

# **Tin and organo-tin species along the Gironde fluvial-estuarine continuum: spatial distribution and solid/liquid partitioning during two contrasting hydrological conditions**

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Butyl-tin (BTs) compounds, especially tributyltin (TBT), were banned forty years ago and are classified as priority substances under the EU Directive (2008/105/EC) due to their high toxicity for aquatic organisms. Despite various past applications including antifouling agents, TBT and its degradation compounds, dibutyltin (DBT), monobutyltin (MBT) and inorganic tin (Sn), are still found nowadays in aquatic environments. This study aims at determining for the first time the current state of BTs distribution in a major European macrotidal system (the Gironde Estuary), by understanding the geochemical behaviour and current potential sources of BTs. Dissolved and particulate BTs and Sn were extensively quantified along the estuarine salinity and turbidity gradients during two oceanographic campaigns (MGTS I and MGTS III) carried out in contrasting hydrological situations (March 2014,  $1203 \text{ m}^3 \cdot \text{s}^{-1}$ ; October 2015,  $248 \text{ m}^3 \cdot \text{s}^{-1}$ ). Target samples including bottom sediments in five local harbours located downstream of the estuary, outflowing solids from Louis Fargues WWTP and a mid-estuary sediment core at Macau cove were also analysed. Results showed (i) positive anomalies for dissolved BTs concentrations ( $\text{BT}_{\text{d}}$ ), ranging between 0.8 and  $41 \text{ ng} \cdot \text{L}^{-1}$  at high salinities ( $S > 20$ ), with  $\text{DBT}_{\text{d}}$  and  $\text{MBT}_{\text{d}}$  showing concave/non-conservative distributions along the entire salinity gradient during both hydrological conditions (10-fold higher during flood), (ii)  $\text{BT}_{\text{sp}}$  concentrations ranging between 0.8 and  $35 \text{ ng} \cdot \text{g}^{-1}$  along the salinity gradient, showing highest  $\text{TBT}_{\text{p}}$  concentrations of  $1.7 \text{ ng} \cdot \text{g}^{-1}$  at high ( $12 \text{ g} \cdot \text{L}^{-1}$ ) suspended particle matter (SPM) concentrations during draught (MGTS III), (iii) increases of  $\text{Sn}_{\text{d}}$  concentrations from 10 to  $155 \text{ ng} \cdot \text{L}^{-1}$  with SPM, and (iv)  $\text{Sn}_{\text{p}}$  concentrations ranging between 5.4 and  $16 \text{ } \mu\text{g} \cdot \text{g}^{-1}$  with slight increases at the estuary mouth during both campaigns when referred to the regional geochemical background ( $\text{Sn}/\text{Th}=0.55$ ). Solid/liquid partitioning along the degradation products, from TBT to Sn, decreases from  $\log_{10} K_{\text{d}} \sim 2$  to  $\sim 6$  suggesting an increasing affinity to the particle phase. Residual organotin concentrations found in water, suspended matters and bottom sediments suggest that both, secondary and ongoing sources linked to anthropogenic activities (i.e., industrial, urban, dredging) are still present today along the Gironde Estuary.