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# Close deciphering of the sediment record of stable and radioactive contamination in the upper Seine estuary, over the last 50 years: a phosphogypsum legacy

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The Seine estuary is the outlet of a huge catchment area (the Basin of Paris), into which fine sediments and anthropogenic elements and substances end in. In the upper Seine estuary, stable and radioactive contaminants come both from upstream and from local in the industrialized agglomeration of Rouen. It is particularly challenging to decipher the relative contribution of these two sources due to the lack of historical data for most of the contaminants of concern. The key issue was the collection of two sediment cores: (1) in a flood plain of the lower Seine River and (2) in an old careening basin of the Rouen harbour which was not dredged since many decades. These cores were carefully dated using artificial radionuclide (atmospheric fallout from Chernobyl and nuclear weapon tests – 137Cs - or discharges in the river - 238Pu) and signal processing techniques and an accurate model age was obtained, yielding a 50 years long sediment record. Major elements, 44 trace elements and 14 naturally-occurring radionuclides (235U, 238U and 232Th series) were measured by ICP-AES, ICP-MS and gamma-spectrometry.

As some elements display no almost constant (Co, Cs, Ga, Ge, Hf, Nb, Rb, Sr, Ta, V) or continuous decreasing concentrations (Ag, As, Pb, Sb, Sn, W, Zn) since the early 60's, some others show much higher concentrations (Ba, Be, Bi, Cd, Cr, Cu, Mo, Ni, Y and lanthanides, U and, to a lesser extent, Th series) from mid-60's to mid-80's, and only in the core collected in the Rouen harbour. Even if some minor contributions cannot be ruled out, it is likely that these huge excess concentrations (up to 50 times) are due to the phosphate fertilizer industry. In fact, between 1960 and 1984, the upper Seine estuary underwent big direct discharges of phosphogypsum (an acidic by-product of the process) from two plants located in the vicinity of the Rouen harbour, which peaked up to 1million tons per year in 1974. Cd and Bi are, from far, the most enriched stable elements (up to 300 times relatively to shale-type material or geochemical background). The activities of 226Ra and some of its daughter elements can exceed 1500 Bq.kg-1 in 1974, namely ca 50 times those expected for shale-type material.

By combining the concentrations of these elements as observed in the upstream core and the annual solid discharge of the Seine River, total annual contaminant input from the catchment area can be derived. In the same way, average phosphogypsum concentrations calculated from excess concentrations combined with annual phosphogypsum discharge lead to the total annual contaminant input from the fertilizer industry. The difference between observed excess concentrations and those expected from annual phosphogypsum discharge are interpreted in terms of residence time of sediment particle in the upper Seine estuary.