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Investigation of the factors that influence lead accumulation onto polyethylene: Implication for potable water plumbing pipes

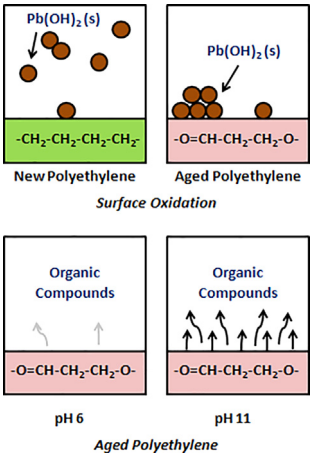
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HIGHLIGHTS

- This study examined Pb deposition onto new and aged plastic at different pH.
- Pb deposition kinetic rates were quantified.
- Aged LDPE leached more organic carbon than new LDPE.
- Pb precipitates had a greater affinity for aged LDPE compared to new LDPE.
- Aged LDPE polar groups acted as nucleation sites for $Pb(OH)_2$.

GRAPHICAL ABSTRACT



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ABSTRACT

The influence of polymer aging, water pH, and aqueous Pb concentration on Pb deposition onto low density polyethylene (LDPE) was investigated. LDPE pellets were aged by ozonation at 85 °C. ATR-FTIR and X-ray photoelectron spectroscopy (XPS) analysis of aged LDPE surfaces showed that a variety of polar functional groups (>C—O<, >C═O, >COO) were formed during aging. These functional groups likely provided better nucleation sites for $Pb(OH)_2$ deposition compared to new LDPE, which did not have these oxygen-containing functional groups. The type and amount of Pb species present on these surfaces were evaluated through XPS. The influence of exposure duration on Pb deposition onto LDPE was modeled using the pseudo-first-order equation. Distribution ratios of 251.5 for aged LDPE and 69.3 for new LDPE showed that Pb precipitates had greater affinity for the surface of aged LDPE compared to new LDPE. Aged LDPE had less Pb surface loading at pH 11 compared to loading at pH 7.8. Pb surface loading for aged LDPE changed linearly with aging duration (from 0.5–7.5 h). Pb surface loading on both new and aged LDPE increased linearly with increasing Pb initial concentration. Greater Pb precipitation rates were found for aged LDPE compared to new LDPE at both tested pH values.

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1. Introduction

Lead (Pb) exposure through tap water is a global concern due to the contaminant's acute and chronic health impacts. Exposure can delay children's mental development, cause behavior disorders, anemia, renal dysfunction, impaired hearing and postnatal growth [1–4]. In the U.S., lead in tap water remains a serious health risk, with large-scale lead drinking water poisonings occurring most recently in Washington, D.C. in 2004 (at up to 7500 μg/L) [2] and in Flint Michigan in 2015 (at up to 13,200 μg/L) [56].

Lead in building tap water can originate from the corrosion of plumbing materials such as lead service lines or lead containing brass appurtenances, solders, and fixtures [7–11]. Pb also can enter buildings from the water distribution piping network and then deposit and release from the surface of metal plumbing components (i.e., galvanized steel) [12]. Pb can be either in particulate or dissolved form and the concentration of each form can be influenced by water conditions (i.e., pH, hardness), as well as network operational conditions (i.e., residence time, temperature, corrosion inhibitor, flow rate) [11,13–18]. Pb levels in U.S. buildings have been reported to range from 0.2 to 13,000 μg/L in first draw water samples, and 0.2 to 7400 μg/L in flushed (30 s) water samples [18,19–22]. If water quality or operational conditions are altered, excessive metal leaching from piping components and pipe scales can occur as evidenced by Pb poisonings in Washington D.C. [2]. and Flint Michigan [5,6]. Due to the complex interactions of hydraulics, temperature, stagnation times, materials, and water chemistry, the fate of Pb in building plumbing is not well understood.

To reduce tap water metal drinking water levels some water utilities and building owners are choosing to replace lead service lines and indoor plumbing with plastic pipes. Galvanize iron coating can have Pb or the pipe scale can become contaminated with Pb from a lead source upstream such as a lead pipe. Even after the pipe is removed the galvanized pipe can remain a persistence source of lead [23,24]. Popular U.S. buried water distribution and service line plastic pipes are composed of high-density polyethylene (HDPE) or polyvinylchloride (PVC). Crosslinked polyethylene and chlorinated polyvinylchloride pipes also are being installed primarily for water service connections and within premise plumbing instead of copper (9% of replumbed households) [25]. Among these materials, it is well-known that PVC and cPVC pipes can leach metal heat stabilizers [26–28], while polyethylene pipes do not leach heavy metals. Ginige et al. reported Fe and Mn deposition onto HDPE pipe [29]. The limited examination of in-service plastic pipes [30,31] however, has revealed heavy metals (including Pb) can accumulate on plastic pipe surfaces (Table 1).

In a survey of 15 U.S. water utilities, very high loadings of As (1416–13,650 μg/g) and Pb (210–9681 μg/g) were detected on PVC drinking water distribution pipes per gram of total scale (Table 1) [32–34]. In our recent study Pb was detected on PEX pipes exhumed from residential green building with greatest magnitude on the service line pipe sample (0.5 mg/m^2 pipe inner wall surface area) [35–36]. Pb adsorption has been detected on drinking well PVC casings, and less Pb adsorption was found at lower pH values, but no adsorption differences were found between 50 μg/L and 100 μg/L Pb concentrations [28]. The role of plastic pipe surface aging on heavy metal sorption and deposition has gone unstudied, as most literature has focused on new (unaged) plastic materials, or plastic pipes in-service, where aging history is unknown and uncontrolled. Plastics pipes are known to undergo oxidative attack and degradation caused by drinking water chlorine disinfectants. Several oxidized functional groups, including >=O,>H, and >—O—C< have been detected on polymer surfaces exposed to chlorinated water [37,38]. These polar functional groups could influence the interactions of heavy metals present in the water with the plastic surface, and hence deserve scrutiny.

