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Assessing the sustainability and safety of polyethylene terephthalate (PET) liners for lead service lines (LSL) upgrades

Amélie Surmont [a,*], Laura Rowenczyk [a], Ivan Viveros Santos [b], Fatemeh Hatam [a], Anne-Marie Boulay [b], Michèle Prévost [a]

[a] Industrial Chair in Drinking Water, Department of Civil Engineering, Polytechnique Montreal, Montreal H3C3A7, QC, Canada

[b] International Reference Center for Life Cycle Assessment and Sustainable Transition (CIRAIG), Polytechnique Montreal, Montreal H3C 3A7, QC, Canada

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ABSTRACT

Polyethylene Terephthalate (PET) liners have been proposed by industry as a more cost effective and less disruptive alternative to lead service lines (LSL) replacement. However, concerns have been raised about their aging under real-use conditions and their potential health and environmental impacts. In this study, two approaches were implemented. First, bench and pilot scale experiments were carried out to investigate the aging of a PET liner under conditions that simulate normal usage. Results show early surface oxidation and leaching of potentially hazardous metals (lead, zinc and titanium). No short-term fragmentation of the PET liner into microplastics was observed. Next, a life cycle assessment (LCA) compared the health and environmental impacts of three LSL upgrade alternatives: PET liners and full LSL replacement using either copper or PEX pipes. The installation phase was shown to be the main contributor to impact scores, while the benefits of PET liners are highly dependent on their lifespan. PEX pipes installed by torpedoing lower impacts as compared to PET liners and copper pipes for equal lifespan, while the use of PET liners remains less impactful regarding human health and ecosystem quality when a complete excavation is needed. LCA derived global human health effects due to ingestion of leached metals from PET liners, copper pipes and unreplaced LSL, and showed that PET liners and copper pipes significantly reduce health impacts by 14 and 80 DALY respectively compared to unreplaced LSL. Finally, the Integrated Exposure Uptake Biokinetic (IEUBK) model was used to assess the impact of lead exposure specifically for drinking water ingestion. Estimated Blood Lead Levels (BLL) in children and infants in households with long unreplaced LSL was up to 263.7% and 207.8% greater compared to either replacement with copper pipes or rehabilitation with PET liners, showing the clear benefits of corrective action. Combining experimental results, LCA and biokinetic modeling provides actionable information for utilities to select the best upgrade options, considering environmental, health and practical constraints, whilst identifying remaining data gaps. The relative benefits of PET liners should be carefully evaluated considering their lifespan under real-life conditions, the complete replacement costs after failure, and the growing evidence of micro- and nanoplastics (MNP) risks.

1. Introduction

1.1. Context

Lead service lines (LSL) represent a significant remaining source of lead exposure through drinking water consumption. Exposure to low levels of lead can result in irreversible neurological and behavioral disorders in young children (Health Canada, 2013). Health Canada (2019a) lowered the guideline for the maximum admissible lead concentration in drinking water from 10 to 5 μg/L. Polyethylene terephthalate (PET) liners, certified to the NFS61 standard, have been proposed by commercial vendors as a more cost-effective and less disruptive alternative than complete LSL replacement with new copper or plastic pipes. PET is a semi-aromatic linear polyester widely used due to its mechanical, chemical and thermal properties, and cost-effectiveness (Panowicz et al., 2021). During installation in a LSL, the liner is inserted and thermoformed with pressurized hot water (approx. 90 °C, 28–43 psi) (Randtke et al., 2017), resulting in a protective layer against lead. Some studies, mostly industry reports, show the potential of PET liners to effectively reduce lead leaching from LSL at

* Corresponding author.
E-mail address: amelie.surmont@polymtl.ca (A. Surmont).

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short term, but long-term performance, resistance and leaching data are lacking (Breault, 2014; Randtke et al., 2017). Aging of plastic liners in direct contact with drinking water raises concerns about their potential degradation over time. Operating conditions, mechanical stress or chemical exposure may release hazardous chemicals and shorten plastic pipes lifespan (Dear and Mason, 2001; Fadel, 2022).

