Response of Lead Cycling in the Surface Sargasso Sea to Changes in Tropospheric Input

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Lead and its stable isotopes have been analyzed in surface water samples (0-600 m) and trapped particles collected at the Bermuda Atlantic Time Series Station (U.S. Joint Global Ocean Flux Study) in April and November 1989. These results are compared with wet atmospheric lead deposition as determined from precipitation continuously collected in Bermuda since August 1988 as part of the Atmosphere-Ocean Chemistry Experiment program. Despite an expected seasonal variability, lead concentrations in surface waters have clearly decreased by 30 to 40% since 1979 in response to a corresponding decline by a factor of 5 to 8 of the tropospheric deposition. This result is corroborated by stable lead isotope measurements with $^{206}Pb/^{207}Pb$ ratios which are significantly less radiogenic (1.18-1.20) in the first 500 m of the 1989 profile than those measured in 1984 (1.20-1.21). This isotopic shift reflects changes of lead ore supply in the United States as well as a relative increase of the Eurafrican contribution to lead input in the northwest Atlantic that is likely due to the reduction of lead emissions from gasoline consumption in North America. This fast response of lead to interannual variations of the tropospheric input into surface waters is related to its efficient bioreactivity, as demonstrated with a sediment trap deployed at 150 m in April 1989. Sediment trap results show the rapid penetration of lead into the first 200 m associated with large particles during a period of high plankton activity. Retrospective isentropic air mass trajectories in three dimensions coupled with the precipitation events collected in Bermuda in 1988-1989 show that 30 to 40% of the annual lead deposition originates from the trade easterly meteorological regime. This input is clearly evidenced with lead isotopic signals observed in surface waters (0-100 m) in April and November 1989. We show that lead accumulated in the seasonal mixed layer (0-50 m) reflects this atmospheric input. Taking into account the isotopic signal measured in this mixed layer (1.178±0.001) as well as the respective contribution of the temperate westerlies and trade easterlies to atmospheric lead deposition to the Sargasso Sea, we calculate that the isotopic signature from the Eurafrican regions is 1.155 (±0.004). Based on these results, we calculate that the actual $^{206}Pb/^{207}Pb$ average ratio in surface waters of the Sargasso Sea is 1.188 (±0.004).

INTRODUCTION

From the 1930s through the 1970s, the deposition of anthropogenic lead increased by a factor of 3 to 5, primarily because of the consumption of leaded gasoline in the northern hemisphere [Murozomi et al., 1969; Shen and Boyle, 1988] causing lead to be the most atmospherically enriched metallic pollutant from anthropogenic activities. As a consequence, contaminant lead constitutes more than 95% of the lead in the troposphere and surface waters of the North Atlantic, perturbating natural lead seawater profiles and cycling [Duce et al., 1976; Ng and Patterson, 1981; Schaule and Patterson, 1983; Patterson, 1987; Patterson and Settle, 1987; Véron et al., 1987; Nriagu and Pacyna, 1988]. U.S. regulations on lead concentrations in gasoline have resulted in a reduction of lead emissions in North America since the mid 1970s. The increase and subsequent decrease in lead deposition has been monitored using stable radiogenic isotopes of lead (M= 206, 207, 208) measured in corals [Shen and Boyle, 1987], in lake sediments [Shirahata et al., 1980] and in stream waters [Erel et al., 1990]. These isotopes display characteristic anthropogenic signatures from the different countries surrounding the North Atlantic owing to the variety of lead ores used to produce alkyl lead or nonferrous alloy [Chow et al., 1975; Church et al., 1990; Hopper et al., 1991]. As such, lead isotopes are efficient tracers

of the sources and transport processes of pollutant trace metals in the marine environment [Shen and Boyle, 1988; Flegal et al., 1989; Maring et al., 1989; Sturges and Barrie, 1987, 1989a,b; Hamelin et al., 1989; Church et al., 1990; Véron et al., 1992].

Anthropogenic lead is very reactive in the ocean, causing 30 to 40% of the contaminant lead deposited in the surface waters to be readily accumulated in surficial pelagic sediments [Véron et al., 1987; Hamelin et al., 1990] because of rapid vertical transport associated with large marine particles generated in the euphotic zone [Lambert et al., 1991] and to a lesser extent with small suspended material repackaged at greater depth [Sherrell et al., 1992]. One of the main uncertainties regarding lead cycling in the ocean is due to the variations of the input function. Lead concentrations in surface waters show seasonal variations related to hydrographic features, biological activity, and aeolian deposition [Boyle et al., 1986], the relative contributions of which are difficult to evaluate. In order to refine budget calculations and better understand the response of surface seawater to modifications of the tropospheric input, we have analyzed lead and its stable isotopes in surface waters (0-600 m) and large particles in April and November 1989 at the Joint Global Ocean Flux Study (JGOFS) Bermuda Atlantic Time Series (BATS) station near Bermuda (31°5N, 64°1W). These results are compared to the atmospheric deposition as calculated from lead concentrations measured in precipitation continuously collected at Bermuda since 1988 as part of the Atmosphere-Ocean Chemistry Experiment (AEROCE) program. The BATS station has been chosen because of its proximity to the North American continent and to Bermuda which represents an ideal platform to sample the atmospheric transport of anthropogenic emissions associated with the North American temperate westerlies [Church et al., 1984, 1990; Galloway and Whelpdale, 1987]. Furthermore, Schaule and Patterson [1983], Boyle et al. [1986], Shen and Boyle [1988], Sherrell et al. [1992] and Sherrell and Boyle [1992] previously reported lead