The literature contains a great number of studies that concern heavy metal adsorption onto plastic materials. Metal ion adsorption to new polyethylene resin pellets has been described using Langmuir and Freundlich isotherms [39]. Aged polyethylene pellets removed from a beach were found to have a greater absorption capacity for Cd, Co, Cr, Cu, Ni, and Pb than new pellets [39]. As water pH increased, adsorption of Cd, Co and Ni increased, Cr adsorption decreased, and Cu adsorption was unaffected [38]. No significant difference was found in the Pb loadings for new and aged polyethylene with increased river water pH from 7.5 to 10.5 at a Pb concentration of 5 μg/L [40]. No studies were found that identified why aged plastics adsorbed a greater mass of these metals than new plastics. Prior studies also did not fully explore the condition of the plastics examined. The working hypotheses in the literature are: (1) metal cations (e.g., Pb^{2+}, Ni^{2+}) or oxy-anions (e.g., $HCrO_4^-$ and CrO_4^{2-}) directly adsorb onto oppositely charge sites or neutral regions of the polymer surface [41–44] and (2) co-precipitation with or adsorption onto Fe and Mn hydrous oxides occurs. However, there has been no testing of these hypotheses.

Because plastic pipes are increasingly being used for potable water distribution and the literature indicates some plastics can adsorb and then release metals into the drinking water, this study was conducted. The goal of this study was to investigate drinking water quality and polymer surface characteristics that influence Pb deposition on and adsorption to LDPE. LDPE is not used for drinking water piping in the U.S., but it was previously used for pipe manufacture in Europe [45], and is widely used for domestic and commercial irrigation [46]. LDPE is a model material for investigating the factors that control contaminant-plastic interactions.

Specific objectives of the work were to: (1) Determine the role of LDPE aging on Pb surface loadings; (2) examine the influence of water pH on Pb precipitation over time for new and aged LDPE; (3) determine the precipitation rates for new and aged LDPE at pH values of 7.8 and 11; and (4) identify the types and amounts of Pb species present on new and aged LDPE surfaces.

2. Methods

2.1. Materials

Low density polyethylene (LDPE) pellets and films (0.1 mm thickness) were purchased from Sigma Aldrich, and LDPE sheets (3.1 mm thickness) were purchased from McMaster-Carr. LDPE plastic sheets and films were cut into 1×1 cm^2 squares prior to experiments. Pb ICP-MS standard (1000 mg/L) was purchased from RICCA Chemical Company. All water used in experiments was treated with an Ultrapure Milli-Q^{TM} (18MΩ*cm) system treated the water used for all experiments conducted in this study. More details described in SI 2-1.

2.2. LDPE aging with ozone and pellet conditioning

A Pacific Ozone Technology Lab 11 ozone generator was used to generate ozone (O_3) from pure (99.5%) molecular oxygen (Fig. SI-1). Polymer aging was conducted at 85 °C. DI water (250 mL) at pH 8 was placed in the glass flask contained 25 g of LDPE pellets and ozone was sparged continually into this flask. The ozone mass flow rate was measured at 3.84 mg/min using the method described by Blatchley et al. [47]. In order to remove residual organics from new and aged LDPE pellets, pellets were conditioned using DI water before Pb exposure and organic leaching experiments (SI 2-2).

2.3. Organic carbon release experiment

The release of organic carbon from new and aged LDPE, measured as total organic carbon (TOC) concentration, was monitored

Table 1
Studies found in the literature that report metal loadings on plastic potable water pipes.

Metal	Water Main						Service Line		
	nr (μg/g solid scale)	PVC	PVC	PVC	PVC	HDPE	PB	HDPE	HDPE
							(mg/cm^2 pipe surface area)		
As	1230	13,650	1416	7842	nr	49.9			
Ba	nr	nr	nr	nr	nr	446			
Cd	0.3	<4.8	36	375	nr	34.1			
Cr	nr	nr	nr	nr	nr	0.82			
Ni	18	6	110	137	nr	330			
Pb	114	210	4,667	9681	0.96 w%	25.1			
U	nr	nr	nr	nr	nr	113			
V	nr	nr	nr	nr	nr	137	0.097[a]	0.010[a]	0.041[a]
Al	4955	1906	1025	2329	19.72 w%	641			
Ca	57,790	22,939	28,859	4455	1.29 w%	2,856			
Mg	21,156	1492	1736	442	0.1 w%	2,435			
Fe	144,200	442,528	77,030	237,293	1.42 w%	5.5 w%			
Mn	882	5,142	290	1267	6.12 w%	2324			
P	10,001	15,410	850	1410	nr	728			
Zn	206	8915	535,783	541,564	0.35 w%	1846			
Ref.	[32]				[31]	[33]	[34]		

nr = not reported; All data reported "as is" from cited studies.
[a] Total mass of scale on inner pipe wall.

after 2, 6, 12, 24, and 48 h and at 5 days of exposure in water at pH 6, 7.8 and 11 (SI 2-3). Surface area to volume ratio S/V was selected as 0.85 cm^2/mL for the pellets.

2.4. Pb exposure procedure

To limit the type of Pb species present in aqueous solution, Pb exposure experiments were conducted in an oxygen-free environment (SI 2-4).