1.2. Plastic aging

In water distribution networks, polyolefin pipe degradation progresses through three main stages (Vertova et al., 2019; Whelton and Dietrich, 2009) influenced by physical factors (temperature, freezing), chemical factors (water type, pH, oxidants) and exposure time. Even if PET degradation due to these factors has not yet been well demonstrated, modifications are expected to resemble those encountered for other polyolefins like polyethylene (Randtke et al., 2017). The first step consists of surface oxidation of the polymer through hydrolysis, thermal, freezing and chlorine effects, often attested by carbonyl and vinyl groups formation (Chamas et al., 2020; Edge et al., 1991; Eng et al., 2011; Khan et al., 2022; Rowenczyk et al., 2024; Vertova et al., 2019). In an advanced stage of degradation, which typically occurs under aging above the polymer's glass transition temperature (Tg, > 80 °C), chain fractures and molecular reorganization can appear in the amorphous phase, leading to an overall increase in crystallinity of the aged material (Dear and Mason, 2001; Hassinen, 2004; Panowicz et al., 2021; Whelton and Dietrich, 2009). In the final stage, increased crystallinity makes materials brittle, leading to microcracks propagation, reducing tensile strength and causing pipe failure (Hassinen, 2004; Vertova et al., 2019; Whelton and Dietrich, 2009).

Pipe degradation can be significantly increased by chlorine-based disinfectants added to water distribution systems by rapidly consuming antioxidants in the amorphous content, leading to the pipe's inner surface oxidation (Castillo Montes et al., 2012; Hassinen, 2004; Vertova et al., 2019). Chlorine diffuses into the polymer matrix, potentially attacking both the additives, especially the antioxidants, and the polymer simultaneously (Bredács et al., 2018). High temperatures can also accelerate PET aging, especially above the Tg (Pirzadeh et al., 2007), and can act as a catalyst for chlorine oxidation (Bredács et al., 2018). Finally, pH contribution in polymer aging, though poorly investigated, could significantly affect chlorine effect due to its speciation (Castillo Montes et al., 2012; Pirzadeh et al., 2007).

1.3. Additives and plastic particles leaching

Several plastic pipe studies have identified leaching of additives (compounds added to plastics to enhance thermal, mechanical or chemical properties) such as phthalates, phenols, organophosphates, salts (Ca, Mg, K) and metals (Ti, Fe, Pb, Sb) (Al-Malack, 2001; Diera et al., 2023; Fadel, 2022; Faust et al., 2017). Previous research conducted by Lane (2015) and Breault (2014) on PET liners showed respectively no phthalate and low antimony leaching at 20 °C over 4 days. However, other studies on PET bottles showed that prolonged storage increases the concentration leached to the water, with the effects of temperature (high and low) and pH depending on the compounds studied (Al-Saleh et al., 2011; Chapa-Martinez et al., 2016; Keresztes et al., 2013; Westerhoff et al., 2008). Additionally, plastics can fragment into micro- (1–5 mm) and nanoplastics (1–1000 nm) (MNP) (Gigault et al., 2018), whose health and environmental effects have not yet been fully assessed (Yong et al., 2020). MNP generation from water pipes (Zhang et al., 2022) and liners (Li et al., 2019) can occur during installation and use, with particles released into air and water. Since these MNP display advanced oxidation, increased crystallinity and reduced hydrophobicity compared to the initial plastic polymer, they can represent a significant health risk by sorbing other pollutants and being more bioavailable (Gigault et al., 2021). However, the leached and fragmented contaminants are still difficult to quantify considering the great chemical variability of additives, field operation conditions and advanced degradation of the polymer.

1.4. Environmental and human health impacts

In light of current climate change, biodiversity loss and pollution-related health impacts challenges and as part of an eco-responsible decision-making process, it is essential to expand the discussion on the sustainability of pipe technologies beyond material aging studies. Using comprehensive and robust tools like Life Cycle Assessments (LCA) allows for a fair comparison of the potential environmental and health impacts of production, transport, use and end-of-life management of various technological solutions considered for a similar application and to identify environmental hotspots (Hajibabaei et al., 2018; Hellweg et al., 2023). In this study, LCA can therefore be an additional important asset in evaluating PET liners relevance as alternatives for LSL upgrades. Previous studies comparing various water pipe materials and installation methods reveal significant differences in environmental and health impacts (Alsadi, 2019; Asadi et al., 2016; Hajibabaei et al., 2018). Plastic pipes show much lower impacts than metal pipes, mainly due to lower material and energy requirements during production (Hajibabaei et al., 2018). Additionally, trenchless installation methods (torpedoing and coatings) offer environmental benefits over traditional trenching, as they use less equipment, require shorter installation times, and involve minimal excavation (Alsadi, 2019).

