MOLECULAR ORGANIC MATTER COMPOSITION CONTROLS MERCURY METHYLATION IN BOREAL LAKE SEDIMENTS

Andrea G. Bravo1,*, Sylvain Bouchet2, Julie Tolu3, Alejandro Mateos-Rivera1, Sari Peura1, Moritz Buck1, Erik Björn2, Stefan Bertilsson1

1Department of Ecology and Genetics, Limnology, Uppsala University, SE-75236 Uppsala, Sweden.
2Department of Chemistry, Umeå University, SE-90187 Umeå, Sweden.
3Department of Ecology and Environmental Science, Umeå University SE-90187 Umeå, Sweden.
andrea.garcia@ebc.uu.se

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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 mediated1 in aquatic systems and occurs mainly in oxygen-deficient sediments or water columns2,3. 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 waters4. 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 tracers5. We concomitantly characterized the molecular composition of the sediment OM by a pyrolysis–gas chromatography/mass spectrometry (Py-GCMS) method6. 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 79057 and the algae Scenedesmus8 were grown in the laboratory. An organic peat soil typical for the discharge area of a boreal forest catchment in northern Sweden was used9.

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.

**References**