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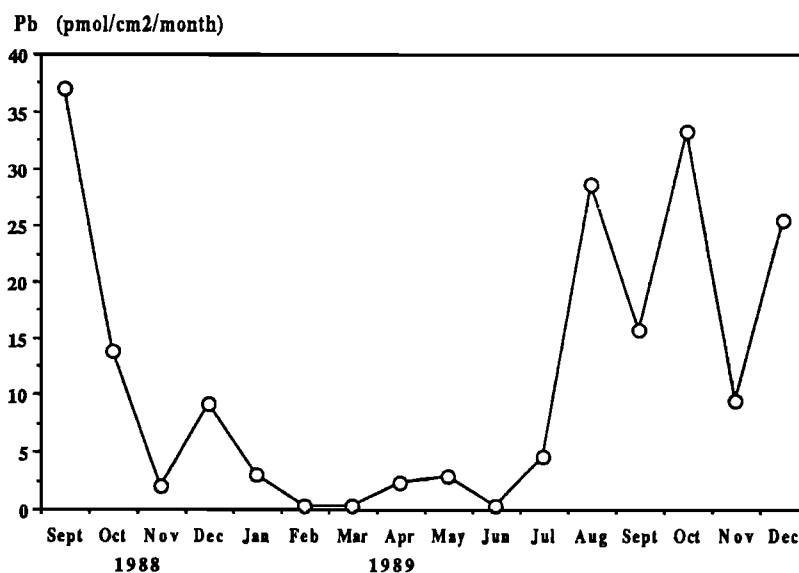
Pb (pmol/cm2/month)

Fig. 1. Monthly wet deposition of lead in Bermuda (Tudor Hill) during 1988 and 1989 as calculated from rain events continuously collected as part of the Atmosphere-Ocean Chemistry Experiment program.

analysis in the Sargasso Sea that can be compared with our data in order to evaluate the temporal dynamics of lead transfer in surface waters in response to the phasing out of leaded gasoline in the western North Atlantic.

METHODS

Rain samples have been collected in Bermuda (West Tower) at Tudor Hill since August 1988 as part of the AEROCE program. The samples are obtained using special automatic collectors with low-density polyethylene (LDPE) buckets. The collector is deployed on a platform at the top of a 20 m anodized aluminium tower. Samples are attended daily and collected after each precipitation event. The rain fall is automatically gauged and compared to the total deposition as recorded in a bucket.

Seawater samples were collected on the R/V *Cape Hatteras* using 30 L Teflon®-coated Go-Flo bottles specially cleaned and mounted on a kevlar line. After sampling, the bottles were transported to a portable clean area where up to 2 L of each bottle were transferred to LDPE bottles and immediately frozen. Each sample was thawed and acidified to pH 1.5 with ultra pure nitric acid a few hours prior to analysis.

Soutar type sediment traps were deployed as a floating array at 150 m between April 18 and 23 at the BATS station. The trap consisted of paired 0.25 m² gel-coated fiberglass cones with swimmer avoidance cod ends designed by *Coale* [1990]. Material in the cod ends was preserved with high-purity buffered formalin. Recovered sediment was dried and all measurements were salt corrected.

Lead in seawater was extracted by a dithizone-chloroform method and determined by isotope dilution thermal ionization mass spectrometry using a ^{208}Pb spike [*Patterson and Settle*, 1976]. The uncertainty on each measurements was determined to be less than 2% of the total concentration. A multicollector VG/Sector solid source mass spectrometer was used to collect multiple sets of stable lead isotopes (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb) ratios on unspiked aliquots. Using the standard SRM 981 from the National Institute of Standards and Technology (Gaitherburg, Md.), the precision of the instrument was found to be 0.1%. Because of a small fractionation factor and better detection limits, ratios of $^{206}Pb/^{207}Pb$ are more accurate (±

0.002) and are used in the discussion. As a consequence, the $^{206}Pb/^{207}Pb$ ratios are referenced more often than other ratios.

Lead in precipitation was analyzed by direct injection of unfiltered samples acidified in 0.4% HCl into the graphite furnace (model 700) of a Perkin Elmer atomic absorption spectrometer (model 1100b) [*Tramontano et al.*, 1987]. The overall uncertainty, including blanks, was less than 10% of the measured concentrations.

A Conductivity/Temperature/depth rosette equipped with Go-Flo bottles was deployed to collect samples for nutrients, oxygen, salinity, temperature, and chlorophyll. Hydrographic parameters were provided by G.A. Cutter (Department of Oceanography, Old Dominion University, Norfolk, Virginia, private communication, 1990).

