Speciation and fate of Hg and Se species in the water column of high altitude lakes (Pyrenees)

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While the speciation of redox reactive and volatile trace elements is a major concern in all aquatic environments, very few studies consider Mercury (Hg) and Selenium (Se) cycles in remote alpine lakes. This work presents an integrated investigation conducted in high altitude pristine lakes from Western Pyrenees (lakes Gentau, Arratille, Sabocos, Azul Superior). Sampling and experiments were performed in June and October 2018. Hg (iHg, MMHg, gaseous Hg-DGM) and Se (Se(IV), Se(VI), gaseous Se) speciation were determined in water samples collected at different depths for each season. Besides, in situ incubation experiments using isotopically enriched Hg species (^{199}iHg, ^{201}MMHg) were conducted to investigate Hg transformation mechanisms in the water column (methylation, demethylation, reduction). While iHg (87-2811 pg/L) showed various seasonal patterns, MMHg (5-426 pg/L) was significantly higher in autumn, except for bottom anoxic waters (i.e. Gentau, Sabocos). Dissolved total Se (11-57 ng/L) was significantly lower in autumn all along the water column. Se was mainly present in Se(VI) form (15-116 $\%Se_{TOT}$; Se(IV)<4 ng/L) except in Gentau bottom waters where reduced Se is predominant. DGM (25-4646 pg/L) emphasized the importance of photoreduction as maximum concentrations occurred around midday, while DMSe remains stable and the predominant volatile Se specie. Hg incubation experiments highlighted an important demethylation yield in surface waters of Gentau (ca. 40%/day, both seasons). In bottom waters, methylation rates were about 2.5 and 0.7%/day (both seasons) respectively for Gentau and Sabocos (<0.04%/day in Arratille). Overall, more reductive and heterotrophic conditions are generally occuring in the hypolimnion of stratified lakes leading to significant changes in Hg and Se speciation (i.e. increasing methylated and reduced species). Due to intense UV radiations, significant photodemethylation and photoreduction of Hg occurs in surface waters of the alpine lakes, while photodegradation of volatile Se compounds must be assessed. The large differences found for Hg and Se speciation and fate between investigated lakes are closely related to their seasonal dynamics including stratification and trophic status.