A combined retrospective and spatial analysis of PFAS in river sediments demonstrates changes in both levels and patterns over the last 40 years (Rhône River, France).

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Per- and polyfluoroalkyl substances (PFAS) are commonly detected in aquatic systems and can be transported across long distances. Concentration levels and patterns of PFAS depend on various factors, including the type of matrix analyzed and the relative contribution of direct and indirect sources. Long-chain perfluoroalky carboxylic acids (PFCA) and sulphonic acids (PFSA) are persistent and more hydrophobic than short-chain PFAS, which may lead to their accumulation in specific compartments such as sediments. Sediment can act as a secondary source of contamination, making this a potential longterm problem. Bed sediments and dated sediment cores were collected in the Rhone River, from both upstream and downstream locations of Lyon (France) and its industrial corridor, to evaluate the spatial and temporal trends of PFAS contamination. Total concentrations of PFAS in upstream sediments were low, ranging between 0.19 and 2.6 ng g-1 dry weight (dw), with a high proportion of perfluorooctane sulfonate (PFOS). Downstream, concentrations reached up to 48.7 ng g-1 dw which is likely due to the presence of a fluoropolymer manufacturing plant in the area. Further downstream, concentrations gradually decreased. The temporal evolution of PFAS emissions was analyzed using three sediment cores sampled from secondary channels located 6 to 42 km downstream from the hotspot. Before 1987, PFAS concentrations were low (2 ng g-1 dw), peaking at 51 ng g-1 dw in the 1990s and high until 2007. Since 2010, trends have been stable with total PFAS concentrations of 4.2 ±1.4 ng g-1 dw near the production site and 2.1 ± 1.0 ng g-1 at the most downstream site. The proportion of PFSA remained constant since the 1980s (~10%), while PFCA showed large variations. Long chain- (C >8) PFCA with an even number of perfluorinated carbons represented 74% of the total PFAS load until 2006. From 2006 to 2020, the relative contribution of long chain- (C > 8) PFCA with an odd number of perfluorinated carbons increased to 80%. The changes observed in the PFAS profiles over time indicate a significant modification in the industrial production process. This study emphasizes the necessity of an improved monitoring of these substances in aquatic systems.