



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

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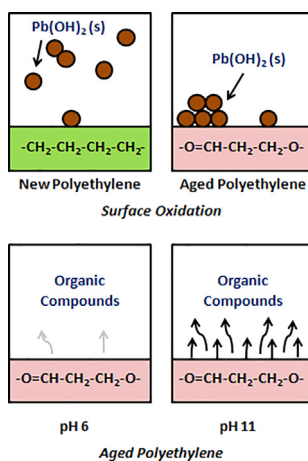
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HIGHLIGHTS

- 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 $\mu\text{g/L}$) [2] and in Flint Michigan in 2015 (at up to 13,200 $\mu\text{g/L}$) [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 $\mu\text{g/L}$ in first draw water samples, and 0.2 to 7400 $\mu\text{g/L}$ 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 $\mu\text{g/g}$) and Pb (210–9681 $\mu\text{g/g}$) 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 $\mu\text{g/L}$ and 100 $\mu\text{g/L}$ 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 $\mu\text{g/L}$ [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 $1 \times 1 \text{ cm}^2$ 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™ (18M Ω ·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

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