RESULTS

Atmospheric Transport and Deposition

Air mass transport is characterized using retrospective air mass trajectories based on isentropic analysis techniques. Meteorological data from the National Meteorological Center (Camp Springs, Maryland) are used to determine geopotential heights and winds from which the isentropic levels are defined. Assuming that air parcels retain their identity over the period of analysis and considering a dry adiabatic motion of the air, the trajectories from Bermuda are calculated up to 1 week back in time. The uncertainties due to these assumptions are mainly related to the paucity of wind data recorded over the open ocean and are largely discussed by *Merrill* [1989]. According to *Stunder et al.* [1990] and W. Ellis and J. Merrill (personal communication, 1991) trajectories are classified as oceanic or continental based on the time spent over the ocean prior to collection (continental with less than 3 days over the ocean and oceanic with 3 days or more over the ocean). Trajectories from North America are generally defined as continental. Among the oceanic trajectories, we sort trajectories associated with the trade easterlies. We observe that 90% of the air masses transported with the trade easterlies are confined from June to October.

Lead concentrations in precipitation have been volume weighted by month in order to calculate monthly deposition fluxes (Figure 1). Deposition is corrected for the marine

recycled component based on sodium concentrations in precipitation and an enrichment factor of 5000 which expresses the enrichment of Pb/salt in sea-salt aerosols compared to the Pb/salt in seawater [Patterson and Settle, 1987]. In this study, sea-salt correction accounts for less than 5% of total lead concentrations in precipitation.

By coupling lead deposition and the air mass trajectories corresponding to each rain event collected in Bermuda, we calculated the relative contribution of the North American westerlies and of the Eurafrican trade easterlies to the tropospheric lead input into surface waters of the Sargasso Sea (Figure 2). We estimate that input from the trade easterly regime could represent as much as 30 to 40% of the overall lead input in 1989 in the northwest Atlantic.

Lead Concentrations in Seawater

Lead concentrations in seawater are reported in Table 1a. The April profile extends to 550 m. Lead concentrations show a minimum in surface waters (70-75 pM), increase rapidly to a maximum of 120 pM at 200 m, and then slightly drop from 200 to 550 m (a 5-10% decline). The minimum in surface waters can be explained by high plankton activity during the time of the sampling as shown by a chlorophyll peak at 100 m and a large oxygen consumption in the first 200 m (Figure 3a). The November profile lies between 0 and 150 m. Lead concentrations display a maximum in surface waters at 10 m (89 pM) and decrease sharply to reach 72 pM at 150 m. A mixed layer (0-50 m) due to a seasonal thermocline is evidenced in the salinity and temperature profiles (Figure 3b). This hydrographic structure inhibits exchanges with the underlying waters and contributes to the accumulation of lead deposited during summertime when plankton activity is low [Boyle et al., 1986]. Therefore we can expect lead concentrations to increase in this seasonal mixed layer during summertime. Despite of seasonal variations, lead concentrations in April and November 1989 are 10 to 50% lower than the previously reported data from the Sargasso Sea (Figure 4).

Lead Isotopes

Lead isotope ratios in surface seawater are shown in Table 1b and are compared to continental sources (Table 2) and atmospheric results obtained by the combination of air mass trajectories and stable lead isotopes in Bermuda [Véron et al., 1992]. In April, the $^{206}Pb/^{207}Pb$ profile displays two different trends with ratios ranging from 1.186 to 1.188 in surface waters (0-90 m) and from 1.197 to 1.199 below (200-550 m). The tropospheric lead isotopic signal shows a clear Euroafrican signature during the time of the sampling, with $^{206}Pb/^{207}Pb$ ratios ranging from 1.13 to 1.18 (Table 2). Likewise, the November $^{206}Pb/^{207}Pb$ profile has two discrete trends including (1) ratios of 1.177 to 1.180 in the first 30 m and (2) ratios of 1.188 to 1.190 between 50 and 150 m. The less radiogenic signature is confined to the seasonal mixed layer and corroborates the accumulation of lead from Eurafrican origin. The underlying water reflects a mixing of North American and Euroafrican sources as expected from the April profile. The $^{206}Pb/^{207}Pb$ ratios measured in surface waters in 1989 are significantly less radiogenic than those observed by Shen and Boyle [1988] in 1984 (Figure 5), while they agree with the Sherrell et al. [1992] profile determined in 1988 below 100 m. These results clearly reflect the impact of the phasing out of leaded gasoline, as already suggested by the decrease of lead concentrations in Sargasso Sea surface waters.

