

RESEARCH LETTER

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Key Points:

- Recent sources of Pb were evident despite a fourfold reduction of Pb in NE Atlantic surface waters since leaded gasoline prohibition
- Enhanced Pb was evident in Mediterranean Outflow Waters, transported >2,500 km across the NE Atlantic
- Sediments represented an important source of Pb to overlying waters, exceeding the atmospheric flux of Pb

Supporting Information:

- Supporting Information S1

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Anthropogenic Signatures of Lead in the Northeast Atlantic

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Abstract Anthropogenic activities have resulted in enhanced lead (Pb) emissions to the environment over the past century, mainly through the combustion of leaded gasoline. Here we present the first combined dissolved (DPb), labile (LpPb), and particulate (PPb) Pb data set from the Northeast Atlantic (Celtic Sea) since the phasing out of leaded gasoline in Europe. Concentrations of DPb in surface waters have decreased by fourfold over the last four decades. We demonstrate that anthropogenic Pb is transported from the Mediterranean Sea over long distances (>2,500 km). Benthic DPb fluxes exceeded the atmospheric Pb flux in the region, indicating the importance of sediments as a contemporary Pb source. A strong positive correlation between DPb, PPb, and LpPb indicates a dynamic equilibrium between the phases and the potential for particles to “buffer” the DPb pool. This study provides insights into Pb biogeochemical cycling and demonstrates the potential of Pb in constraining ocean circulation patterns.

Plain Language Summary Lead (Pb) is a toxic element to all living organisms and may cause health impacts upon exposure to high levels. Humans have introduced large amounts of Pb into environment over last 150~years mainly through combustion of leaded gasoline and coal. The anthropogenic Pb is transported over long distances in the atmosphere and deposited in remote ocean regions resulting in elevated Pb concentrations. Since the implementation of stricter environmental regulations over last decades, Pb levels in surface waters have declined. In this study we report five times lower Pb concentrations in European surface waters compared with those from the 1980s and also present recent Pb inputs. Once anthropogenic Pb is introduced into the marine environment, it can be used to trace specific water mass transport throughout the global ocean. In this study we use Pb signal to demonstrate transport of Mediterranean Sea surface water over 2,500~km across the Atlantic Ocean. Our results serve as evidence of the positive impact of environmental regulations on Pb inputs and highlight the requirement of continuing efforts in regulating Pb emissions.

1. Introduction

Lead (Pb) is one of few elements for which the impact of human activity on the marine environment is clearly evident. Anthropogenic perturbation of the natural biogeochemical cycle of Pb in the ocean dates back to circa 1850 (Kelly et al., 2009), with coal and leaded gasoline combustion serving as major sources of Pb to the atmosphere (Kelly et al., 2009; Wu & Boyle, 1997). Anthropogenic Pb is transported in the atmosphere in the form of fine aerosol particles that can travel long distances and are deposited in remote areas resulting in enhanced DPb surface ocean concentrations (Kumar et al., 2014; Véron & Church, 1997), reaching >190 pmol kg⁻¹ during the peak of the Pb emissions in 1970–1980 (Laumond et al., 1984). Anthropogenic Pb has entirely masked signals of naturally sourced Pb (approximately 2.2 pmol kg⁻¹ in surface waters; Henderson & Maier-Reimer, 2002). To date, leaded gasoline has been virtually phased out (except in three countries, as of March 2017, United Nations Environment Programme), and Pb concentrations have decreased significantly from ~170 to <15 pmol kg⁻¹ in surface waters (Boyle et al., 2014; Schaule & Patterson, 1983), leading to recent evidence for natural Pb signatures reemerging in the North Atlantic (Bridgestock et al., 2016).

The spatial and temporal variable historic Pb inputs to the marine environment can be used to investigate the reactivity and cycling of this element and trace long-range ocean circulation patterns (Chen,

