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Tracking Zn and Pb isotope signals from antifouling paints in coastal environments

BRUNO CUNHA[1], JÉRÉMIE GARNIER[2], DANIEL FERREIRA ARAUJO[3], MYLLER TONHÁ[4], WILSON MACHADO[5], MARCOS FERNANDEZ[6], CARLOS EDUARDO SOUTO DE OLIVEIRA[1], IZABEL RAMOS RUIZ[1] AND **MARLY BABINSKI**[7]

[1]Universidade de São Paulo
[2]Universidade de Brasilia
[3]IFREMER
[4]Universidade de Brasília
[5]Universidade Federal Fluminense
[6]Universidade Estadual do Rio de Janeiro
[7]Instituto de Geociências - Universidade de São Paulo

Presenting Author: babinski@usp.br

Trace metal contamination in marine ecosystems is an environmental issue of great concern on a global scale. Even those marine environments free from traditional pollution sources (urban and industrial effluent) are frequently exposed to metals derived from antifouling paints (AFP). Copper-based AFP (~30% w/w) also has Zn (up to 26 % w/w) and Pb in their composition to a lesser extent. In this work, we tested Zn and Pb isotopes to identify records of antifouling paints and quantify Zn and Pb contribution over time in a marina on the southeastern Brazilian coast (Rio de Janeiro State). This marina, built in 1978, has never been dredged and has no contribution from major anthropogenic contamination, such as sewage and industries. Three common AFP in Brazil analyzed in this study presented Zn and Pb contents about 12% w/w and 380 mg/kg, respectively, and $\delta^{66}Zn_{JMC}$ and $^{206}Pb/^{207}Pb$ values of +0.08‰ ± 0.04 2s and 1.19 ± 0.02 2s. Sediment core analysis discriminated the historic evolution: pre-marina moment ($\delta^{66}Zn_{JMC}$ +0.20‰ ± 0.02 2s and $^{206}Pb/^{207}Pb$ 1.41 ± 0.00 2s), the start of marina operation ($\delta^{66}Zn_{JMC}$ + 0.09‰ ± 0.02 2s and $^{206}Pb/207Pb$ 1.36 ± 0.00 2s) and the present day ($\delta^{66}Zn_{JMC}$ -0.02‰ ± 0.04 2s and $^{206}Pb/^{207}Pb$ 1.21 ± 0.00 2s). The $\delta^{66}Zn_{JMC}$ vs. $^{206}Pb/^{207}Pb$ variability reveals distinct Zn and Pb isotopic groups that may represent AFP influence in the background marina sediment signatures shifting towards AFP signals. This influence was calculated with a simple zinc binary mixing model, which shows that Zn contributions from AFP accounted for up to 89%. Our study uses Zn and Pb isotopes as a tracker of antifouling paints signals in coastal environments for the first time. While discriminating from the natural background is feasible, AFP's Zn and Pb isotopic signatures alone may overlap with other commonly known sources. Nevertheless, the multi-isotopic study proved to be a powerful tool to discriminate sources and may be used in complex systems with multiple anthropogenic pressures.