DISCUSSION

Impact of the Phasing out of Leaded Gasoline in the Northwest Atlantic

Lead inventory in the Sargasso Sea surface water. As determined from precipitation samples (AEROCE program), the wet input accounts for 126 pmol cm⁻² of lead in Bermuda in 1989 (Figure 1). Assuming that dry deposition represents 5 to 20% of the total lead flux over the ocean [Settle and Patterson, 1982; Dedeurwaerder et al., 1985; Jickells et al., 1987; Church et al., 1984, 1991], we calculate that the total lead
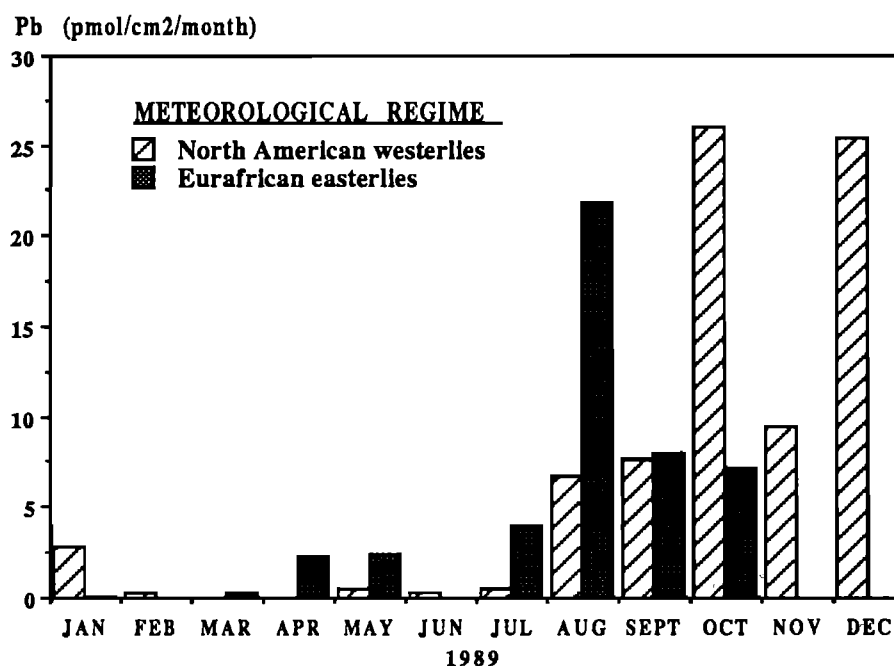
Pb   (pmol/cm2/month)

Fig. 2.   Relative contribution of North American (hatched columns) and Eurafrican regions (shaded columns) to the tropospheric input of lead in Bermuda using isentropic trajectories coupled with each rain event.

TABLE 1a. Dissolved Lead Concentrations (pmol L⁻¹) in Surface
Waters of the Bermuda Atlantic Time Station (BATS)
in April and November 1989

Depth, m	April 1989	November 1989
0.5	nd	83.2
10	nd	89.0
15	75.5	nd
30	73.8	88.4
70	nd	78.5
90	102	nd
150	nd	72.1
200	120	nd
350	112	nd
550	115	nd

Here nd means no determination.

deposition to the Sargasso Sea in 1989 is 150 ± 30 pmol cm⁻². Similar estimates and calculations made by Schaule and Patterson [1983] in the Sargasso Sea (about 250 km northwest of Bermuda) indicate tropospheric lead fluxes of 950 ± 120 pmol cm⁻² in 1979, suggesting a reduction of the lead input by a factor of 5 to 8 between 1979 and 1989 to the western North Atlantic Ocean. We can verify that the decline of lead concentrations observed in surface waters between 1979 and 1989 (Figure 4) is directly related to this reduction of the atmospheric input. To do so, we consider the surface water inventory of Pb in 1989 from (1) measured lead concentrations in April and November 1989 seawater profiles (this study) and from (2) a mass balance calculation using Pb inventory in 1979 [Schaule and Patterson, 1983]. We determine (2) using a 10-year atmospheric input function and the oceanic vertical and advective in and out Pb transport in the Sargasso Sea as described by Boyle et al.. [1986], Shen and Boyle [1988], Lambert et al. [1991], Véron et al. [1991, 1992], Sherrell et al. [1992], Sherrell and Boyle [1992]. This inventory (2) is calculated according to the following expressions:

$$\Delta A_1/\Delta t = f(t) + (K_{21})A_2 + (K_i)A_1 - (K_{P1})A_1 - (K_{12})A_1 - (K_e)A_1 \quad (1)$$

$$\Delta A_2/\Delta t = (K_{12})A_1 - (K_{23})A_2 - (K_{P2})A_2 \quad (2)$$

at t=0 (1979) $A_1=A_0Z_1$ and $A_2=A_0Z_2$. Expressions (1) and (2) refer to a box model for lead cycling into surface waters of the Sargasso Sea where the sizes of the oceanic boxes are set at 0-

100 m (box 1) and 100-600 m (box 2). Box 1 corresponds to the surface mixed layer [Boyle et al., 1986] (Figure 3) and box 2 corresponds to the subtropical mode water (18°C water) and to the upper thermocline. The terms $\Delta A_1/\Delta t$ and $\Delta A_2/\Delta t$ represent the response of the 2 reservoirs (as expressed by the lead inventory, A_1 and A_2, in each reservoir) from 1979 to 1989 as a function of

f(t)　time dependent tropospheric lead input to the mixed layer (box 1);

A_0Z_1　lead accumulated in box 1 at t=0 (1979) (1592 pmol cm⁻²) [Schaule and Patterson, 1983];

K_{P1}　rate constant for removal of lead from box 1 by large particles (0.3 year⁻¹) [Véron et al., 1987; Hamelin et al., 1990; Lambert et al., 1991];

K_{12}　rate constant for lead ventilation from box 1 to box 2 (0.5 year⁻¹) [Nozaki et al., 1976];

K_e　rate constant for lateral export of lead from box 1 (0.1 year⁻¹) [Véron et al., 1991];

K_i　rate constant for lateral import of lead into box 1;

K_{21}　rate constant for water transfer from box 2 to box 1 (0.1 year⁻¹);

A_0Z_2　lead accumulated in box 2 at t=0 [1979] (8208 pmol cm⁻²) [Schaule and Patterson, 1983];

K_{P2}　rate constant for removal of lead from box 2 by small particles (0.013 year⁻¹) [Lambert et al., 1981; Sherrell et al., 1992; Sherrell and Boyle, 1992];

K_{23}　rate constant for lead ventilation from box 2 to the main and lower thermocline (0.06 year⁻¹) [Jenkins, 1980].