2.4.1. Five day Pb exposure experiments

Experiments were conducted at pH values of 7.8 and 11 for new and 10 h aged LDPE pellets during 5 day exposure period. In these experiments the initial aqueous Pb concentration was 300 μg/L. 13 mL of the Pb solution was added to 20 pellets in 50 mL PTFE bottles. During each experiment, three replicates (three 50 mL PTFE bottles) were removed at each exposure period. To quantify Pb on the pellets, the pellets were removed from the water and digested with 2% HNO_3 for 24 h. In order to distinguish the soluble fraction of Pb from its precipitate, the 300 μg/L Pb solution was filtered through 0.22 μm Millipore GSTF filter. Control experiments were also conducted and described in SI 2-4-1.

2.4.2. Influence of aging duration, organics content, and initial Pb concentration on Pb deposition

The role of polymer oxidation on Pb surface loading was examined. LDPE pellets were aged for 0.5, 2.5, 5, 7.5, and 10 h and exposed to Pb aqueous solution (300 μg/L) for 48 h. The pH was adjusted to 7.8. A solution of residual organic matter leached from the pellets was prepared. This solution was used to examine the effect of organics leached from the aged LDPE on Pb surface deposition. To prepare this solution, 10 h aged LDPE pellets were exposed to water at pH 11 for 5 days. Then the aqueous solution was removed and TOC was measured at 47 mg/L. This solution (TOC of 47 mg/L) was diluted to achieve 28 mg/L and 13 mg/L TOC levels. Then Pb (300 μg/L) was added to all three solutions. The pH was adjusted at 7.8. Then 10 h aged LDPE pellets were exposed to these solutions in triplicate for 48 h. The impact of initial Pb concentration on Pb surface deposition also was tested. New and 10 h aged LDPE pellets were exposed to initial Pb concentrations of 100, 600, 1,000, 3000 and 5000 μg/L. For each condition, the pH was adjusted to either 7.8 or 11.

2.4.3. Pb exposure to LDPE film prior to XPS analysis

New and aged LDPE plastic film (aged for 5 h or 10 h) was exposed to a Pb concentration of 5000 μg/L for 48 h at pH 7.8. The plastic film was not conditioned before the experiment. The S/V ratio was 1 cm^2/mL. After the exposure period, each film was removed from solution. They were dried in the glove box with slow purging N_2. After drying, each film was transferred to the XPS facility in a N_2 filled bottle.

2.5. ATR-FTIR analysis, water contact angle and BET surface area measurement

ATR-FTIR spectra were recorded on a Perkin-Elmer Spectrum 100 FT-IR Spectrometer from 4000 to 650 cm^{-1} with A 2 cm^{-1} resolution. Carbonyl bond intensity previously has been used to examine polymer oxidation [48–51]. In this study the degree of polymer oxidation (carbonyl index) was estimated by comparing the ratio of the carbonyl peak intensity (1710 cm^{-1}) to the reference peak intensity at 720 cm^{-1} [52]. The surface wettability changes were examined by application of static water contact angle titration. New and 10 h aged LDPE sheets were studied (SI 2-5). BET surface area measurement was conducted for 10 hr aged LDPE powder using a Micromeritics Tristar 3000 analyzer. The Brunauer, Emmet and Teller (BET) model was applied to the N_2-adsorption data to evaluate any surface area changes due to LDPE aging.

2.6. TOC measurement and Pb quantification

TOC was measured with a Shimadzu TOC-L CPH/CPN in accordance with USEPA method 415.1. Standard calibration curves were constructed from 0 to 100 mg TOC/L (r^2 = 0.995) using potassium acid phthalate as the standard [53]. Pb surface loadings and residual floc were quantified with a Perkin Elmer Elan-DRCE II Inductively Coupled Plasma Mass Spectrometer (ICP-MS), details are explained in SI 2-6.

2.7. FE-SEM and XPS analysis

Field emission scanning electron microscopy (FE-SEM) imaging was conducted to examine the polymer surface morphology changes. New and 10 h aged LDPE sheets were used for FE-SEM imaging. The FE-SEM instrument was FEI Nova Dual Beam with an Oxford 80 EDX detector. Before analysis, sheets were coated with

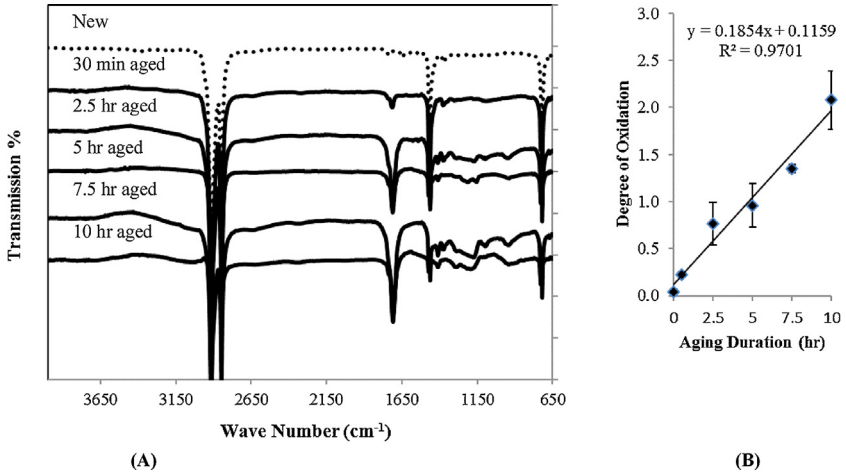


Fig. 1. (A) ATR-FTIR spectra of new and aged LDPE, (B) Degree of oxidation over the aging duration (error bars represent standard deviation for three samples).

gold and palladium under vacuum conditions. X-ray photoelectron spectroscopy (XPS) measurements were conducted using a Kratos Axis Ultra DLD spectrometer with monochromatic Al Kα radiation (hυ = 1486.6 eV) (SI 2-7).

