

# Simultaneous investigation of the biogeochemistry of selenium and mercury compounds in natural waters of a coastal range watershed: Analogues or Antagonists?

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While the speciation of redox reactive and volatile trace elements is a major concern in all aquatic environments, no studies have considered simultaneously selenium (Se) and mercury (Hg) cycles in waters and their potential coupling. Se and Hg in aquatic systems exhibit both inorganic oxidized, methylated and reduced gaseous compounds and their interaction may have further biogeochemical and environmental “health” implications. To better constrain how Hg and Se aquatic biogeochemistry evolves from mountain headwaters to downstream end-member, investigations were conducted (2017-19) in mountain lakes (Western Pyrenees) and in the Adour river and estuary (Bay of Biscay). In lakes, annual sampling was carried out in spring and fall, while in the Adour, seasonal sampling was performed in spring, fall and winter. Se (TDS<sub>Se</sub>, Se(IV), Se(VI), gaseous Se-TVSe) and Hg (THg, iHg, MMHg, gaseous Hg-DGM) speciation were determined in water samples for different locations and depths. In upstream lake waters, total dissolved Se (TDS<sub>Se</sub>) ranged from 7-80 ng.L<sup>-1</sup>, mainly as Se(VI) in most cases (63% TDS<sub>Se</sub>), and was correlated with sulfate concentrations. Se(IV) and TVSe were low (TVSe: 3-120 pg.L<sup>-1</sup>). While iHg(II) (0.11–1.19 ng.L<sup>-1</sup>) did not show significant seasonal variations, MMHg (<0.03–0.062 ng.L<sup>-1</sup>) was significantly higher due to biotic methylation in anoxic bottom waters. DGM (20–680 pg.L<sup>-1</sup>) and exhibit high variations levels (photoreduction). In river and estuarine waters, TDS<sub>Se</sub> concentration varied from 71-771 ng.L<sup>-1</sup> and correlate with nitrate levels. Se(VI) was the main species (50% TDS<sub>Se</sub>), while Se(IV) and reduced Se fraction averaged 11% and 42% TDS<sub>Se</sub>. TVSe ranged from 51-2757 pg.L<sup>-1</sup>. Total and dissolved concentrations of inorganic mercury (IHg) and methylmercury (MeHg) are rather homogeneous (Total IHg: 0.51–3.42 ng.L<sup>-1</sup>; MeHg: 0.03–0.08 pg.L<sup>-1</sup>) and depends on suspended particulate matter and organic carbon content. Overall, significant differences were observed between Hg and Se species distribution and fate among the investigated water bodies that are

closely related to their seasonal dynamics such as water discharge, thermal stratification and biological productivity. In our case study, Hg distribution seems better controlled by regional atmospheric inputs, while for Se, local geogenic, atmospheric and agricultural land use inputs are all significant.