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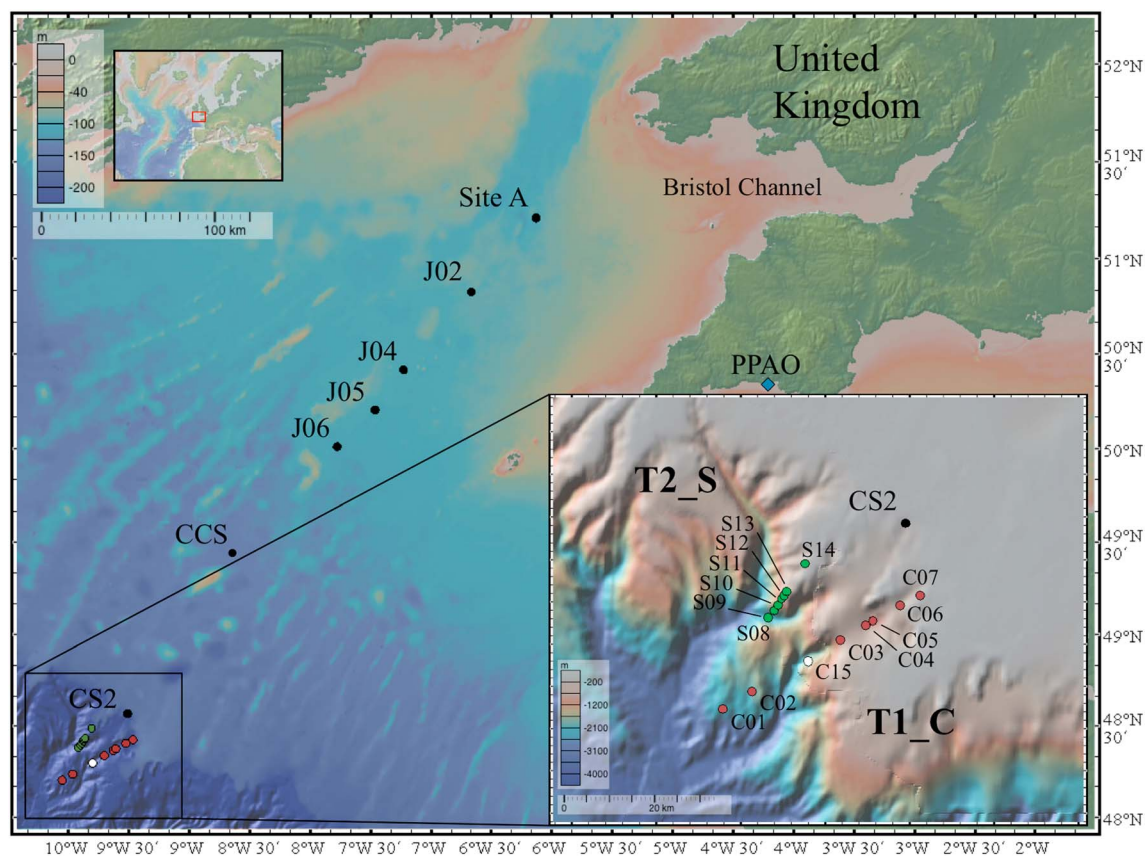


Figure 1. Station locations across canyon T1_C (white and red circles), spur T2_S (green circles) transects, and on-shelf transect (black circle) during three research expeditions in November (DY018), April (DY029), and July (DY033). Blue diamond represents PPAO station. Map was generated using GeoMapApp, <http://www.geomapapp.org> (Ryan et al., 2009).

Goodkin, et al., 2016; Fine, 2010; Lee et al., 2015). Anthropogenic Pb has been utilized as a tracer of subducted surface waters in the Indian (Lee et al., 2015) and Pacific Oceans (Chien et al. 2017) and ventilated surface waters of the Northwest Atlantic (Boyle et al., 2014). Anthropogenic perturbation of natural Pb concentrations in the ocean has been described as an “evolving global experiment” (Boyle et al., 2014) that demonstrates the magnitude of human impact on the environment.

Lead is a particle reactive element in marine waters and is typically removed through scavenging, with the sediments acting as repositories. However, the role of particulate matter and the physicochemical processes that influence the fate of dissolved Pb (DPb) and facilitate long-range transport are poorly constrained. A slow release of DPb from particles and a rapid isotopic exchange with particulate matter that can influence the fate of particulate Pb (PPb) has recently been reported (Chen, Boyle, et al., 2016). Therefore, in order to gain insights into biogeochemical cycling of Pb in the marine environment, both phases should be considered.

Here we report and evaluate the first extensive seasonal study of DPb, PPb, and leachable Pb (LpPb) distributions in Northeast Atlantic marginal seas since the phaseout in Europe of leaded gasoline use in 1980–2011 (European Communities, 1978; Economic Commission for Europe, 2014). This study provides insights into the dynamic relationship between dissolved and particulate phases and demonstrates the role of benthic Pb release and Pb as a tracer of North Atlantic circulation patterns.

2. Study Region, Materials, and Methods

Full details of the study region, sampling and methods are provided in the supporting information (SI) Text S1. Briefly, samples for trace metal analysis were collected during three different seasons: November–December 2014 (DY018), April 2015 (DY029), and July–August 2015 (DY033) in the Northeast Atlantic continental margin (Celtic Sea) (Figure 1), on board *RRS Discovery*. Two off-shelf transects were conducted along a canyon (T1_C,