This study aimed to assess the physico-chemical modifications of a PET liner under accelerated aging conditions in laboratory (PET coupons static aging) and pilot (PET lined pipes static and dynamic aging) experiments. Additionally, it sought to verify the health and environmental benefits of trenchless LSL upgrades through a comprehensive LCA, including water quality evaluation, and an analysis of the potential impact of leached lead on young children's BLL. The ultimate objective is to help stakeholders identify durable, eco-friendly options and understand system constraints for reducing impacts during LSL upgrades.

2. Materials and methods

Quality Assurance/Quality Control steps are shown in **SI. A1**.

2.1. Composition of the PET liner polymer

A commercial PET liner available in Canada was thermoformed and provided by a local product vendor. The PET liner composition (additives/polymer ratio, organic and inorganic additives identification) was assessed following the protocol detailed in **SI. A2**.

2.2. Laboratory and pilot aging procedures

2.2.1. Laboratory experiments

PET liner coupons (size ≈ 1*5 cm^2; thickness = 0.71±0.22 mm) (**SI. A3**) were aged following 45 different conditions, adapted from NSF/ANSI-61 certification (International/ANSI, 2016) with aging durations of 4, 8 and 12 weeks, temperatures of 20 °C, 20 °C with 2 freeze-thaw cycles (FTC) monthly and 40 °C. Simultaneously, coupons were exposed to five types of aging water: phosphate buffer at pH 6.5 or 8 (20 mM), with or without an initial dosage of 25 mg/L chlorine to simulate commissioning shock disinfection procedures, and municipal tap water (**SI. A4**). Each coupon was placed in a glass vial and completely immersed in a sufficient volume of water and kept in the dark. For aging at 40 °C, quadruplicate samples were placed in an incubator. Tubes undergoing FTC were repeatedly stored at 20 °C for 10 days, then frozen at -15 °C for 72 hours, and finally thawed gradually in a water bath at 80 °C for 5 hours. After aging, the coupons were extracted, rinsed with Type I water and stored in a desiccator.

2.2.2. Pilot aging and operating procedures

The experimental pilot involves two main steps. First, copper pipes (length ≈ 80 cm; internal diameter = ½-inch) lined with a PET liner (thickness = 0.26±0.06 mm) were aged in quadruplicate for a static aging procedure under 12 conditions. Two types of aging water were used: phosphate buffers at pH 8, with and without chlorine. The three remaining pipes were installed on a closed-circuit pilot system (**Fig. A1**) for dynamic aging. Pilot operation is detailed in **SI. A5**.

2.3. Water quality analysis

Chlorine concentration and pH were measured using Standard Method APHA 4500-Cl (G) and using a Fisher Scientific Accumet® pH meter (±0.01). Chlorine monitoring is displayed in **Table A1** (laboratory) and **Table A2** (pilot) and pH (±0.4) remained stable during experiments. Total metal concentrations (Al, Ti, Cr, Fe, Cu, Zn, Zr, Cd, Sb and Pb) were analyzed by inductively coupled plasma mass spectroscopy (ICP-MS), adapted from Method 200.8 of the United States Environmental Protection Agency. Analysis was conducted using a NexION 5000 instrument (Perkin Elmer, MA, USA). Samples were acidified to 1% vol/vol with Trace Metal Grade HNO_3. Metal contamination during acidification and collection was checked by analyzing Type I water following the same preparation as the samples.

2.4. PET liner analysis

2.4.1. Composition of the PET liner

TGA analysis revealed a polymer and additives content of 89.05 ±0.72% and 10.95±0.72%, respectively (**Fig. A2**). Additives identification is provided in **SI. A6**.

