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Transient Isotopic Variations of Pollutant Lead in the Mediterranean Sea

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The phasing out of leaded gasoline combustion in the EEC after the 1980's has provided a unique transient tracer in the Mediterranean Sea, variable both in deposition flux and source signatures. Variations in lead concentrations have been observed in the Mediterranean troposphere likely as a response to the reduction of lead emissions. However, uncertainties persist regarding the impact of this phasing out on lead cycling in Mediterranean seawater. Indeed, lead concentrations in seawater from different areas in the western Mediterranean display significant variations owing to the variability of emission sources, biological activity in surface waters and physical mixing. Stable lead isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb) are not fractionated by such processes in seawater. Here we report the first stable lead isotope ratios measured in the western Mediterranean seawater collected as part of the DYFAMED-JGOFS and MATER-EEC programs. The ^{206}Pb/^{207}Pb ratios measured in 1983 in the Gibraltar Straits show a clear contrast between the Atlantic inflow in sub-surface waters (1.179±0.001) and the Mediterranean outflow at depth (1.159±0.002). This is due to the more radiogenic composition of North American origin in the North Atlantic Central Basin. The ^{206}Pb/^{207}Pb of the outflow is close to that measured in the Ionian Sea (1.157±0.004), significantly different from the Ligurian Sea (1.147±0.002). This corroborates the larger contribution of the eastern basin to the Mediterranean outflow indicated by the hydrographic data. Considering lead concentrations and isotopic compositions in seawater collected from the western basin in 1995, we calculate that the isotopic signatures of the anthropogenic lead input vary from [^{206}Pb/^{207}Pb]ANTHR = 1.156±0.002 to 1.160±0.002 in the Gulf of Lions and the Ligurian Sea. The pollutant lead isotopic signature changed from [^{206}Pb/^{207}Pb]PL= 1.147±0.002 in 1983, to 1.160±0.002 in 1995 in the Ligurian Sea. We show that this is due to the relative increase of industrial versus automotive emissions resulting from the phasing out of leaded gasoline. These results allow us to determine an average industrial signa ture of 1.166. This radiogenic composition is consistent with recent measurements in industrial areas. The increase in ^{206}Pb/^{207}Pb ratio, and the decrease in [Pb] concentration are simulated with a four-box non-steady state model. New constraints are obtained from this simulation on the input function and the internal fluxes in the western Mediterranean sea.