stations C01–C07 and C15), nearby a spur (T2_S, stations S08–S09) (SI Text S1) and one on-shelf transect in the Celtic Sea (stations CS2, CCS, J02–J06, and Site A). Trace metal samples were collected following GEOTRACES protocols (Cutter et al., 2010). Dissolved Pb and Mn (DMn) were filtered using a 0.2 μm cartridge filter (Sartobran 300, Sartorius), preconcentrated using an automated system (SC-4 DX SeaFAST pico; ESI), and analyzed by high-resolution inductively coupled plasma-mass spectrometry (HR-ICP-MS; Thermo Fisher Element XR) (Rapp et al., 2017). Dissolved Fe (DFe) (0.2 μm filtered) was analyzed by flow injection with chemiluminescence detection (Obata et al., 1993) as detailed in Birchill et al. (2017). Particulate Pb was collected on clean 25 mm Supor[®] polyethersulfone membrane disc filters (Pall, 0.45 μm) and subjected to a 25% acetic acid-hydroxylamine hydrochloride leach (LpPb) (Berger et al., 2008) and subsequently an acid digestion (PPb) (Ohnemus et al., 2014). Particulate samples were analyzed using ICP-MS (Thermo Fisher X Series 2). Dissolved aluminum (DAI) (0.2 μm filtered) was analyzed using spectrofluorometry following the method by Hydes and Liss (1976). Evaluation of the accuracy and efficiency of these methods was carried out using Certified Reference Materials with the results showing good agreement (SI Table S1). Some data points were identified as outliers and were excluded from consideration (SI Text S2). Radium (Ra) isotopes ^{223}Ra and ^{224}Ra were extracted from large seawater volumes (60–100 L) by adsorption onto Mn acrylic fibers (Sun & Torgersen, 1998). Ra activities were analyzed at sea by Radium Delayed Coincidence Counting following standard methodology (Annett et al., 2013; Garcia-Solsona et al., 2008; Moore, 2008; Moore & Arnold, 1996). Ra activities used here ($^{224}\text{Ra}_{\text{xs}}$ and $^{223}\text{Ra}_{\text{xs}}$) are in excess of activity supported by the parent isotopes in the water column (SI Text S1). Water mass distribution was quantified using extended Optimum Multiparameter (extOMP) analysis (Hupe & Karstensen, 2000; Karstensen & Tomczak, 1998; Pollard et al., 2004). The propagation time of Mediterranean Outflow Water (MOW) from the Gulf of Cadiz was calculated analogously to (Waugh et al., 2003), using chlorofluorocarbon-12 data available in the GLODAPv2 data product (Olsen et al., 2016). Aerosol samples were digested using hydrofluoric acid and HNO_3 following the method adapted from Morton et al. (2013) and analyzed by the ICP-MS (Thermo Fisher X Series 2). Section plots figures were created with Ocean Data View (2015) software (Schlitzer, 2015) with Data-Interpolating Variational Analysis gridding settings.

3. Results and Discussion

DPb concentrations in the Celtic Sea region ranged between 29.6 and 122 pmol kg^{-1} (Figures 2a–2c and S1). Off-shelf distributions showed elevated DPb concentrations ($50.8 \pm 3.0 \text{ pmol kg}^{-1}$ ($n = 20$)) in the seasonal mixed layer (SML) along the canyon transect in November and also at stations S08 and S09 along the spur transect. Below the SML, DPb distributions were generally consistent along both transects for all seasons and decreased down to 38 pmol kg^{-1} in the subsurface waters and increased at depths of ~550–1,500 m to $46.6 \pm 5.6 \text{ pmol kg}^{-1}$ ($n = 84$). In deeper waters (>1,500 m), DPb concentrations decreased to $37.0 \pm 3.2 \text{ pmol kg}^{-1}$ ($n = 39$). On the continental shelf, DPb concentrations were generally higher in comparison to the off-shelf transects and ranged between 36.1 and 122 pmol kg^{-1} . Elevated DPb concentrations were measured in surface waters in April ($96.8 \text{ pmol kg}^{-1}$) and July ($99.1 \text{ pmol kg}^{-1}$) with somewhat lower levels in November ($72.6 \text{ pmol kg}^{-1}$), whereas DPb was persistently elevated in bottom waters (up to 121 pmol kg^{-1}) across all seasons. No correlations between DPb and macronutrients were observed.

Our surface water DPb concentration of $40.2 \pm 7.5 \text{ pmol kg}^{-1}$ ($n = 109$) along the shelf break in 2014–2015 showed at least a fourfold decrease compared to previous reports for the region (Figure 2e and Table S2) (Brügmann et al., 1985; Cotté-Krief et al., 2002; Helmers & Van der Loeff, 1993; Lambert et al., 1991; Muller et al., 1994) and were generally lower in comparison to other European shelf waters over the last 4 decades (Kremling & Streu, 2001; Laumond et al., 1984; Monteiro et al., 2015; Pohl et al., 2011; Prego et al., 2013; Waeles et al., 2008). The diminishing DPb concentrations over the last two decades form a success of the phaseout process of leaded gasoline. Nevertheless, the observed concentrations in our study region exceeded predicted natural levels of Pb (Henderson & Maier-Reimer, 2002) by at least an order of magnitude, indicating that the vast majority of Pb still has an anthropogenic origin. Thus, the elevated DPb concentrations we report in surface waters indicate the presence of recent anthropogenic Pb inputs to Northeast Atlantic waters from sources such as coal burning, smelting, or mining (Lee et al., 2014; Nriagu & Pacyna, 1988).

3.1. Long-Range Pb Transport in MOW

Elevated DPb concentrations of $46.6 \pm 5.6 \text{ pmol kg}^{-1}$ ($n = 84$) were a persistent feature in the depth range ~550–1,500 m ($27.30\text{--}27.75 \text{ kg m}^{-3} \sigma_\theta$) in the slope region of the Celtic Sea. The DPb maximum coincided

