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Application of lead isotope ratios for pollution source investigation in the marine environment

Emiliya Vassileva, Anna Maria Orani, and Sergey Assonov
International Atomic Energy Agency, Environmental Laboratories

Lead is a non-essential toxic element that at high levels of human exposure causes damage to many organs of the human body. This element naturally occurs in the Earth crust, but its biogeochemical cycle has been altered by anthropogenic activities, which have introduced high amount of this element from different sources. Among inorganic contaminants, Pb is perhaps the most studied, but the determination of its total concentration only is not sufficient for a proper evaluation of contamination sources. Discrimination of anthropogenic and geogenic lead sources requires both precise and accurate isotope ratio determination as well as high versatility due to the complexity of environmental matrices, such as sediments, biota and seawater. This element has a partially radiogenic isotopic composition with 208Pb, 206Pb and 207Pb originating from the radioactive decay of 238U, 235U and 232Th respectively and 204Pb representing the only natural stable isotope. This characteristic isotopic composition represents a powerful analytical tool as it allows to trace the sources, fate and effects of possible Pb contamination. The most common way to express the Pb isotopic composition is using the ratio 206Pb/207Pb, because of the easy interference-free determination and isotopes' abundance. The determination of 204Pb by ICP-MS is quite challenging as this is also the least abundant among Pb isotopes (about 1.4%) and it is also affected by isobaric interference from 204Hg. The latter derives from both sample matrices and from plasma/sweep gas supplies and it represents a big analytical challenge, especially for marine biota samples, where the amount of Hg can be up to 100 times higher than Pb.

In this work we present the development and the application of analytical methodology for the accurate and precise determination of Pb isotope ratios by HR-ICP-MS in different marine environmental matrices (sediments, seawater and biota). Analytical procedures are involving a separation of Pb from the sample matrix and mercury, present in the sample. For seawater samples, the use of the SeaFAST automated system allowed simultaneous matrix separation and analyte pre-concentration before ICP-MS analysis. A comparison of results for lead isotope ratios obtained with MC-ICP-MS and HR ICP-MS in the same samples, in all cases, showed very good agreement . The total uncertainty associated to each result was estimated and all major contributions to the combined uncertainty of the obtained results were identified. As all such studies involve companions of different datasets, the uncertainty estimation is critical to ensure correct companions. The developed methodology was applied to different marine samples, namely sediments from Caribbean, Baltic and Namibian coasts, biota samples from French Polynesia, seawater samples from Mediterranean and Arctic seas.