3. Results and discussion

3.1. Low density polyethylene (LDPE) degradation

3.1.1. ATR-FTIR analysis

Several oxidized surface functional groups were identified on aged LDPE (Fig. 1). The 1710 cm^{-1} carbonyl functional group revealed the LDPE pellets were oxidized to some degree. This functional group has been previously found on aged HDPE water pipes exposed to disinfected drinking water [54]. For LDPE samples aged 2.5 to 10 h, a wide peak was observed at 938 cm^{-1}. This wide peak was attributed to >—O—C< and was not found on new LDPE [38]. Several other peaks were observed for 2.5 to 10 h aged LDPE that may be attributed to >—O—C< functional groups (1096, 1154, 1411, 1214, and 1291 cm^{-1}) [36]. Formation of the >—O—C< bond was also detected on polyethylene water pipes exposed to hot chlorinated and nonchlorinated waters [38,55]. In our recent study we examined one year old PEX pipes exhumed from a residential building, and we have found the absorption peak at 1263 cm-1 (>C—O—C<), at 1024 cm-1 and 1100 cm-1 (>C—O<) for pipes exiting the water heater tanks and also for the cold water pipes [35,36]. With increased aging duration (from 0 to 10 h), the amount of oxidation increased linearly (r^2 = 0.97) (Fig. 1B**).** Using the FT-IR data reported by Whelton et al. [37], for the water utility high-density polyethylene (HDPE) pipes exhumed from the field after 7, 20, and 25 year of operation, we calculated the carbonyl indexes as 0.05, 0.15 and 0.38, respectively. The carbonyl index for the 10 h aged LDPE pellets was 2.08 and was more than 5 times greater than the carbonyl index for the PE pipe in service for 25 years. The carbonyl index for 25 year old PE pipe is between the carbonyl indexes found for 30 min (0.2) and 2.5 h (0.76) aged LDPE pellets. It should be noted that LDPE resin compositionally and structurally is different from HDPE pipes removed from the field, and it is unknown if this comparison is fully appropriate. ATR-FTIR analysis of LDPE sheet and film are discussed in SI 3-1.

3.1.2. Influence of polymer aging on surface wettability, morphology and surface area

Water contact angle measurement showed significantly lower water contact angles (*p-value* <.01) for aged LDPE (98.6° ± 1.6°) compared to the new LDPE (104.6° ± 2.8°). This finding is likely due to the aged LDPE's surface being more hydrophilic. It is well-known that water adsorbs to polar polymer surfaces, and oxidation can increase wettability [56]. pH (2–12) did not consistently influence water contact angle for aged LDPE (*p-value*. 076). Water contact angles were greater for new LDPE at pH values of 8, 9, and 10 (*p-value*. 006), but not at any other pH values (Fig. SI-3). No surface morphology differences were observed for new and aged LDPE by FE-SEM **(**Fig. SI-4**)**. BET surface area was significantly greater for 10 h aged LDPE powder (0.12 ± 0.0 m^2/g) compare to new LDPE powder (0.03 ± 0.0 m^2/g).

3.2. Organic carbon release by new and aged LDPE

Organic carbon release from aged LDPE pellets was significant over the five day experiment. The observed TOC concentration by aged LDPE was a function of pH. As Fig. 2 shows, organic carbon released from the aged LDPE was significantly greater at pH 11 (>48 mg/L) than pH 6 (<3 mg/L) and pH 7.8 (<1.3). For new LDPE, organic carbon release was low (<1.7 mg/L at all pH conditions). The release of organic carbon is likely altering the polymer nanostructure and could influence heavy metal interaction with the plastic surface. LDPE can hydrolyze at room temperature [57]. By increasing pH from 5.0 to 8.1 polycarbonate hydrolysis was shown to result in greater chemical leaching [58]. Greater carbon release could be due to the aged LDPE degradation at high water pH. Excess OH^- ion could attack vulnerable bonds created from chemical oxidation of polymer chains. Oxidized carbon functional groups formed on polymeric chain after ozone aging made the polymer vulnerable. As a result electron withdrawing by oxygen caused the positive charge for adjacent carbon atom. This positively charged carbon atom can be attacked by OH^- ion present in water [59,60]. High reactivity of the radicals formed due to OH^- attack could initiate unsaturated bond formation [61]. In the present study exposure of aged LDPE to pH 11 water caused the vinyl bond formation (>C=C<). This bond was not detected after polymer exposure to either pH 6.0 or 7.8 (Fig. SI-5). For new LDPE, significantly lower levels of organic carbon was released into the water at pH 11 compare to the aged LDPE. For new LDPE, there is negligible covalently-bound oxygen in the polymer that would make the LDPE more susceptible to attack by OH^- at pH 11.

3.3. Pb precipitation on new and aged LDPE

3.3.1. Pb speciation in DI water

Pb chemical speciation in DI water when no oxygen was present, was determined through solving Pb hydroxyl complexation and

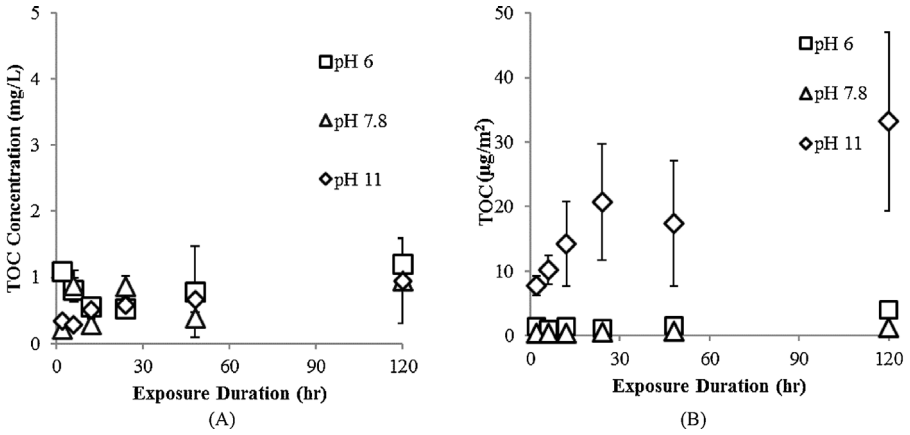


Fig. 2. Organic release over the time for (A) New and (B) 10 h aged LDPE (Mean ± std).