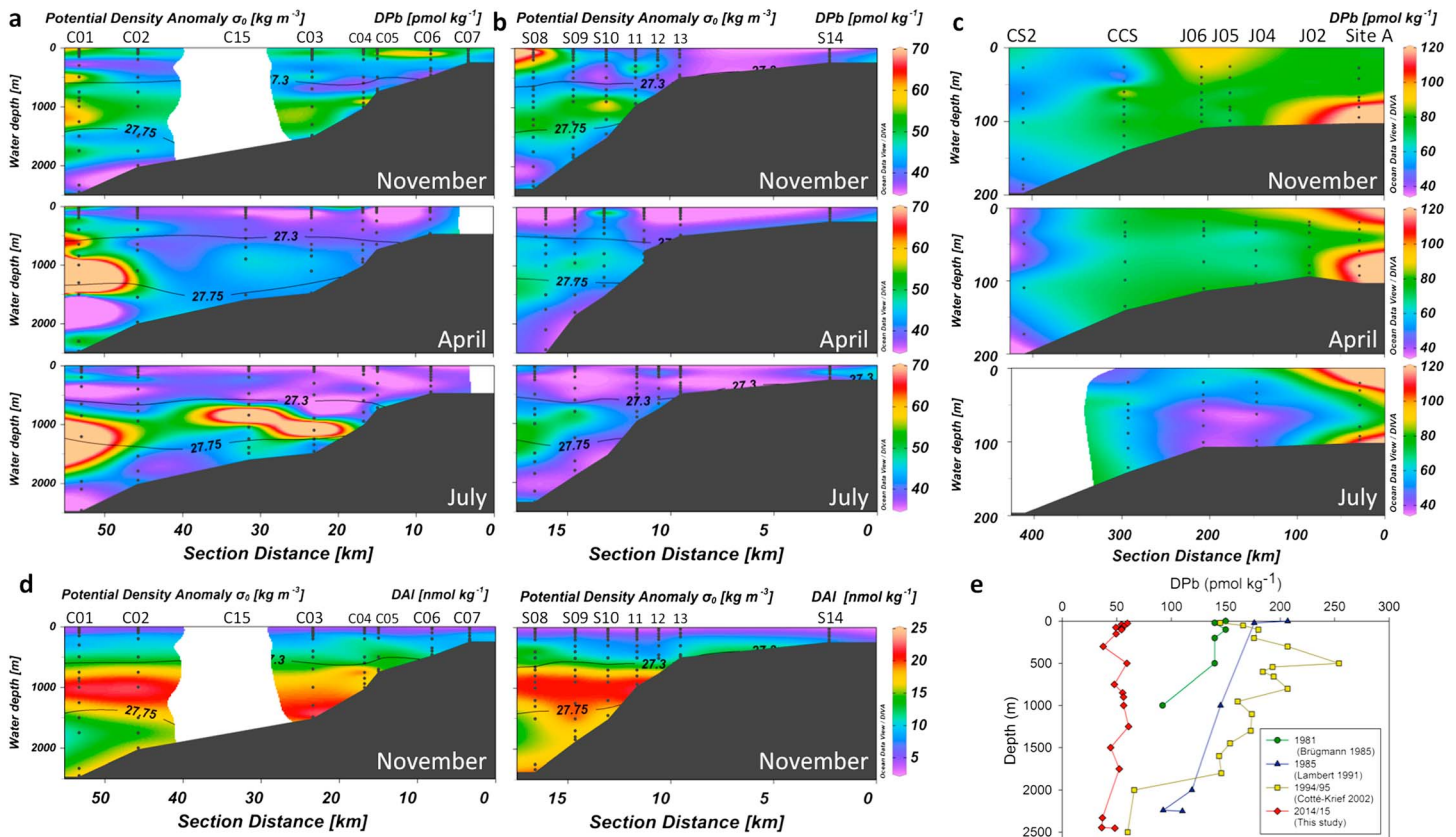


Figure 2. Upper panel: dissolved lead distribution plots (a) along the canyon transects (T1_C, left), (b) along the spur transects (T2_S, middle), and (c) and along the on-shelf transect (left) in November (DY018) (top), April (DY029) (middle), and July (DY033) (bottom). Bottom panel: (d) example of dissolved aluminum distribution plots from November (DY018) along the canyon (T1_C) transect (left) and spur (T2_S) transect (right). For the full dissolved aluminum results please see SI Figure S2. Black lines represent Mediterranean Outflow Water density range contour plots. (e) Reduction of DPb concentrations in the Celtic Sea slope region over the last 30 years. Data are from Brüggmann et al. (1985, green circles), Lambert et al. (1991, blue triangles), and Cotté-Krief et al. (2002, yellow squares), and this study is represented by the S08 station in April (DY029) by red diamonds.

with salinity (35.74) (Figure 3a) and DAI ($17.3 \pm 2.6 \text{ nmol kg}^{-1}$, $n = 88$) maxima (Figure S2), signatures of MOW (Measures & Edmond, 1988; Rolison et al., 2015). Surface waters in the Mediterranean Sea received enhanced atmospheric Pb inputs during the period of leaded gasoline use, with maximum surface DPb of $>190 \text{ pmol kg}^{-1}$ (Laumond et al., 1984). Mediterranean waters also receive enhanced eolian fluxes of Al from Saharan dust (Rolison et al., 2015). Deep water formation occurs in the Levantine Basin and Gulf of Lions, and the saline deep Mediterranean waters with enhanced DPb ($40\text{--}80 \text{ pmol kg}^{-1}$) (Rolison, 2016) and DAI ($125\text{--}170 \text{ nmol kg}^{-1}$) (Rolison et al., 2015) exit the Strait of Gibraltar as bottom waters and mix with Eastern North Atlantic Central Water (García-Ibáñez et al., 2015). The MOW spreads across NE Atlantic at a depth $\sim 550\text{--}1,500 \text{ m}$ and propagates along the continental slope toward the Celtic Sea shelf break. The mean propagation time of MOW from the Gulf of Cadiz to the Celtic Sea slope region is ~ 5 years (Figure S3). The presence of MOW at intermediate depths in the study region has previously been reported (Cotté-Krief et al., 2002; Lambert et al., 1991) and was confirmed by the extOMP analysis (Figures 3b and S4). The core of the MOW (up to 55%) was identified at $\sim 1,000 \text{ m}$ depth with a Gaussian decay (20% at 550 m and 1,500 m).