2.4.2. Coupon properties

Coupons mass variations were assessed at each aging stage. A Spectrum Fourier Transform Infrared (FTIR) spectrometer Spectrum™ 65 in attenuated total reflection (ATR) mode with a zirconium crystal was used (PerkinElmer, MA, USA) to characterize the surface chemistry of the PET liner coupons and to identify particles recovered during the pilot experiment (**SI. A7**).

Coupons crystallinity was evaluated with a differential scanning calorimeter DSC Q2000 (TA Instruments, DE, USA) to obtain the melting enthalpy (ΔH_f) (**SI. A8**). Crystallinity percentage (X%) was obtained using the method described by Sichina W.J. (2000) based on the melting enthalpy of 100% crystallized PET (ΔH_f^0) (140.1 J.g^{-1}) (Eq. 1).

$$X\% = \frac{\Delta H_f}{\Delta H_f^0} * 100 \qquad \text{(Eq. 1)}$$

Values were then corrected by the percentage of polymer in the PET determined by TGA (Rowenczyk et al., 2024).

2.4.3. PET fragmentation

At the end of the pilot experiment, a filtration system installed at the pilot drain (**Fig. A3**) filtered the circulated water and the equivalent of three times the pilot volume of fresh filtered water, recovering PET liner particles at 300 and 28 μm cut-off points. White/beige particles recovered were counted under a microscope Stemi 305 MAT (Zeiss, CA, USA) with 20- or 10x magnification and then chemically identified as PET using ATR-FTIR if matching PET spectrum.

2.5. LCA

2.5.1. Goal and scope

This LCA aimed to evaluate the potential environmental and human health impacts of three scenarios for LSL upgrades in a participating utility (QC, Canada): 1) PET liner installation, 2) replacement with PEX pipe and 3) replacement with copper pipe. It also sought to compare the health impacts from direct chemical leaching exposure during use. The LCA was conducted following the ISO 14040/14044 standards (International Organization for Standardization (ISO), 2006a; b). This study established cradle-to-grave boundaries, encompassing production, installation, use and end-of-life phases. The assessment was conducted on the following functional unit (FU): "Upgrading a LSL (internal diameter ½-inch) for 50 years in a participating water utility (QC, Canada) in 2024″. An average LSL of 2.8 m length, based on available field data, was considered and assumed to supply a two-person household with a water demand of 184.5 LCD (Hatam et al., 2023) and an average drinking water intake of 1.242 LCD, regardless of age (Phillips et al., 2021). Operational phases were considered equivalent in terms of energy and maintenance due to data limitations and professional judgment. While it is recognized that different wastewater pipe materials exhibit varying levels of hydraulic roughness (Alsadi, 2019), it can be assumed that the impact on pumping energy consumption would not be significant considering the length of piping involved. Additionally, based on information from the partner utility, no maintenance is expected to be performed over the lifespan of each service line. However, in the event of local copper pipe failure, damaged sections can be repaired, which may lead to additional impacts. Conversely, PET liners would require full replacement due to the lack of demonstrated repairability. Given the uncertainties surrounding these factors, a sensitivity analysis was conducted to assess the effect of the lifespan of the various alternatives under consideration. If two service line replacements are needed to meet the FU, because of a shorter lifetime, the assumption was made that de-installation was part of the new system's installation and did not require any extra energy or material.

2.5.2. Life cycle inventory

The life cycle inventory (LCI) involves the quantification of all pollutants released into the air, water and soil, as well as the natural resources utilized throughout the entire operational process of the three systems over their 50-year service life. Data for the LCI were sourced from the participating utility, published literature, internet resources and complemented for the background with ecoinvent database (version 3.8) for the three systems, the material production (PET liner, copper and PEX pipes), the installation by partial excavation (PET liner), complete excavation or torpedoing (PEX and copper pipes) (energy consumption, excavation, backfilling materials and material transport), and the end-of-life (transport and waste management). Specifically for the use phase, an exposure to leached metals at the tap by average (mean) and highly exposed consumers (high-90%) was determined and compared for PET liners, copper pipes, and a reference scenario of no LSL replacement. This excluded the PEX pipe scenario due to a lack of expected leaching data. Metal concentrations leached from copper and lead pipes and the PET liner were determined based on LSL pilot studies and the current laboratory study at 20 °C and assumed constant over 50 years. To validate these estimates, the quantity of metals leached from the PET liner over the 50-year period was estimated and represented 7% of the inorganic additives initially present in the liner. A linear relationship based on computer simulation was established to link the leached and ingested metals at the tap (**Table A3**). Detailed methodology is provided in **SI. A9**. OpenLCA 2.1 software was employed to model the product systems and specific information is provided in the **SI** as an Excel file.

