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Lead isotopic composition of sediments from the Scheldt estuary (SW-Netherlands - Belgium).

A. Taillez, J. Petit, S. Verheyden, L. Chou, L. Rebreanu, C. Maerschalk and N. Mattielli.

Universite Libre de Bruxelles, Department of Earth and Environmental Sciences, Brussels, Belgium.

Pb isotopes are now well-known to be used, in environmental studies, as a powerful tool for tracing Pb origins. A large variety of materials (atmospheric aerosols, lake sediment, peat, ice cores, ...) have already been investigated, but riverine and estuarine systems remain significantly less documented. However, estuaries constitute key area, located at the interface between continental freshwaters and marine waters, and characterized by very dynamic biogeochemical and physical properties (contrasting with the state of quasi-equilibrium of oceans and lakes). Estuarine problematic can be summarized by two major characteristics: (i) the long residence time of both waters and particles and (ii) the multiplicity of the particulate matters sources. This is particularly true for the macrotidal estuaries such as the Scheldt estuary that shows the additionnal specificity to be one of the more polluted estuary in particulate trace metals of the W-Europe. The purpose of the present study is to characterize the variations in Pb isotopic composition of subsurface sediments of the Scheldt estuary, and to identify relevant Pb pollution sources. 40 sediment samples were collected during 6 cruises (on the Belgica vessel) covering all seasons and showing contrasted hydrological conditions. After a classical digestion of the samples using sub-boiled HF-HNO3-HCl in Savilex teflon beakers, Pb fractions were obtained by a one step-chromatographic separation (Weis et al., 2005). Pb isotopic compositions were analysed on a Nu Plasma MC-ICP-MS at ULB. Pb/Tl ratios for the samples equalled 5, reproducing the Pb/Tl value of the standards. The total Pb ion beam intensities varied from 20 to 40 V/ppm. During the analysis sessions, 44 analyses of the NBS981 Pb standard gave mean values of 208Pb/204Pb = 36.7151 +/- 27 (2SD), 207Pb/204Pb = 15.4967 +/- 10 (2SD) and 206Pb/204Pb = 16.9401 +/- 11 (2SD).

As a whole, 208Pb/204Pb, 207Pb/204Pb, 206Pb/204Pb, 208Pb/206Pb and 207Pb/206Pb range from 37.9196 to 38.4582 +/- 33 (2SD), 15.5894 to 15.6329 +/- 12, 17.9914 to18.4823 +/- 15, 2.07479 to 2.10769 +/- 7 and 0.84538 to 0.86650 +/- 2, respectively. Pb isotopic ratios of the sediments show a positive correlation between 206Pb/207Pb and 208Pb/207Pb and describe a positive linear trend with the salinity. These observations suggest a main control of the Pb isotopic signatures of the Scheldt sediments, as well as Pb concentrations, by the relative contributions of marine and fluvial inputs. This is consistent with interpretations of Petit et al. (2005) obtained on Cu and Zn isotopes data. In order to better constrain the isotopic signatures of our fluvial and marine input endmembers, our isotopic data have been compared with literature data. Fluvial signature seems to be largely influenced by the contribution of an anthropogenic component: treated and untreated urban wastes (206Pb/207Pb ranges from 1.147 to 1.161) and, to a lesser degree, airborne particles from Western Europe (1.1342 - 1.1634). Marine signature overlaps the isotopic field of oceanic sediments (for Atlantic sediments 206Pb/207Pb ranges from 1.201 to 1.219). In details, Pb isotopic ratios show larger variations with time at the upstream stations. These variations are likely due to seasonal variability of the hydrological conditions (freshwater flow): in winter, high river discharges tend to displace longitudinally the area under influence of anthropogenic inputs to the middle part of the estuary. In contrast, Pb isotopic compositions are relatively constant in the middle part of the estuary (from the Estuarine Turbidity Maximum -ETM- to the mouth). Both the 208Pb/207Pb vs. salinity profile and the mixing plot (inverse of Pb concentration vs. the 208Pb/207Pb) suggest two additional sources especially active in the middle estuary. Those latter recorded in the ETM (208Pb/207Pb = 2.4569 and 2.4543) are probably related to anthropogenic wastes of the Antwerp city (km 20). Unusual Pb isotopic compositions at the mouth (208Pb/207Pb = 2.4487 and 2.4506) could be associated with either the Ghent city anthropogenic activities via the Ghent-Terneuzen Canal (km 80), or the input in the Scheldt estuary of polluted coastal water masses flowing from the Rhine/Meuse estuary (Lacroix et al., 2004). Further investigations will be performed to better constrain the origin and relative contribution of those Pb pollution sources.