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Does surface seawater actively trace the Pb isotope ratios of aerosol Pb? A case study of the low-dust South Pacific and Southern Oceans

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Seawater lead (Pb) is a valuable atmospheric flux tracer supplied to surface waters from both anthropogenic activities and lithogenic weathering. Here, we quantify the relative flux of natural and anthropogenic Pb to the South Pacific and Southern Ocean in the U.S. GEOTRACES GP17-OCE cruise using stable Pb isotope ratios. Surface water dissolved Pb (dPb) concentrations were extremely low (4-12 pmol/kg) and had distinctly anthropogenically-influenced $^{206}Pb/^{207}Pb$ that decreased from the northernmost South Pacific Gyre stations (~20°S, $^{206}Pb/^{207}Pb$=1.1733) to the Southern Ocean (south of 55°S, $^{206}Pb/^{207}Pb$ as low as 1.1575). We interpret this range of surface dPb isotopes as mixing between anthropogenic Australian Pb ($^{206}Pb/^{207}Pb$ <1.15) and lithogenic Australian, New Zealand, and/or Antarctic Pb ($^{206}Pb/^{207}Pb$=1.175-1.215). In contrast, the $^{206}Pb/^{207}Pb$ of aerosols collected simultaneously with the surface seawater were significantly more crustal than the surface seawater, whether the aerosol was totally digested ($^{206}Pb/^{207}Pb$=1.1879-1.2066) or partially leached with acetic acid-hydroxylamine-hydrochloride ($^{206}Pb/^{207}Pb$=1.1821-1.2102). Hence, partial solubilization of the labile aerosol Pb does not explain the more anthropogenic Pb isotope signatures of surface seawater. Instead, in the South Pacific and Southern Ocean at least, where aerosol inputs are quite low (0.1-2.4 pmol/m^3 measured at our stations), Pb inputs to the seawater circulation pathway during the residence time of 2-3 years matter more than *in situ* aerosol Pb deposition. While modeled seawater back trajectories are required to prove this, we hypothesize that south of the Polar Front (~55°S) in the Southern Ocean, upwelling of Upper Circumpolar Deep Water (UCDW) brings an anthropogenic-crustal mixed $^{206}Pb/^{207}Pb$ signature of ~1.16 to surface waters, which matches the signature observed in subsurface UCDW along the rest of the transect. HYSPLIT air mass back trajectories reveal Antarctic aerosol provenance south of 55°S, and the aerosol Pb fluxes are variable but likely too low to overcome the greater UCDW Pb inventories (and thus $^{206}Pb/^{207}Pb$). Farther north in the Subtropical and Subantarctic South Pacific (20-55°S), we observed a meridional trend of decreasing $^{206}Pb/^{207}Pb$ in both the surface seawater and the aerosols, consistent with less surface mixing and/or advection of dPb.