2.5.3. Life cycle impact assessment

The regionalized LCA methodology IMPACT World+ (Bulle et al., 2019) was used for life cycle impact assessment (LCIA). Three midpoint and two endpoint indicators from IMPACT World+ Combined (v2.0.1) were considered, covering all potential impact pathways. Midpoint indicators focused on short-term climate change (CC) (100 years post-emission) to measure carbon footprint [kg CO_2eq], water scarcity (WS) [m^3 world$_{eq}$] and fossil and nuclear energy use (FNEU) [MJ deprived]. Endpoint indicators assessed human health (HH) in healthy life years lost [DALY] and ecosystem quality (EQ) in potentially disappeared fraction of species over a square meter during a year [PDF.m^2.an]. Details concerning the HH characterization impact factors and endpoint indicators are respectively provided in **SI. A10 and SI. A11**.

2.5.4. BLL model

IEUBK model (version 2.0 Build 1.66) (SRC, 2021) was used to estimate potential increases in blood lead levels (BLL) in children aged 0–84 months. The model considered exposures to lead in tap water, soil, dust, air and diet described by Deshommes et al. (2013) and used mean and high-90% lead values at the tap (**Table A3**).

2.6. Statistical analysis

The software package JASP version 0.18.0.0 was employed for statistical analysis. Non-parametric tests (Kruskal-Wallis) were conducted to assess the statistical significance ($p < 0.05$) of the conditions on the measured parameters.

3. Results and discussion

3.1. Impact of pH, temperature and chlorine on PET liner aging – laboratory scale

No mass variation or visual modifications were measured during aging under all conditions tested (**Fig. A4**).

3.1.1. PET surface oxidation

Oxidation Index (OI) as a function of the aging time plot for laboratory experiment is shown in Fig. 1. OI of unaged PET liner is not null, indicating the initial presence of ester and vinyl ester groups in the PET structure. Partial oxidation may have already occurred during high-temperature PET thermoforming (Panowicz et al., 2021), or additives with such chemical groups may be present in the liner. After 4 weeks of aging across most water types and all three thermal aging conditions, OI shows significant increases compared to unaged PET. Indeed, hydrolysis and thermal aging can break PET ester bonds into carboxyl (COOH) and hydroxyl (-OH) (Allen et al., 1991; Chamas et al., 2020; Edge et al., 1991), and oxidize polyethylene skeleton resulting in shorter polymer chains and CI increase (Panowicz et al., 2021). FTC and aging at 40 °C accelerate the oxidation rate: OI raises significantly between 4 and 8 weeks, then decelerates between 8 and 12 weeks, appearing to reach a plateau. Both high temperature and FTC may contribute to the formation of additional C=C bonds (Chamas et al., 2020; Edge et al., 1991; Rowenczyk et al., 2024) along with C=O bonds, resulting in the observed pronounced increase in OI. For chlorination at pH 8, OI values are systematically higher in chlorinated buffers as compared to unchlorinated ones, except under FTC at 8 weeks. In fact, chlorine addition can accelerate polyethylene oxidation (Castagnetti et al., 2011; Castillo Montes et al., 2012; Eng et al., 2011; Khan et al., 2022). There are no significant differences in oxidation rates among the various pH. This differs from findings by Castillo Montes et al. (2012) testing Cl_2 concentrations from 0 to 100 mg/L that suggests that pH could influence the depth of chlorine migration into the polymer, with deeper penetration occurring around pH 6.5, when $HOCl^-$ predominates.

