EGU2020-19867
https://doi.org/10.5194/egusphere-egu2020-19867
EGU General Assembly 2020

Determination of gaseous elemental mercury air-sea exchange in the Baltic Sea

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Ocean waters store approximately 400 Gg of mercury (Hg) and exchange it with the atmosphere at a high rate. Air-sea exchange of gaseous elemental mercury (Hg^0) is a key process in global Hg cycling because evasion lowers the reservoir of Hg(II) available for methylation and subsequent bioaccumulation in marine fish and prolongs the atmospheric lifetime and subsequently global cycling of Hg. However, global estimates on the air-sea flux are not well constrained (1.9 to 4.2 Gg a^{-1}) mainly because high-resolution measurements of Hg^0 in seawater are largely lacking and parameterization of the Hg^0 transfer velocity introduces uncertainties in Hg^0 flux modelling. We present estimates of the net Hg^0 flux for the Baltic Sea derived from land-based marine measurements of Hg^0 in air and seawater as well as micrometeorological techniques. We found that coastal waters at the ICOS field station Östergarnsholm, located east of Gotland, Sweden, were typically supersaturated with seawater Hg^0 (mean ± SD = 13.5 ± 3.5 ng m^{-3}; ca. 10 % of total Hg) compared to ambient Hg^0 (1.3 ± 0.2 ng m^{-3}). The Hg^0 flux calculated using gas-transfer wind speed relationships ranged from 0.1 to 1.3 ng m^{-2} h^{-1} over the course of the campaign (May 10 – June 20, 2017). The modeled Hg^0 flux showed a distinct diel pattern with an average daytime flux of 0.6 ng m^{-2} h^{-1} and nighttime flux of 0.4 ng m^{-2} h^{-1}, indicating that temperature and light induced production of seawater Hg^0 was of significance in shallow waters. Preliminary calculations of the average coastal Hg^0 flux simultaneously measured using direct, non-intrusive gradient-based, aerodynamic gradient and relaxed eddy accumulation techniques were 0.5 ± 1, 0.6 ± 3.8 and 0.6 ± 37 ng m^{-2} h^{-1}, respectively. Although, these flux estimates were in good agreement, there were indications in the temporal patters of the observations, which suggest that there is a need to reconsider the modeled flux with the support of more direct flux measurements. Direct flux measurements revealed not only Hg^0 evasion but also periods of Hg^0 dry deposition. In addition,

direct measurements indicated a stronger wind speed dependence of the Hg^0 transfer velocity compared to CO_2 which appears to coincide with whitecap formation in the open sea flux footprint (wind speed > 5 m s^{-1}). Hence, we anticipate this study as a starting point for more land-based, marine, continuous measurements of seawater Hg^0 concentration in combination with micrometeorological fluxes in order to improve Hg^0 flux estimates in regional and global scale models. In this context, directly measured Hg^0 fluxes will be pivotal to improve transfer velocity estimates of Hg^0 especially during periods of high wind speed.