

# Speciation of ultra-trace mercury(II) in natural waters: Insights from a DNA-based biosensor and hydrochemical modeling

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An understanding of the speciation of aqueous mercury(II), in addition to its total concentration, is of vital importance to elucidate the mobility, cycling and toxicity of mercury (Hg) in the environment [1], and assess the risks to ecosystems and drinking water sources. Research on Hg biogeochemistry, however, is hindered by the lack of sensitive and reliable detection methods that can measure trace-level Hg(II) directly on site and in situ. Unravelling the fate of Hg in remote regions also calls for field-deployable sensing tools that are easy to operate and can reveal Hg(II) speciation in variable aquatic environments [2].

In a previous study [3], we presented a DNA-DGT sensor for aqueous Hg(II) that combines the DNA-functionalized biosensing material with the Diffusive Gradients in Thin Films (DGT) technique. Here we show that the sensor can detect ultra-trace Hg(II) concentrations in hydrochemically diverse groundwaters and surface waters by adjusting the deployment time of the sensor. Quantification of aqueous Hg(II) speciation with the sensor, combined with temperature-calibrated hydrochemical models, indicates that the (in-situ) water temperature and dissolved organic matter (DOM) concentration strongly affect the partitioning of aqueous Hg(II) between inorganic and organic species, which in turn affects its bioavailability. Furthermore, DNA-DGT sensor data, when interpreted along with additional results from laboratory-based hydrochemical analyses, suggest that the mobilization of Hg(II) is linked to the biogeochemical cycling of sulfur in groundwater systems.

[1] Ullrich *et al.* (2001) *CREST* **31**, 241-293. [2] Douglas *et al.* (2012) *ENVIR CHEM* **9**, 321-355. [3] Pi *et al.* (2020) *ES&T* **54**, 13680-13689.