

# The Evolution of Anthropogenic Lead in the Ocean, 1976-2000

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The evolution of anthropogenic Pb in the ocean has now been directly observed for nearly 25 years, beginning with the pioneering North Pacific (1976) and North Atlantic (1979) deep-sea profiles of Pb by Schaule and Patterson. Schaule and Patterson demonstrated that Pb concentrations were high in the upper ocean and low in the deep sea, with North Atlantic Pb > North Pacific Pb > South Pacific Pb. They argued that this pattern was ample demonstration of the anthropogenic influence on oceanic Pb because the enrichments corresponded to the respective eolian fluxes and to the upwind consumption of leaded gasoline. A skeptic could point out that the same pattern of enrichment is observed for non-anthropogenic Al. The ongoing response of the ocean to decreases in the utilization of leaded gasoline by North America, western Europe, and Japan now provides overwhelming confirmation of the anthropogenic contribution to oceanic Pb. In the surface waters of the north Atlantic Ocean near Bermuda, Pb has decreased by fivefold, from ~160 pmol/kg in 1979 to ~30 pmol/kg at the end of 1999, and significant decreases in Pb are observed in subsurface waters down to ~1500m depth. In the eastern north Atlantic south of the Azores, surface Pb has decreased from ~130 pmol/kg (1981) and ~100 pmol/kg (1989) to ~40 pmol (1999), with subsurface decreases observed down to ~1200m. Mid-depth (~700m) waters now have higher Pb concentrations than above or below because of the lag due to the decadal ventilation rates. In the north Pacific Ocean near Hawaii, surface water Pb has decreased from ~65 pmol/kg in 1976 to ~30 pmol/kg (so North Atlantic and North Pacific surface waters now have comparable levels of lead). It is likely that Asian industrial sources now play a larger relative role in Pb emissions to the Pacific ocean compared to leaded gasoline. The information from total Pb concentration is supplemented by limited data on Pb isotope ratios, which allow some source discrimination (e.g., European vs. U.S. leaded gasoline). Shen and Boyle (1988), Sherrell et al. (1992), Veron et al. (1993, 1999), Hamelin et al. (1997) and Alleman et al. (1999) have established that Pb in the North Atlantic surface waters dominantly derived from the U.S. as would be expected from the 3-4 fold dominance of U.S. leaded gasoline utilization relative to western Europe and the prevailing wind directions. Recent developments in multiple collector plasma mass spectrometry will transform this difficult

measurement into a tool that can be more widely applied in the future. Coral Pb and Pb isotope observations of Shen and Boyle (1987) extend the perspective back to the turn of the 20th century, when industrial Pb emissions (smelting, coal burning, and other high temperature processes) began their rise until the 1940's, when they were overtaken by Pb gasoline emissions as the dominant source. The decreases in Pb gasoline emissions over the past two decades have probably restored industrial Pb emissions as the dominant source of Pb to the ocean. This ongoing large scale geophysical experiment allows oceanographers to test their knowledge of mechanisms of chemical dispersal in the oceans. The Pb variations that we have observed can be reconciled with our oceanographic understanding of the late 1990's. In the upper euphotic zone, the "scavenging" process documented by <sup>210</sup>Pb indicates that surface Pb should trace emissions with a lag consistent with a 2-year residence time. In the well-mixed thermocline (200-1000m) of the subtropical western North Atlantic, Pb follows the pattern predicted by decadal isopycnal "ventilation" of deep levels as calibrated by <sup>3</sup>He-<sup>3</sup>H studies (Jenkins, 1998), with relatively little in-situ scavenging on the decadal timescale. In the more laminarily-ventilated eastern North Atlantic, the Pb distribution follows the pattern predicted by the Luyten et al. (1983) model as calibrated by <sup>3</sup>He-<sup>3</sup>H studies (Jenkins, 1998; Robbins and Jenkins, 1998). It now seems appropriate to insert our understanding of Pb input evolution and oceanic behavior into large-scale 3D ocean circulation models and compare the observed evolution of Pb in the deep sea with our calculations.

- Schaule, B. & Patterson, C., *Earth Planet. Sci. Lett.*, **54**, 97-116, (1981).
- Schaule, B. & Patterson, C., *Trace Elements in Seawater, Plenum*, 487-504, (1983).
- Shen, G. & Boyle, E., *Earth Planet. Sci. Lett.*, **82**, 289-304, (1987).
- Shen, G & Boyle, E., *J. Geophys. Res.*, **93**, 15715-15732, (1988).
- Veron, A, Church, T, Flegal, R, Patterson, C & Erel, Y, *J. Geophys. Res.*, **98**, 18269-18276, (1993).
- Hamelin, B, Ferrand, J, Alleman, L & Nicola, E *Geochim. Cosmochim. Acta*, **61**, 4423, (1997).