Enhanced DPb concentrations were also observed along GEOTRACES transects in corresponding MOW density layers ($27.22\text{--}27.82 \text{ kg m}^{-3} \sigma_\theta$): in the Gulf of Cadiz ($49.0 \pm 2.6 \text{ pmol kg}^{-1}$, $n = 14$, GA04; Rolison, 2016), north ($46.1 \pm 6.1 \text{ pmol kg}^{-1}$, $n = 18$, GA01), and south of the Gulf of Cadiz ($55.1 \pm 5.5 \text{ pmol kg}^{-1}$, $n = 10$, GA03, Noble et al., 2015) (Figure S5), in agreement with our observations. Our study region is $\sim 2,500 \text{ km}$ away from the Strait of Gibraltar; therefore, the concentration of DPb and DAI may be expected to decrease through scavenging and/or dilution processes during transit. While DAI concentrations and salinity correlated well ($r^2 = 0.68$) and decreased from $27.8 \pm 7.2 \text{ nmol kg}^{-1}$ and $35.6\text{--}36.4$ (Gulf of Cadiz)

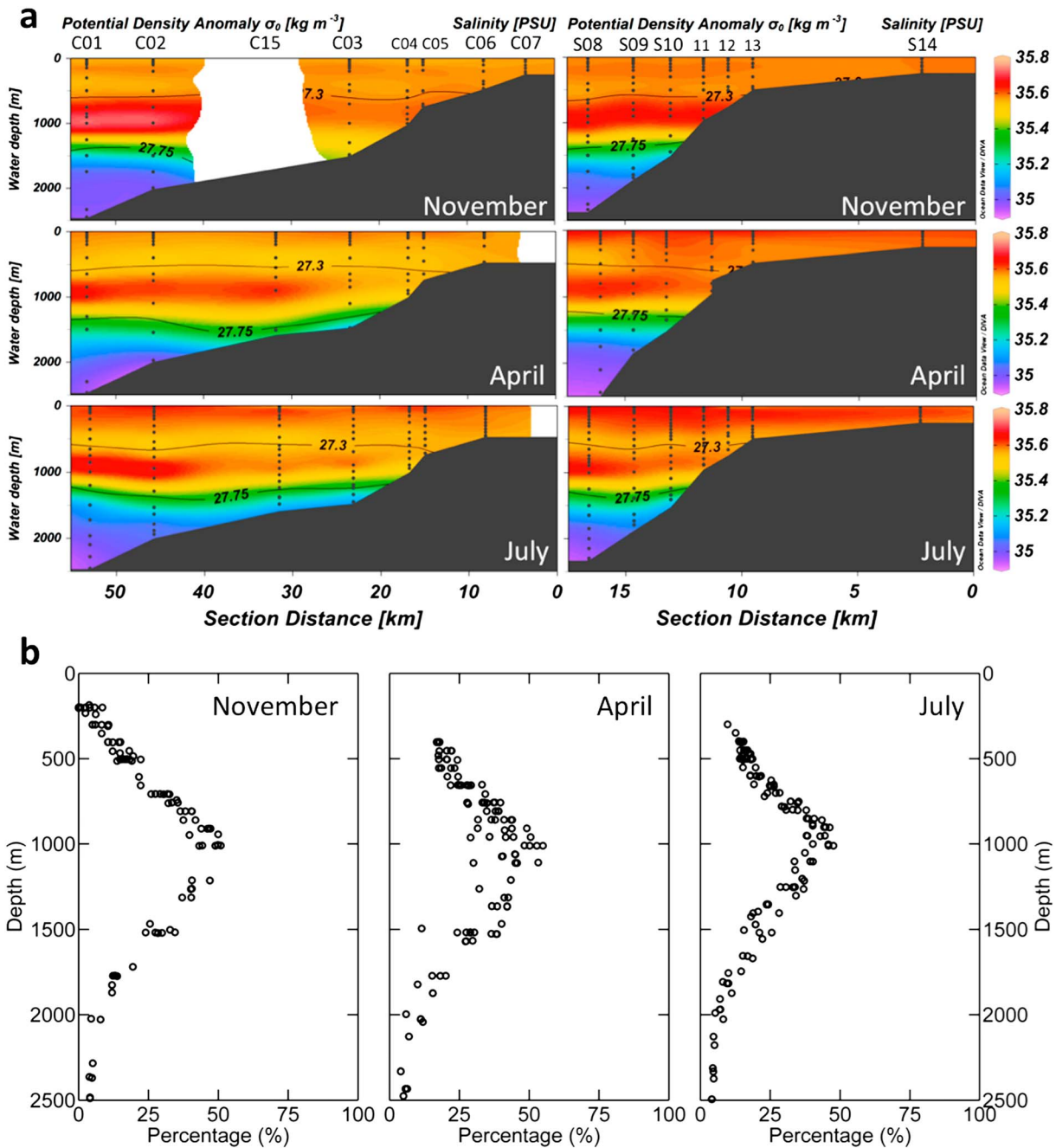


Figure 3. Upper panel (a): salinity distribution plots along the canyon transects (T1_C, left) and along the spur transects (T2_S, right) in November (DY018) (top), April (DY029) (middle), and July (DY033) (bottom). Bottom panel (b): the percentage distribution of Mediterranean Outflow Water along the canyon (T1_C) and spur (T2_S) transects in November (DY018) (left), April (DY029) (middle), and July (DY033) (right). For the full Optimum Multiparameter results see SI Figure S4.