This simple box model is presented as that for the Lasaga [1980] expression. Inventories are integrated over a 10-year period (1979-1989) according to equations (1) and (2). The input function f(t) corresponds to the atmospheric deposition as calculated by Schaule and Patterson [1983] in 1979 and this study (1989). Assuming a linear input, we estimate an annual deposition of 590 pmol cm⁻² of lead to the Sargasso Sea between 1979 and 1989. This assumption leads to an atmospheric input of 630 ± 95 pmol cm⁻² in 1983 which is in good agreement with Boyle et al.'s [1986] and Jickells et al.'s [1987] calculation of total lead deposition to the Sargasso Sea in 1983 (748 pmol cm⁻²). Lead exported from the North American basin is likely to partially return with North Atlantic Deep Waters at depth (when entrained northward by the North

TABLE 1b. Isotopic Composition of Dissolved Lead in Surface Waters of the BATS Station
Near Bermuda in April and November 1989

| Depth, m	April 1989			November 1989		
	$^{206}Pb/^{207}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{206}Pb/^{207}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
0.5	1.186	18.50	37.95	1.180	18.45	38.11
10	nd	nd	nd	1.177	18.25	37.75
15	1.186	18.42	37.91	nd	nd	nd
30	1.187	17.55	36.12	1.180	nd	nd
70	nd	nd	nd	1.188	18.62	38.26
90	1.188	18.29	37.64	nd	nd	nd
150	nd	nd	nd	1.190	18.56	38.00
200	1.197	nd	nd	nd	nd	nd
350	1.199	18.61	38.02	nd	nd	nd
550	1.198	18.69	38.18	nd	nd	nd

Here nd means no determination.
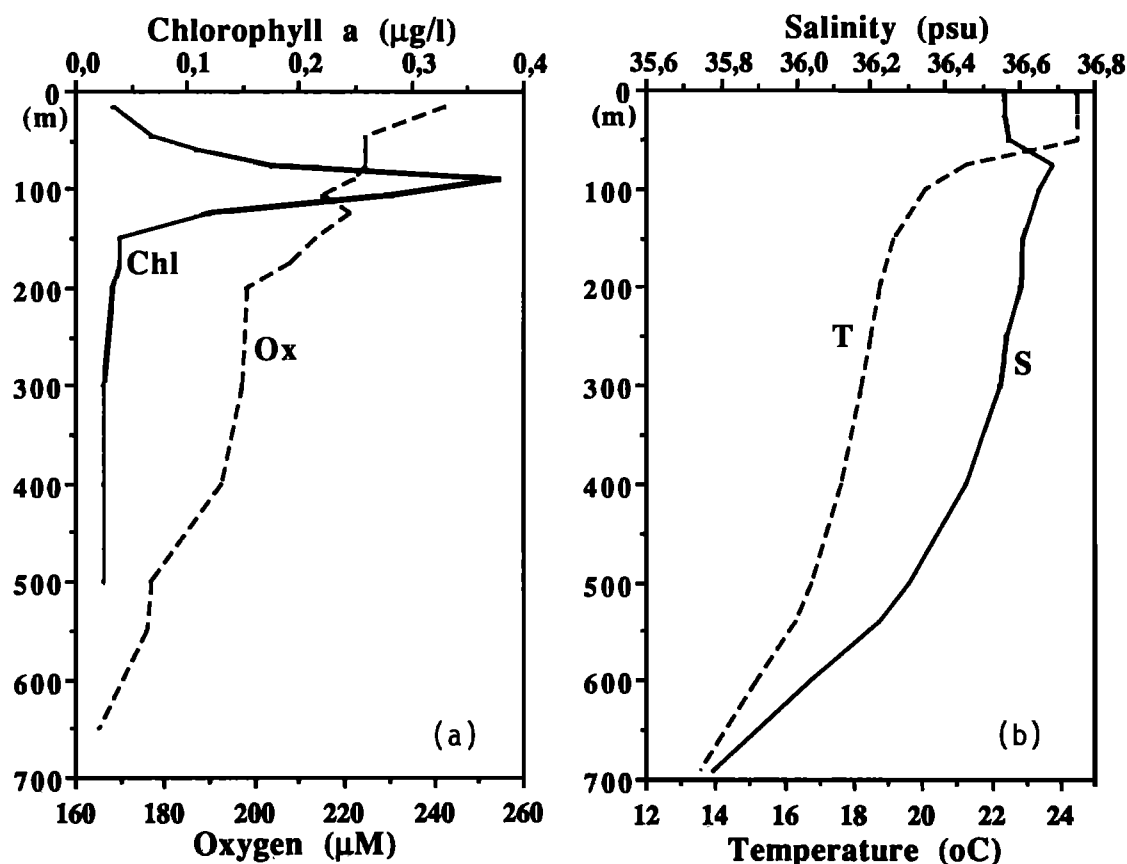
Fig. 3. (a) Chlorophyll (solid line) and oxygen (dashed line) concentrations in surface waters at the Bermuda Atlantic Time Series (BATS) station near Bermuda in April 1989. (b) Salinity (solid line) and temperature (dashed line) in surface waters at the BATS station near Bermuda in November 1989.

