Spatial distribution of per- and polyfluoroalkyl substances (PFAS) in natural and restored intertidal wetlands in the Scheldt estuary

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Due to high human activity in coastal ecosystems, coastal and estuarine wetlands have become one of the most threatened and used natural ecosystems on the planet, with a significant decline in their ecosystem services as a result. Many of these ecosystems are impacted by long-term exposed to pollutants, including per- and polyfluoroalkyl substances (PFAS), coming from various sources. However, the environmental fate and behaviour of many pollutants in these habitats are poorly understood. Especially intertidal areas, which flood at high tides and drain at low tides, are expected to be at risk because tidal supply and deposition of pollutants is a predominant process in these areas. The three eco-geomorphic zones, i.e. tidal marshes, flats and channel networks, of estuaries each have their own distinct hydrodynamics, sedimentology and ecology which may affect the environmental fate and behaviour of pollutants. To gain better insights in the environmental fate and behaviour of PFAS in these ecosystems, we investigated the spatial distribution of PFAS in sediments of intertidal areas in the Scheldt estuary, and to what extent this varies between different eco-geomorphic zones and between an old natural intertidal site (drowned land of Saeftinghe) and recently restored intertidal site (Hedwige-Prosperpolder; HPP), where tides were re-introduced. Furthermore, we investigated whether this distribution is affected by sediment characteristics and distance from the estuarine main channel. Finally, we compared PFAS levels prior and after re-introduction of tides in the restoration site.

Eleven PFAS were detected in at least one of the surface sediment samples from Saeftinghe. PFAS profiles were often dominated by PFBS and 6:2 FTS. The concentrations and profiles did not show clear patterns related to specific zones nor with distance from the estuarine main channel, but overall, higher concentrations were observed in the vegetated tidal marshes. PFAS concentrations in the surface sediments were not correlated to the sediment characteristics, whereas PFBA, PFOS, and 6:2 FTS concentrations in sediment core samples were positively correlated to clay content. Prior to re-introduction of tidal flooding in the Hedwige-Prosperpolder, only PFBS, PFOS and PFOA were detected in the surface sediments. After re-introduction, five additional PFAS (6:2 FTS, PFNA, PFDA, PFDoDA, and PFTrDA) were also detected. Concentrations of PFBS, PFOS and PFOA were respectively 5x, 12x, and 3x higher after re-introduction, although only the increase of PFOS was significant ($p = 0.034$). Before re-introduction the concentrations were not related to distance from the main channel, but after re-introduction significant differences were observed for PFOA ($p = 0.029$) and PFBS ($p = 0.024$). For both PFAS, the concentrations were highest closest to the main channel. Prior to re-introduction, the PFAS concentrations were not correlated to the sediment characteristics, but afterwards the concentrations of PFOA were positively correlated to clay and OM content, whereas those of PFNA and PFTrDA were respectively negatively and positively correlated with sand content.

Our results show signs of both historical and recent PFAS emissions, and we hypothesize that the vegetation and the higher intertidal elevation in the marshes slow down currents, causing more deposition of finer sediment and higher PFAS concentrations. Distance from the estuarine main channel, grain size and organic matter content were less determining factors in the environmental fate. The re-introduction of tidal flooding in HPP leads to an enrichment with PFAS. Although environmental risks in the intertidal areas were not assessed in this study, the enrichment could be beneficial for the Scheldt estuary and North Sea, as PFAS are filtered from the estuary and ecological risks in the estuary and adjacent sea are likely reduced. Moreover, removal of contaminated sediment beds and plants from intertidal areas would allow for remediation of the ecosystem, something which is more difficult when the pollution stays in the estuarine channel or sea.

Keywords
Eco-geomorphology; Intertidal Areas; PFAS; Sediment