to $17.3 \pm 2.6 \text{ nmol kg}^{-1}$ and 35.4–35.7, there was no correlation of DPb with salinity and DPb concentrations remained unchanged (Figure S6). We suggest the following processes that maintain elevated DPb signal during MOW transit: (i) benthic inputs from the European continental slopes. Shelf break sediments have been suggested as a Pb source in the Philippine Sea (Chien et al. 2017). Local sediment resuspension events in the Bay of Biscay as MOW propagates along the continental slope have also been reported (McCave & Hall, 2002). (ii) Reversible Pb sorption onto particle surfaces. This mechanism has been suggested to supply DPb to North Pacific deep waters (Wu et al., 2010). Pb isotope exchange between these two phases has been demonstrated (Chen, Goodkin, et al., 2016; Sherrell et al., 1992), and the

potential of particle reversible sorption has been shown using thorium isotopes (Bacon & Anderson, 1982). Potentially, DPb ($<0.2 \mu\text{m}$) may be released from particles in the form of small, low specific density inorganic particles (colloids $0.02\text{--}0.2 \mu\text{m}$) with longer residence time. However, PPb dissolution within MOW is also plausible. Our results show that the major portion of PPb was in LpPb form ($78 \pm 10\%$, $n = 205$), while overall the majority of the total Pb pool (PPb + DPb) was in the DPb fraction ($70 \pm 18\%$, $n = 171$), thus implying a significant role of particles in DPb distributions.

Partial mixing with other historically Pb polluted waters, such as Northeast Atlantic ventilated surface waters and Labrador Sea Water (LSW) transported across the North Atlantic (Bridgestock et al., 2018; Zurbrick et al., 2018), has been shown to influence DPb concentrations at intermediate depths and thus also need to be considered. Low-salinity and high-oxygen LSW underlies the warm, saline MOW (Talley & McCartney, 1982). Our extOMP confirmed a layering of the water masses with MOW at a core depth 27.60 kg m^{-3} transiting into the LSW core at 27.79 kg m^{-3} (Figure S4) and a potential for vertical mixing of bottom layers of MOW with LSW. Within waters identified as LSW ($27.76\text{--}27.85 \text{ kg m}^{-3} \sigma_0$), concentrations of DPb ($42.9 \pm 4.5 \text{ pmol kg}^{-1}$ ($n = 42$)), and DAI ($15.8 \pm 1.0 \text{ nmol kg}^{-1}$ ($n = 40$)) were somewhat lower, and salinity ($34.98\text{--}35.4$) and temperature (from 8.8 ± 1.5 to $5.1 \pm 1.0^\circ\text{C}$) were lower in comparison to overlying MOW. These values were higher in comparison to DPb ($36.9 \pm 7.4 \text{ pmol kg}^{-1}$, $n = 60$), DAI ($13.0 \pm 1.2 \text{ nmol kg}^{-1}$, $n = 60$), salinity ($34.94\text{--}35.2$), and temperature ($4.3 \pm 0.6^\circ\text{C}$) observed within LSW ($27.68\text{--}27.81 \text{ kg m}^{-3} \sigma_0$) in the NW Atlantic (GA02 section, 2010) (Mawji et al., 2015). Densities of MOW and LSW are similar, and a complete differentiation is therefore challenging, preventing us from determining the exact fraction of each water mass. Our findings indicate the potential for MOW penetration into deeper waters, altering properties of LSW although the upward vertical mixing cannot be ruled out. We conclude that the DPb maximum in the Celtic Sea region was a result of anthropogenically perturbed MOW masses reaching NE Atlantic continental margins.

3.2. Sediment Release of a Particle Reactive Element

Elevated DPb ($65.9\text{--}121 \text{ pmol kg}^{-1}$) and PPb concentrations ($149\text{--}806 \text{ pmol kg}^{-1}$) were observed in bottom waters at Site A during all seasons and on the continental slope at C03–C04 stations in November (DPb: $52.5 \pm 5.6 \text{ pmol kg}^{-1}$, $n = 7$, PPb: $29.3 \pm 13.9 \text{ pmol kg}^{-1}$, $n = 8$) (Figure 4). Following fluvial or atmospheric inputs to marine waters, Pb is typically scavenged and transferred to the seafloor (Bastami et al., 2015; Marani et al., 1995). Tidal currents, wind-driven waves, and storm events cause resuspension of sediments (Kalnejais et al., 2007), thereby supplying Pb-enriched pore waters and particles to the water column. This mechanism has been reported for deep ocean (Lee et al., 2015; Noble et al., 2015), coastal (Annibaldi et al., 2009; Chien et al., 2017; Sañudo-Wilhelmy & Flegal, 1994), estuarine (Rivera-Duarte & Flegal, 1994), and riverine systems (Ferrari & Ferrario, 1989) and observed in sediment chamber experiments (Kalnejais et al., 2007; Zago et al., 2000).