Atlantic Current) or to be entrained within the Subtropical North Atlantic Gyre into the North African basin. According to *Jickells et al.* [1987] calculations, this latter recirculation and upwelled mixing in the upper thermocline do not significantly contribute to the lead inventory in the Sargasso Sea surface waters at the latitude of Bermuda. Therefore we consider Ki as negligible in our calculation.

According to expressions (1) and (2), we calculate that the integrated lead inventory between 0 and 600 m (box 1 and 2) should be 5480±1100 pmol cm^{-2} in 1989. The measured inventory from our two 1989 profiles at the BATS station is 6500±300 pmol cm^{-2}. These two determinations are in good agreement, suggesting that the decline of atmospheric deposition is likely to be mostly responsible for the observed decrease of lead inventory in surface waters of the Sargasso Sea, although this result should be considered with caution given the uncertainties on our calculation and the observed seasonal variation of lead concentrations in the mixed layer [*Boyle et al.*, 1986 and this study].

Evidence of a rapid transfer of lead in surface waters. Comparison of the 1979 (9800 pmol cm^{-2}) and 1989 (6500 pmol cm^{-2}) lead inventories between 0 and 600 m confirms the strong reactivity of lead in seawater. This reactivity is evidenced by the observed seasonal change of isotopic composition in surface waters (0-100 m) between April and November 1989 (Figure 5) without much change in concentration. Biological processes are likely to generate this efficient scavenging of lead in surface waters as suggested by a

sediment trap deployed during 5 days at 150 m in April 1989. Lead fluxes measured from this trap (1.1 pmol cm^{-2} for 5 days) account for 20% of the previous 3 months of aeolian deposition (5.9 pmol cm^{-2}). Such an efficient transport is related to high productivity in the first 100 m as shown by the oxygen and chlorophyll profiles (Figure 3a). During this particuliar period, lead is rapidly scavenged from surface waters associated with plankton and fecal pellets [*Lambert et al.*, 1991]. Stable isotopic ratios confirm this assumption, with large particles displaying a $^{206}Pb/^{207}Pb$ ratio of 1.187 at 150 m, similar to those observed in surface waters of the April profile and different from the $^{206}Pb/^{207}Pb$ ratio at 200 m (1.197). These results suggest that lead is effectively transported through the surface water column in a few days during a period of high plankton activity. The homogeneity of the $^{206}Pb/^{207}Pb$ signal in the first 100 m (1.186-1.188) can be explained by the mineralization of large sinking particles (>10 µm) [*Lambert et al.*, 1991].

Stable lead isotopic signal. A change of the U.S. isotopic signature from 1.21-1.23 to 1.19-1.20 has been observed in the western North Atlantic troposphere during the past 20 years [*Shen and Boyle*, 1987; *Patterson and Settle* 1987; *Véron et al.*, 1992]. This variation has been mainly attributed to the phasing out of leaded gasoline in North America, involving a relative change of atmospheric pollutant lead sources and a reduction of lead ore supply from the very radiogenic Missouri ores. The atmospheric change of stable lead isotope composition is clearly evidenced in surface waters of the
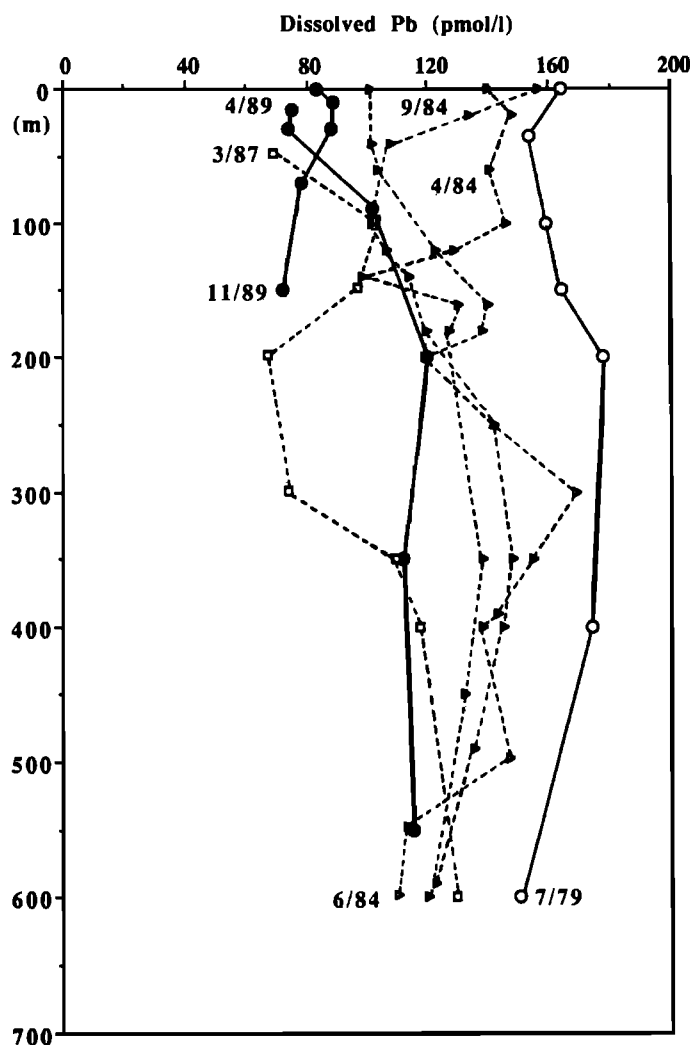
Dissolved Pb (pmol/l)

