were determined in surface sediments (0-1 and 1-2 cm) using enriched isotope tracers[5]. We concomitantly characterized the molecular composition of the sediment OM by a pyrolysis–gas chromatography/mass spectrometry (Py-GCMS) method[6]. We conducted additional laboratory incubation experiments to assess the relative importance of three OM sources, with differentiated molecular OM compositions, on the k_m of two lakes that presented low k_m. The substrates consisted in an algae, cyanobacteria and a humic soil extract. Briefly, the cyanobacteria *Aphanizomenon flos-aquae PCC 7905*[7] and the algae *Scenedesmus*[8] were grown in the laboratory. An organic peat soil typical for the discharge area of a boreal forest catchment in northern Sweden was used[9].

Results

The highest k_m values were found in lakes dominated by planktonic derived OM (0.038–0.075 day^{-1}, n=4). Lower values were observed in lake sediments enriched in terrigenous OM (0.0095–0.013, n=5) or in invertebrate chitin associated compounds (0.013, n=1). Bacterial production rate (BP, µg C L^{-1} d^{-1}) in sediments dominated planktonic and chitin derived OM was significantly higher than in sediments characterized by terrigenous OM (p-value<0.001). We identified and quantified 110 organic compounds in all studied lakes. An orthogonal partial least square model (OPLS) built with both k_m and BP as Y variables showed that the abundance of planktonic-derived OM (chlorophyll, protein, and cell wall lipids) predicted the variability in both the k_m and BP. However, the second predictive component suggested that among the planktonic-derived compounds, Hg methylation rate constants would be higher in the presence of fresh chlorophyll compounds and cell wall lipids.

All substrates added to lake sediments boosted k_m although the net response differed between the two lakes, likely due to the differences on in habitant microbial communities. While we did not observed significant differences between the addition of the algae and the humic soil, we saw that in both lakes the presence of cyanobacteria led to an enhanced Hg methylation in lake sediments. Incubations experiments demonstrated that planktonic derived OM, concretely that derived from cyanobacteria, enhances k_m in lake sediments by enhancing the activity of the overall bacterial community. Bacterial community composition and Hg methylating bacteria (*hgcA*) composition varied among lakes independently of k_m.

Conclusions

A deep characterization of molecular OM composition is needed for understanding Hg methylation processes in aquatic systems. Besides cyanobacterial blooms produce a wide range of potent toxins, the results of this study show that they can also increase MeHg formation in aquatic systems, both with adverse health effects on humans.

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