At Site A, the enhanced DPb and PPb concentrations near the seafloor coincided with persistently elevated turbidity signals (Figure 4), indicating particle resuspension and subsequent DPb and PPb remobilization from the sediments to overlying waters. The benthic Pb supply is supported by increased levels of the short-lived isotopes $^{223}\text{Ra}_{\text{xs}}$ and $^{224}\text{Ra}_{\text{xs}}$ (half-lives 3.66 and 11.4 days, respectively) near the seafloor (Figure 4), indicating recent sedimentary supply. A similar feature was observed on the continental slope along the canyon transect in November where salinity maximum associated with MOW decreased toward the continental shelf break (Figure 3a) from 35.74 (C01) to 35.61 (C04) ($\sim 1,000 \text{ m}$ depth); thus, DPb concentrations were also expected to decrease. Yet DPb concentrations remained unchanged (Figure 2a) and PPb concentrations were elevated, coinciding with increased $^{223}\text{Ra}_{\text{xs}}$, $^{224}\text{Ra}_{\text{xs}}$ and turbidity signals (Figures 4 and S7), indicating a sedimentary Pb source to overlying waters.

Benthic DPb remobilization has been reported by Noble et al. (2015) and Chien et al. (2017). To our knowledge, we provide the first clear evidence of a benthic Pb source supported by PPb and Ra field measurements. Furthermore, our results showed a strong positive correlation between DPb and PPb ($r^2 = 0.97$, $n = 12$), and DPb and LpPb ($r^2 = 0.97$, $n = 12$) at Site A and C03/C04 stations (Figure S8), indicating a dynamic equilibrium between the phases. Although little is known about biogeochemical processes facilitating DPb sedimentary release, we suggest that benthic remobilization could be facilitated through Pb association with Fe/Mn oxo-hydroxide precipitates (Allen et al., 1990; Bastami et al., 2015; Fernex et al., 1992; Kalnejais et al., 2007) with subsequent reductive dissolution of the solid Fe and Mn forms in sediments (Fernex et al., 1992). This mechanism is supported by elevated DFe and DMn concentration toward the