Fig. 4. Dissolved lead concentrations in Sargasso Sea surface waters in 1979 [*Schaule and Patterson*, 1983] (open circles), 1984 [*Boyle et al.*, 1986] (3 profiles; open triangles), 1987 [*Sherrell et al.*, 1992] (open square) and 1989 (this study) (2 profiles; solid circles). Isotope dilution mass spectrometry measurements are represented by solid lines and atomic absorption measurements are indicated by dashed lines.

Sargasso Sea (JGOFS station) with $^{206}Pb/^{207}Pb$ ratios varying from 1.203 in 1984 [*Shen and Boyle*, 1988] to 1.187 in 1989 (this study) (Figure 5). This difference propagates to 500 m. Below 200 m, the isotopic signal in the April 1989 profile reflects an older signal which extends down to the main

thermocline with no noticeable change (1.198 ± 0.001). The similarity between the profiles at 500-600 m suggests that the downward perturbation induced by changes in the tropospheric signal in the past 10-15 years has little or no effect on the reservoir of dissolved lead in the main and lower thermocline.

Seasonal Variations and Eurafrican Input

Lead concentrations show significant variations between the April and November 1989 profiles (Table 1). As shown by *Boyle et al.* [1986] in the Sargasso Sea, lead concentrations in surface waters can display seasonal variations of 25% or more. The low concentrations observed in the first 50 m of the April profile (74-75 pM) have been explained by a rapid scavenging of lead associated with large biogenic particles during a period of high plankton activity. The maximum measured at 200 m (120 pM) could be due to a partial mineralization of these aggregates from which lead is released. Lead concentrations in the first 50 m are 15 to 20% higher in the November profile (83-89 pM) than in the April profile (74-75 pM). In addition, the concentrations in the first 50 m in November are 10 to 20% higher than in the underlying waters (72-78 pM). This relative increase in concentration can be attributed to an accumulation of lead as suggested by the presence of a seasonal thermocline which generally inhibits vertical mixing from May to October in the Sargasso Sea (Figure 3) [*Boyle et al.*, 1986].

The November isotopic signal measured in the seasonal mixed layer at 0-50 m ($^{206}Pb/^{207}Pb = 1.177\text{-}1.180$) is clearly less radiogenic than in April ($^{206}Pb/^{207}Pb = 1.186\text{-}1.187$) and in the underlying waters (50-150 m) of the November profile ($^{206}Pb/^{207}Pb = 1.188\text{-}1.190$). The $^{206}Pb/^{207}Pb$ ratios in this mixed layer suggest a mixing of North American and Eurafrican components (Table 2). While the eastern North American isotopic signal is well defined in 1989 ($^{206}Pb/^{207}Pb = 1.204\pm0.004$) [*Church et al.*, 1990; *Véron et al.*, 1992], the Euro-African signal, associated with the trade easterlies, is more diffuse ($^{206}Pb/^{207}Pb = 1.11\text{-}1.16$) because of the variation and extent of the different European sources. According to the air mass trajectories, the Eurafrican component is a significant tropospheric source in Bermuda during summertime (Figure 2) when lead is trapped by a water column thermostratification. Based on Figure 2, we calculate that lead from the Eurafrican region represents 51% of the tropospheric input into the Sargasso Sea from May to October 1989. Lead accumulated in the seasonal mixed layer (90 ± 10 pmol cm^{-2}) accounts for 80% of the atmospheric deposition during that period (110 ± 20 pmol cm^{-2}). Assuming that lead in