solubility equations (Table SI-1, SI 3-2**).** Based on Pb hydroxyl complexation and solubility equations, the concentration of soluble Pb species was 19.0 μg/L at all tested Pb initial concentrations (100 μg/L–5000 μg/L) at pH 7.8. It was almost 0.0 for all tested Pb initial concentrations (100 μg/L–5000 μg/L) at pH 11 (Fig. SI-7). This soluble Pb level was neglected compare to total Pb concentration (100 μg/L–5000 μg/L). In the present work, Pb was precipitated onto new and aged LDPE, as the major portion of Pb species in the solutions was solid. To confirm this, 300 μg/L Pb solution (pH 7.8) was filtered through 0.22 μm filter and only 2.3% of total Pb concentration was found in the filtered solution (soluble Pb). So the majority of Pb was solid.

3.3.2. *Pb precipitation on LDPE over 5-day exposure period*

Pb precipitation on new and aged LDPE during the 5 day exposure period was examined using Pseudo-first-order model (Eqs. (1) and (2)). In this study we considered Pb precipitation onto LDPE as a mass transfer process, since pb species only transferred from solution to LDPE surface and no chemical reaction occurred.

$$\frac{dq}{dt} = -K(q - q_{eq}) \tag{1}$$

$$q = q_{eq}(1 - e^{-Kt}) \tag{2}$$

$$t_{1/2} = \frac{\ln 2}{k} \tag{3}$$

where q and q_{eq} are the Pb surface loading on LDPE (μg/m^2) at the time t (h) and at the equilibrium. The first-order mass transfer constant was shown by K (h^{-1}), and it was determined using the experimental data. The half-life ($t_{1/2}$) represents the time required for polymer to uptake half of equilibrium Pb surface loading and calculated using Eq. (3).

In this study the authors defined the distribution ratio (*D*) as shown in Eq. (4). The distribution ratio shows Pb distribution between LDPE surface and aqueous solution.

$$D\ (\text{mm}) = \frac{Pb\ Surface\ loading\ (\frac{\mu g}{m^2})}{Pb\ floc\ left\ in\ solution\ (\frac{\mu g}{L})} \tag{4}$$

3.3.2.1. Effect of polymer aging on Pb precipitation kinetics. Aged LDPE had a greater Pb surface loading than new LDPE and loading was a function of exposure duration (2 h–5 days) (Fig. 3A). Pb loading on aged LDPE was greater than new LDPE during all exposure periods. Aged LDPE achieved 45% of its surface loading equilibrium during the first 2 h exposure. But for new LDPE only 1.7% of equilibrium was obtained in the first 2 h. As Table 1 shows the half-life ($t_{1/2}$) determined for new LDPE (46.21 h) was greater than aged LDPE (33.00 h). At pH 7.8, Pb surface loading increased sharply for both new and aged LDPE during the first 12 h exposure period, and gradually approached equilibrium. Pb surface loading was quantified for LDPE pellets aged from 30 min to 10 h, after 48 h exposure to Pb aqueous solution. Pb surface loading increased linearly (r^2 = 0.966) with increased aging duration up to 7.5 h, then it became constant (Fig. 3B). Polar functional groups on the aged LDPE (>C=O, >—O,>—O—C<) could be responsible for greater Pb surface loading compared to new LDPE. Other researchers have reported detecting greater Pb mass loading on aged polyethylene pellets compared to the new polyethylene pellets [37,38].

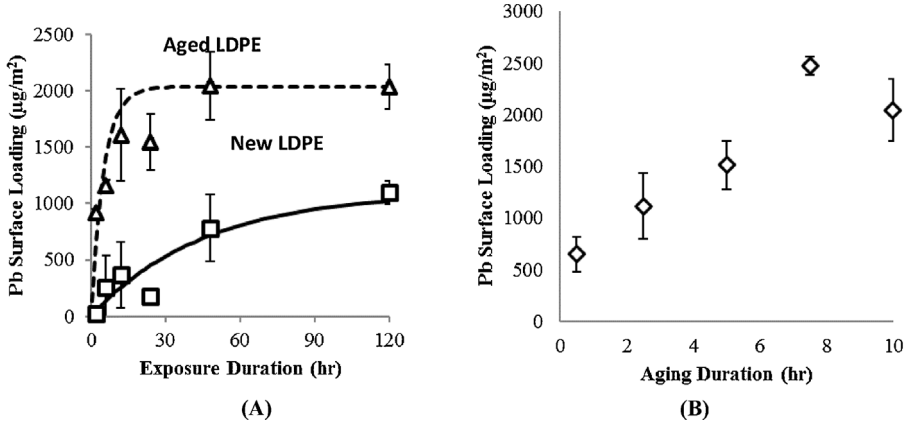


Fig. 3. (A)Pb surface loading on new and aged LDPE over the time at pH 7.8 (Mean ± std)**, (B)** Influence of LDPE aging duration on Pb surface loading.