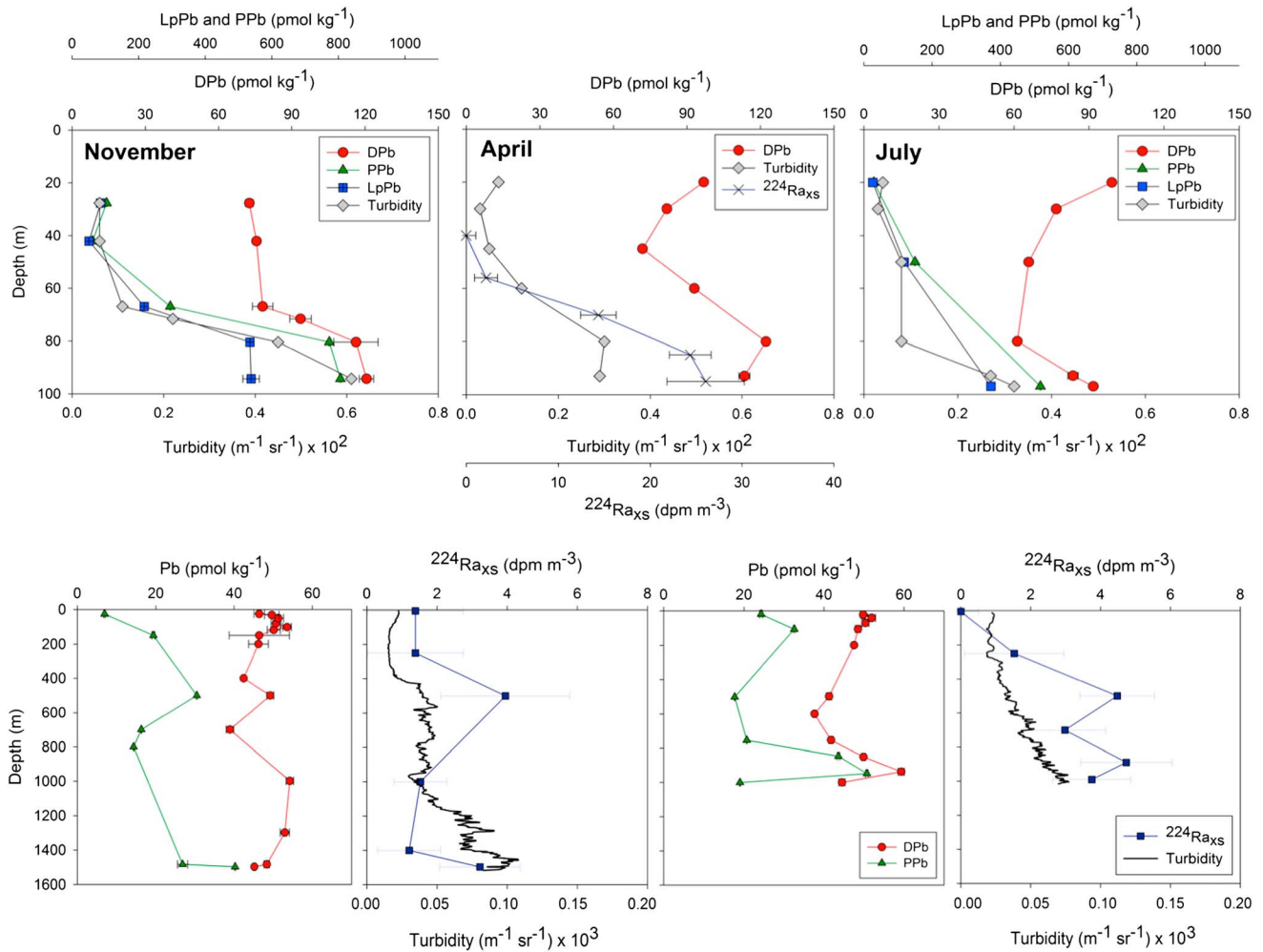


Figure 4. Upper panel: depth profiles of DPb (circles), PPb (triangles), LpPb (squares), turbidity (diamonds), and $^{224}\text{Ra}_{\text{XS}}$ (crosses) at Site A in November (DY018) (left), April (DY029) (middle), and July (DY033) (right). Bottom panel: depth profiles of DPb (circles), PPb (triangles), $^{224}\text{Ra}_{\text{XS}}$ (squares), and turbidity (black line) at station C03 (left) and C04 (right) in November (DY018).

seafloor (Figure S9) and a buildup of Fe(II) in sediments at Site A (Klar et al., 2017) but not at stations C03/C04. Potentially, DPb may be supplied in the form of small colloids deposited onto sediments (Muller, 1996; Sen & Khilar, 2006), with these Pb-enriched resuspended fine particles having a longer residence time in comparison to bulk sediment particles (Ferrari & Ferrario, 1989; Kalnejais et al., 2007).

We determined a benthic Pb flux to overlying waters of $27\text{--}41 \times 10^{-9} \text{ mole Pb m}^{-2} \text{ d}^{-1}$ ($n = 3$) at Site A in April (SI Text S4) using short-lived Ra isotopes (Moore, 2000). To our knowledge, this is the first benthic Pb flux estimate from field observations. Furthermore, this sedimentary Pb flux was up to 2 orders of magnitude higher than the atmospheric flux ($0.03\text{--}12.2 \times 10^{-9} \text{ mole Pb m}^{-2} \text{ d}^{-1}$) observed at nearby Penlee Point Atmospheric Observatory in 2015 (Figures 1 and S5; Arimoto et al., 2003). The benthic DPb flux was likely a result of accumulation in sediments of Pb deposited over time, while the recent relatively low atmospheric Pb flux reflects the implementation of strict European air emission regulations. We therefore stress that sediments containing legacy Pb will continue to serve as an important, if not the major source of Pb to overlying waters.

3.3. Recent Pb Sources in the NE Atlantic Shelf Region

A strong spatial and temporal variability in surface water DPb concentrations were observed over the various transects across the seasons. Elevated surface DPb concentrations ($36.1\text{--}122 \text{ pmol kg}^{-1}$) were observed on the continental shelf during all sampling seasons, with lower levels along off-shelf transects (Figure 2c). Elevated DPb concentrations observed in surface waters at Site A in April and July in comparison to

November indicated recent Pb inputs (Figure 4). A strong inverse correlation of DPb with salinity $r = -0.96$ (April, $n = 6$) and $r = -0.94$ (July, $n = 4$) between Site A and CS2 stations suggests a freshwater source of DPb, which could be continental runoff or wet deposition (Figure S10). On the continental slope, elevated DPb concentrations in the SML were observed along the canyon (T1_C) transect in November (50.8 ± 3.0 pmol kg⁻¹, $n = 20$) and at S08 and S09 stations on the spur (T2_S) transect. Lower DPb concentrations were observed during other seasons; 39.6 ± 6.9 pmol kg⁻¹ ($n = 23$, T1_C) and 35.0 ± 4.2 pmol kg⁻¹ ($n = 30$, T2_S) in April and 35.2 ± 3.8 pmol kg⁻¹ ($n = 9$, T1_C) and 37.4 ± 4.9 pmol kg⁻¹ ($n = 6$, T2_S) in July (Figures 2a and 2b). The distinctive changes in DPb over short spatial scale observed between our closely spaced stations (<20 km) suggest presence of different water masses with different DPb input histories over the last months to years. These observations coincided with distinct temperature and DMn concentration differences (Figure S11), which enforce this suggestion.

4. Conclusions

Our observations demonstrate the widespread impact of anthropogenic Pb emissions on the marine environment. The elevated Pb signal (46.6 ± 5.6 pmol kg⁻¹) in MOW is transported long distances (>2,500 km) at intermediate depths across the Northeast Atlantic following anthropogenic Pb emissions during the past century. Following implementation of stricter environmental regulations in Europe, this oceanic Pb signal will behave similarly to chlorofluorocarbons in that it is predicted to decrease with time. However, considering the residence time of Pb in the deep ocean of ~100 years (Nozaki & Tsunogai, 1976), the presence of recent Pb sources (>90 pmol kg⁻¹ in surface waters), and the “resupply” of Pb to the water column from sediments containing legacy Pb ($27\text{--}41 \times 10^{-9}$ mole Pb m⁻² d⁻¹), we expect the anthropogenic Pb signal to remain in the marine environment for decades to come. The role of the particulate phase in buffering Pb concentrations in the water column needs to be considered in the interpretation of oceanic DPb distributions due to a close relationship between the dissolved and particulate phases.

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