

Probing Organic Carbon Associations with Iron Oxide and Clay Minerals in Sediments of the St. Lawrence Estuary (Canada) Using Chemical Fractionation, Stable/Radiocarbon Analysis as well as Integrated Machine Learning and Multivariate Analysis

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Marine sediments serve as a major sink of organic carbon (OC) on the planet, with the bulk of this OC being sequestered within coastal and deltaic sediments where terrestrial and marine OC are deposited in different proportions along the fresh- to seawater continuum. The long-term storage of reduced organic compounds within these environments helps maintain the global redox balance at the surface of the planet and constitute an important removal vector for atmospheric CO₂. As dominant constituents of the sediment mineral matrix, redox-sensitive nanophase iron oxides and aluminosilicate clay minerals play an important role in the preservation of sedimentary OC through the formation of mutually protective organo-mineral associations. Despite the importance of these associations in the balance between the preservation and oxidation of OC in coastal systems, very little is known about the type of OC most likely to be found associated with these distinct minerals, and to what extent the formation of these associations takes place *in situ* within the sediment or on land before minerals are delivered to the sediment. We used a chemical fractionation scheme developed in our laboratory to quantify and characterize OC in four different fractions in a series of sediment samples collected along the St. Lawrence Estuary and Gulf continuum: (i) total OC, (ii) OC associated with iron(III) oxides; (iii) OC associated with clays; and (iv) residual insoluble OC not associated with minerals. At each of the seven stations, results for surface sediments (0-5 cm) were compared to those obtained for diagenetically stabilised sediment (26-31 cm). Isotopic mass balance calculations allowed apportioning OC among the different fractions, and each fraction was characterized through elemental (C and N), isotopic ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, $\Delta^{14}\text{C}$), spectroscopic (FTIR), microstructural (pXRD, XPS, BET surface area) and microscopic (SEM) analysis. Preliminary results show varying proportions of OC found in each fraction, with differences along both the temporal (surface vs. deep) and spatial (distance from land) dimensions. Our results shed light on the complex interplay between OC and minerals in coastal sediment environments, highlighting the need for further