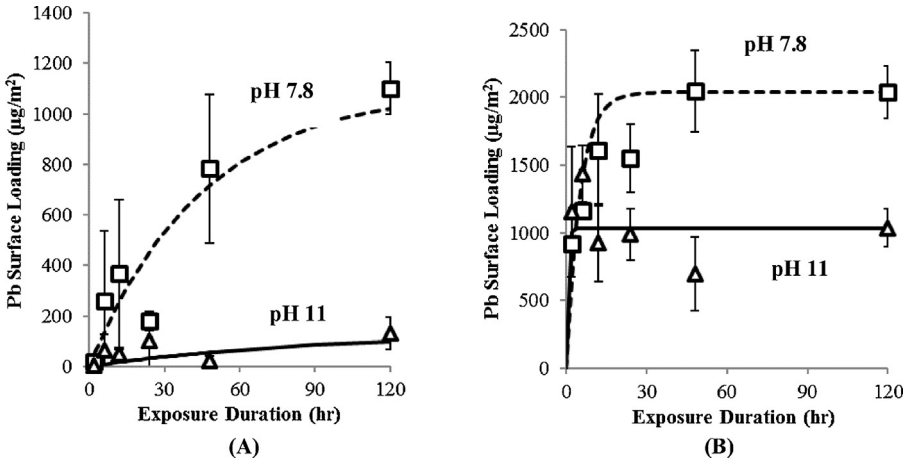


Fig. 4. Pb surface loading on (A) new and (B) aged LDPE over the time at pH 7.8 and 11 (Error bars are standard deviation for three replicates).

Table 2
The behavior of Pb precipitation over the time on new and aged LDPE at pH 7.8 and 11.

LDPE	Water pH	Pseudo first order model			Distribution ratio (mm)
		K (h^{-1})	R^2	$t_{½\,(h)}$	
New	7.8	0.022	0.999	31.65	69.26
	11	0.011	0.980	60.27	1.37
Aged	7.8	0.195	0.996	3.55	251.50
	11	1.700	0.999	0.40	12.97

3.3.2.2. Effect of water pH on Pb precipitation over the time. As Fig. 4 shows less Pb surface loading was found on both new and aged LDPE at pH 11 compared to pH 7.8. Despite Pb surface loading great changes due to the pH increase from 7.8 to 11 much lesser changes occurred in Pb speciation. At pH 7.8, 93.7% of the lead was present as $[Pb(OH)_2]_{(s)}$ and at pH 11, the lead was 100% $[Pb(OH)_2]_{(s)}$ (Fig. SI-6). Pb surface loading on both new and aged LDPE was time dependent. While the pH 11 condition was outside recommended U.S. drinking water pH standard (6.0–8.5), in some cases long water stagnation periods in iron pipes cause pH increase up two units and approach pH 11 [62,63].

The new LDPE first-order mass transfer constant was 2-fold greater at pH 7.8 compare to pH 11 (Table 2). In contrast, *K* for aged LDPE was more than 8-fold greater at the high pH condition. For aged LDPE the half-life ($t_{1/2}$) was significantly less at pH 11 (24 min) compare to pH 7.8 (3.55 h). According to distribution ratio (*D*), Pb precipitates demonstrated a greater affinity toward aged LDPE compared to new LDPE. At the high pH condition, the distribution ratio (*D*) was 50-fold lower than at pH 7.8 for new LDPE, and it was 19-fold lower for aged LDPE.

Pb surface loading was significantly reduced due to the increased pH after 48 h for new LDPE and after 24 h for aged LDPE. As mentioned earlier, a great amount of organic carbon was released from aged LDPE due to the exposure at pH 11. The influence of these organic chemicals on aged LDPE Pb surface loading was examined. The results showed that the presence of organic material leached by aged LDPE pellets reduced aged LDPE Pb surface loading (Fig. SI-8).

3.3.3. Influence of Pb concentration on its precipitation on LDPE

At all Pb concentrations, greater Pb surface loading was detected for aged LDPE compare to new LDPE. Pb mass on polymer surface was graphed versus the residual Pb mass remaining in aqueous solution, where the pellet surface area was 1108 mm^2 and aqueous solution volume was 13 mL (Fig. 5). Less Pb surface loading at higher pH, could not be related to the Pb species very small change due to pH changes. The high organic material release for aged LDPE at pH 11 may have caused lower Pb surface loading. No study was found that has investigated the influence of organic materials, released from plastics, on Pb adsorption. In other studies, the the type of organic material present, their concentration, contact time, and metal concentration can influence Pb adsorption to soil [64]. As described at Section 3.3.2.1 with increased TOC level, less Pb surface loading was found for aged LDPE (Fig. SI-8). By applying a linear model for the results, the slope was defined as the precipitation mass rate. This mass rate was greater for aged LDPE at both pH values (at pH 7.8 as 4.58 and at pH 11 as 0.54) compared to mass

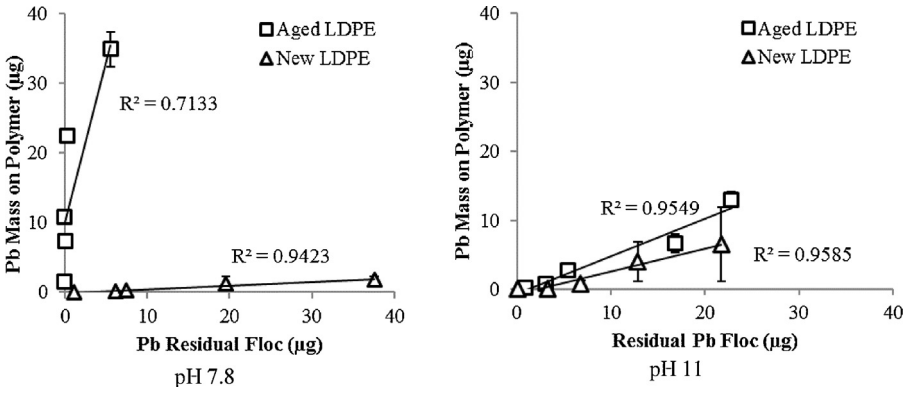


Fig. 5. Pb mass loading on LDPE versus Pb residual mass in the solution at pH 7.8 and 11 (error bars are standard deviations for three replicates).

