

## **Spatial and temporal (30 yr.) variations of lead geochemical signature in a macrotidal highly polluted estuary**

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Two dated sediment cores (<sup>210</sup>Pb) were sampled in 2005 on tidal flats of the Scheldt estuary at the vicinity of the heavily industrialized harbour city of Antwerp. Coring was close to the estuarine turbidity maximum and in the reactive 0-10 psu zone. The cores were compared with suspended particulate matter (SPM) and surface sediments (SS), sampled between 2002 and 2006 from the Scheldt river to the southern bay of the North Sea. Pb isotopic compositions measured by MC-ICP-MS were compiled regarding to grain-sized distributions as well as sedimentological, mineralogical and geochemical data. The present study aims at characterizing the sources and the evolution of Pb inputs during last 30 yr. in that complex environment.

As a whole, Pb isotopic data for SPM, SS and cores from the Antwerp area show a narrow range of variations. <sup>206</sup>Pb/<sup>207</sup>Pb data vary of 1.9 ‰ (RSD) from the mean value of 1.1629 (with 1.1630 for SS and SPM, and 1.1628 for cores) suggesting that ~70% of particulate Pb has an anthropogenic origin. A close look at the Pb isotopic ratios highlights systematic variations reflecting different controlling mechanisms. In the SPM and SS samples, the larger Pb isotopic variations (from  $1.1582 \pm 0,000010$  -2sd- to  $1.1697 \pm 0,000017$ ) are related to daily tidal cycles, where the relative contribution of fluvial and marine waters controls the isotopic signature. In the cores, the relative lower variation range (from  $1.1608 \pm 0,000011$  to  $1.1646 \pm 0,000007$ ) may result from a larger time-series integration and homogenization of the surface layers by bioturbation. Pb isotopic variations are correlated with grain-sized distributions, that are controlled by changes in deposition modes, mainly related to neap tide/spring tide cycles. In contrast, the decrease in Pb concentration, recorded from the 75-80's period to 2005, does not imply any significant isotopic answer. Therefore, Pb sources in the Scheldt estuary may remain unchanged, in component and relative contribution, over 30 yr. The water quality improvement, in terms of Pb contents, is not related to the removal of the leaded gasoline, but only to industrial emission control policies and establishment of water treatment plants.