TABLE 2. Recent $^{206}Pb/^{207}Pb$ Signatures in the Troposphere of the Regions Potentially Contributing to the Atmospheric Isotopic Signal in the Northwest Atlantic

| Source	$^{206}Pb/^{207}Pb$	Date	Location	Reference
Western Europe	1.115-1.150	1990	Ireland, Suiss, Fr., Ger.	A.R. Flegal et al. (pers. comm.1992)
	1.130-1.155	1988	Sweden	*Hopper et al.* [1991]
Eastern United States	1.198-1.203	1989	Western N.Atlantic	*Véron et al.* [1992]
	1.205-1.210	1988	Western N.Atlantic	*Church et al.* [1990]
Trade easterlies	1.150-1.160	1988	Southern N.Atlantic	*Church et al.* [1990]
Mixed United States/Eurafrican	1.193	April 19, 1989	JGOFS BATS station	*Véron et al.* [1992]
	1.186	April 20, 1989	JGOFS BATS station	*Véron et al.* [1992]
	1.179	April 21, 1989	JGOFS BATS station	*Véron et al.* [1992]
	1.182	April 21, 1989	JGOFS BATS station	*Véron et al.* [1992]
	1.131	April 22-23, 1989	JGOFS BATS station	*Véron et al.* [1992]
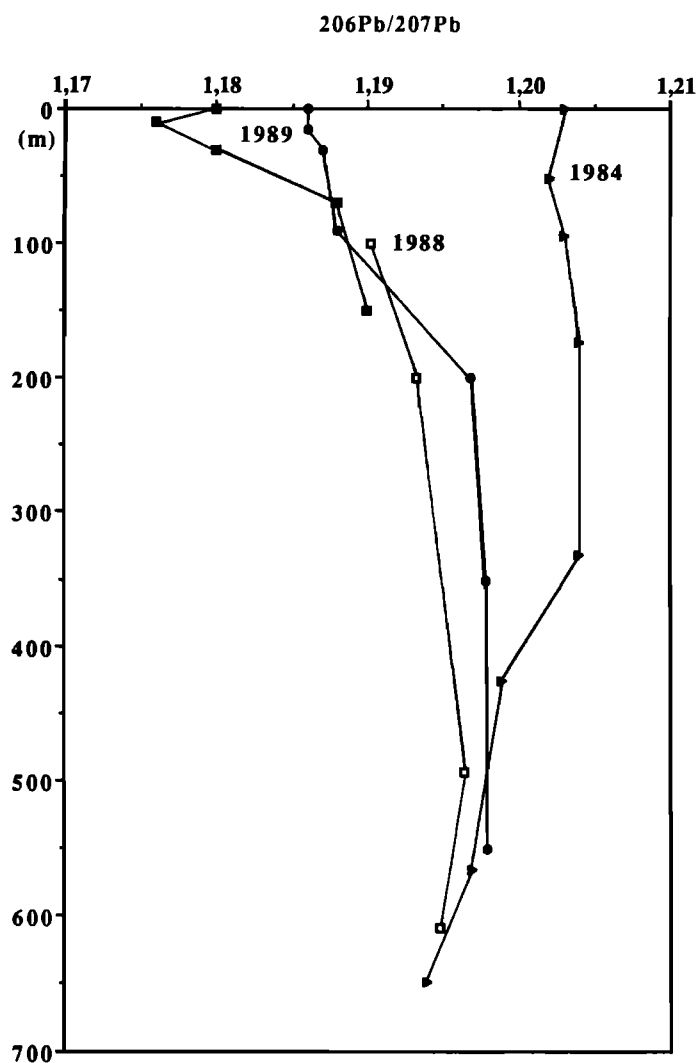
206Pb/207Pb

Fig. 5. Stable lead isotopes (^{206}Pb/^{207}Pb) in surface waters at the BATS station near Bermuda in 1984 [*Shen and Boyle*, 1988] (open triangles), 1988 [*Sherrell et al.*, 1992] (open squares) and 1989 (this study) (April, closed circles; November, solid squares).

the mixed layer reflects summertime atmospheric input and considering ^{206}Pb/^{207}Pb ratios in the mixed layer (1.178±0.001) and in the North American westerlies (1.204±0.004), we calculate that the integrated ^{206}Pb/^{207}Pb signal from Eurafrican sources is 1.155±0.004 in the northwest Atlantic troposphere. This result is in good agreement with *Church et al.* [1990] measurements in the trade easterlies which displayed ^{206}Pb/^{207}Pb ratios ranging from 1.150 to 1.160 in August 1988. Considering these isotopic signatures and the atmospheric sectoring at Bermuda, we calculate that the average ^{206}Pb/^{207}Pb ratio in the Sargasso Sea surface waters should be 1.188 (±0.004).

CONCLUSIONS

We use the combination of 3 years of atmospheric deposition records with surface seawater collections in 1979 and 1989 to determine the impact of seasonal and long-term changes of the tropospheric input function on lead cycling in the northwest Atlantic. We observed that the tropospheric lead input to the Sargasso Sea surface waters has declined by a factor of 5 to 8 since 1979. This significant decrease is mostly due to

the phasing out of leaded gasoline in North America. By coupling each precipitation event to the corresponding retrospective isentropic air mass trajectories, we estimate that 30 to 40% of lead deposition in the western North Atlantic is associated with the Eurafrican trade easterlies. Considering these results, we calculate the average isotopic signal (^{206}Pb/^{207}Pb) in the Sargasso Sea surface water and in the Euro-African trade easterlies to be 1.188 (±0.004) and 1.155 (±0.004) respectively. These isotopic signatures as well as the relative increase of the Eurafrican contribution and its impact on surface waters should be taken into account when modeling the future response of lead cycling in relation to changes of the tropospheric input in the western North Atlantic Ocean.

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