Table 3
Elemental atomic concentration % in sample of LDPE.

Sample Name	C 1s				O 1s	Pb 4f
	C—C	C—O	C=O	O—C=O		
New LDPE	98.3				1.7	0.0
	98.4	1.6	0.0	0.0		
New LDPE + Pb	99.3				0.7	0.0
	99.1	0.9	0.0	0.0		
5 h aged LDPE	90.6				9.4	0.0
	91.5	3.7	2.7	2.1		
5 h aged LDPE + Pb	89.0				10.6	0.5
	91.6	3.4	3.0	2.0		
10 h aged LDPE	92.1				7.9	0.0
	92.2	4.1	2.0	1.7		
10 h aged LDPE+ Pb	83.8				16.0	0.1
	86.1	4.9	4.1	4.9		

rates observed for new LDPE (at pH 7.8 as 0.05 and at pH 11 as 0.33). For aged LDPE, the precipitation mass rate reduced with increased water pH from 7.8 to 11. However, for new LDPE with increased pH from 7.8 to 11 this mass rate increased. Increased Pb surface loading with increased aging duration supports this hypothesis (Fig. 3B).

The surface of both new and aged LDPE pellets served as the nucleation sites for Pb precipitation and crystal growth. Different Pb mass loadings for new and aged LDPE could be due to the different number of nucleation sites present on each polymer. The aged LDPE oxidized surface provided more nucleation sites for Pb precipitation compared to new LDPE. For aged LDPE at pH 11, a high mass of organic compounds were released into the contact water. It is possible these organic compounds decreased Pb surface loading by (1) making non-adsorbable complexes with Pb, and (2) competed with Pb for available surface sites [65]. Both phenomena could have resulted in less Pb precipitated on the polymer surface.

3.4. Polymer surface chemistry analysis using XPS

XPS analysis was conducted on LDPE films to understand the influence of polymer aging on Pb precipitation. As Fig. 6 shows the C1s peak at 284.8 eV [66,67] is the dominant feature associated with the LDPE structure. A weak O1s peak at 531.9 eV [68] was observed for new LDPE and its intensity was greater for aged LDPE indicating chemical oxidation had occurred. The weak O1s peak for the new LDPE sample was probably due to intrinsic surface oxidation. All new, 5 h and 10 h aged LDPE samples were exposed to Pb aqueous solution (5000 μg/L), but Pb 4d and 4f peaks in survey spectra were only detectable on aged LDPE samples. This finding indicates a persistent Pb residue resulting from LDPE aging/oxidation.

The atomic % of C, O, Pb were determined with 0.1% uncertainty based on the peak area from XPS analysis (Table 3). Aged LDPE samples had a greater percentage of oxygen present on their surface than new LDPE (Table 3). As ICP-MS results show, a greater amount of Pb was deposited onto aged LDPE compared to the new LDPE.

To verify that the observed oxygen percentage increase for aged samples was due to polymer oxidation, high-resolution C1s spectra was measured. Carbon atom chemical state was also studied. Analysis of C1s spectra for aged LDPE samples revealed four different peaks at 284.7 eV (>C—C<), 286.4 eV (>C—O), 287.7 eV (>C=O) and 289.2 eV (>O—C=O) (Table 2, Fig. SI-9) similar to the peaks found by Briggs et al., (2003) who analyzed oxidized LDPE [67]. No differences were found between the surface oxidation of 5 h and 10 h aged LDPE. As the 10 h LDPE may have experienced additional aging relative to the 5 h sample, but the oxidation region measured by XPS was saturated after 5 h aging. ATR-FTIR analysis revealed greater degree of oxidation for 10 h aged LDPE film compare to 5 h aged LDPE film (Fig. SI-10). ATR-FTIR analysis depth is around 0.5 to 5 μm however, XPS is only sensitive to the outermost 8–10 nm of material.

There was no change in the carbon envelop for new LDPE before and after the Pb solution treatment. From this result, the Pb solution treatment did not itself impact the LDPE. XPS analysis, coupled with the ATR FTIR results, indicates that deposited Pb is likely electrostatically bound to the oxidized carbon. The doublets characteristic of lead derived from spin-orbit split appeared respectively, at 139 eV (assigned to Pb 4f7/2) and at 143.9 eV (assigned to Pb 4f5/2) after exposure of 5 h aged LDPE sample to Pb aqueous solution (Fig. SI-11). The peak observed at 139 eV agrees with the 138.6 eV value reported for $Pb(OH)_2$ [70,71]. The binding energies of C and O were also not changed by Pb(II) adsorption. This indicated no significant electron transfer occurred between unoxidized LDPE and Pb(II). As XPS and ATR-FTIR results demonstrated, the aging process clearly oxidized the LDPE, creating electrostatic dipoles at the surface. Results from this study indicate that Pb precipitation on LDPE was due to the electrostatic interaction between polymer charged surface and $Pb(OH)_2$. Aged LDPE with variety of polar groups (>C—O<,>=O, >OO) on its surface provided more nucleation sites for $Pb(OH)_2$ deposition than new LDPE.

3.5. Implication for potable water plumbing pipes

Nationwide, regulators, water suppliers, and building officials generally believe plastic pipes are inert and that these materials will solve their drinking water problems associated with heavy metals. Plastics can sorb heavy metals, and material characteristics and water chemistry conditions controlling their accumulation and release have yet to be studied. As a result, regulators and water suppliers have limited ability to minimize the risk of metals being adsorbed or precipitated onto, and subsequently released from plastic pipes into drinking water. Changes to drinking water chemistry could ultimately contribute to drinking water contamination associated with heavy metals deposited onto plastic pipes. Results of this study provide a foundation for continued exploration of heavy metal fate in water infrastructure. Results are applicable to where new or aged plastic plumbing materials are exposed to Pb from fixtures and pipes. Results show that water pH and plastic aging influences the magnitude of Pb deposition onto the plastic. Aging may cause plastic pipes to become more susceptible to Pb contamination.

4. Conclusion

The influence of polymer aging, water chemistry and Pb initial concentration on Pb deposition onto LDPE was examined. Pb precipitation over 5 day exposure period for new and aged LDPE was determined at pH 7.8 and 11. The type and amount of Pb species and carbon functional groups present on new and aged LDPE surface were determined through XPS. Both ATR-FTIR and XPS analysis of aged LDPE showed that several oxidized carbon functional groups formed on the polymer surface. Greater Pb surface loading was found for aged LDPE compare to the new LDPE at both water pH conditions. Pb surface loading increased linearly ($r^2 = 0.966$) with longer aging duration, up to 7.5 h. Polar functional groups formed due to the aging could be responsible for greater Pb surface loading on aged LDPE compared to the new LDPE. Modeling Pb transfer from solution to LDPE surface with Pseudo-first-order equation showed a greater rate of Pb deposition onto aged LDPE compared to new LDPE. Less Pb surface loading on both new and aged LDPE was found after 5 days at pH 11 compared to pH 7.8. XPS analysis revealed several oxidized functional groups were formed on aged LDPE, these functional groups likely provided better nucleation sites for $Pb(OH)_2$ deposition compared to new LDPE.

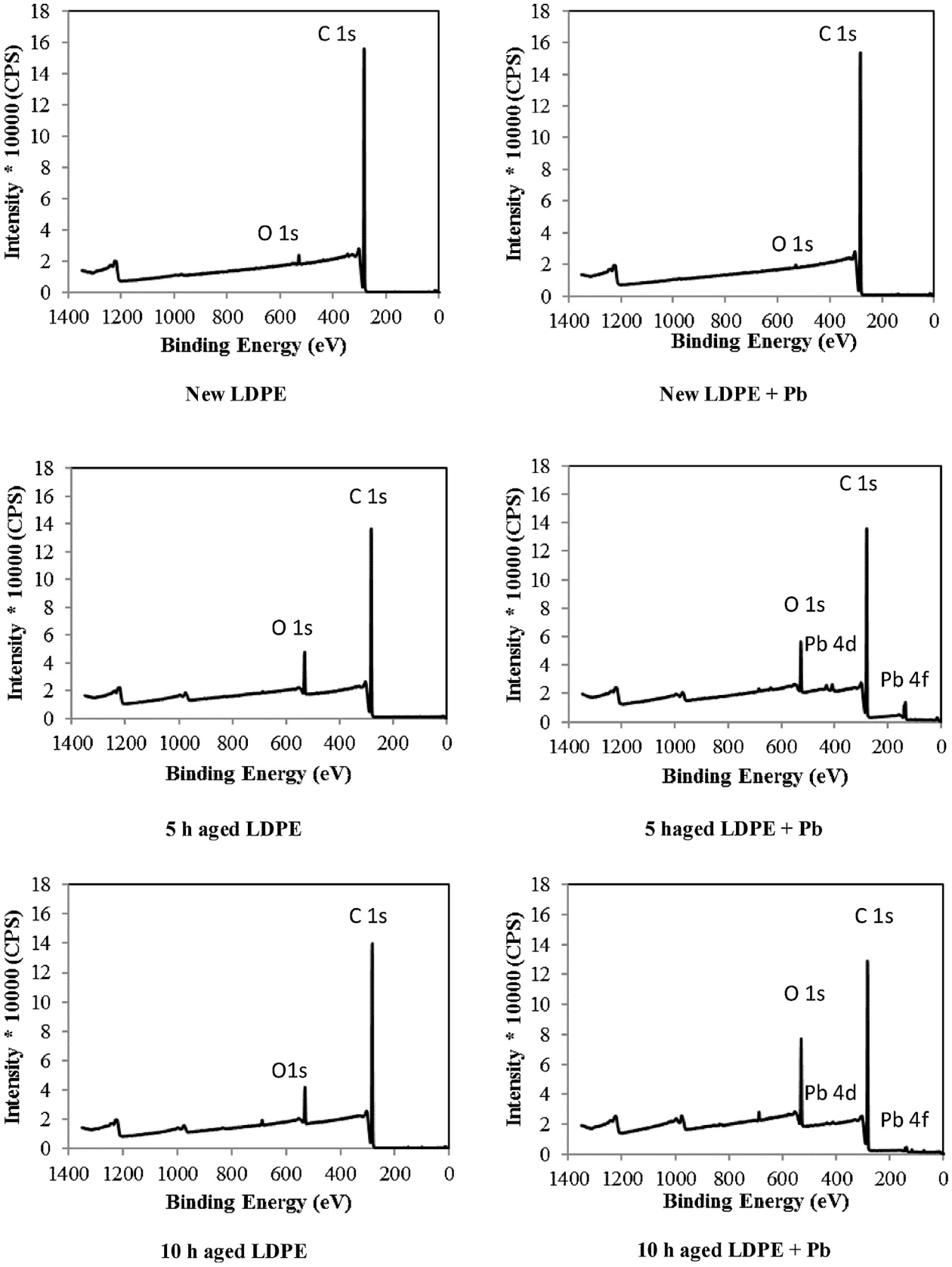


Fig. 6. XPS survey spectra for LDPE samples before and after exposure to Pb.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2017.12.066

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