3.1.2. Crystallinity modification

Crystallinity as a function of the aging time plot for laboratory experiments is displayed in **Fig. A5**. The crystallinity of aged PET liner coupons after laboratory aging remained consistent for all water types, thermal and temporal conditions in reference to unaged PET liner coupons with a crystallinity content of 12.5±1.8%. Aging is expected to increase the percentage of crystallinity with the polymer's amorphous content slow degradation (Allen et al., 1991; Castillo Montes et al., 2012; Hassinen, 2004; Panowicz et al., 2021). Thermal conditions tested remained well below PET's Tg, so polymer modifications primarily occurred on the surface layer, leaving the internal structure largely unaffected, especially for the short-term conditions tested. As a bulk measurement technique, DSC cannot detect small variations on the plastic surface. Notably, aged PET coupons show some degree of crystallinity variability (from 9.0% to 19.2%) depending on the conditions. It can be attributed to the thermoforming, leading to thickness variations of the installed liner, measured at 0.71±0.22 mm across 80 coupons cut from the same liner. Hence, different cooling rates occur depending on the local thickness and cooling conditions, yielding uneven thermal stresses and variations in crystallinity (Erchiqui et al., 2021).

Although no significant changes in crystallinity were observed, the previous oxidation results suggest that more pronounced degradation could occur over the long term. Ultimately, substantial changes in crystallinity would lead to mechanical weakening of the liner (Vertova et al., 2019; Whelton and Dietrich, 2009), resulting in liner rupture or failure and compromising its protective function against lead and its actual service life.

3.1.3. Metal leaching

PET liner coupons have shown significant leaching of titanium, zinc and lead (Fig. 2 and **Table A4**). All are presumed to be metallic additives added into the PET liner based on the plastic deformulation results (**SI. A6**), although titanium was not detected by MEB-EDX, likely due to

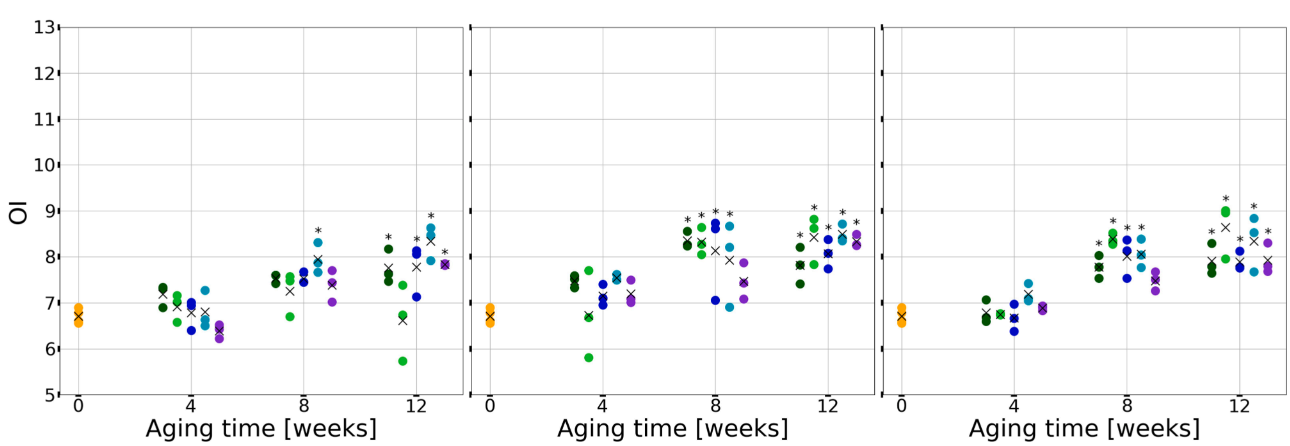

Fig. 1. Aging time against OI for laboratory experiments conducted at 20 °C (left), 20 °C and 2 FTC per month (center) and 40 °C (right). Legend: Triplicates of unaged (orange), and static aged coupons in buffer solution at pH 6.5 unchlorinated (dark green) and chlorinated (light green) and pH 8 unchlorinated (dark blue) and chlorinated (light blue) and tap water (violet). Significance relative to the unaged PET liner (*) and mean on triplicates (x).

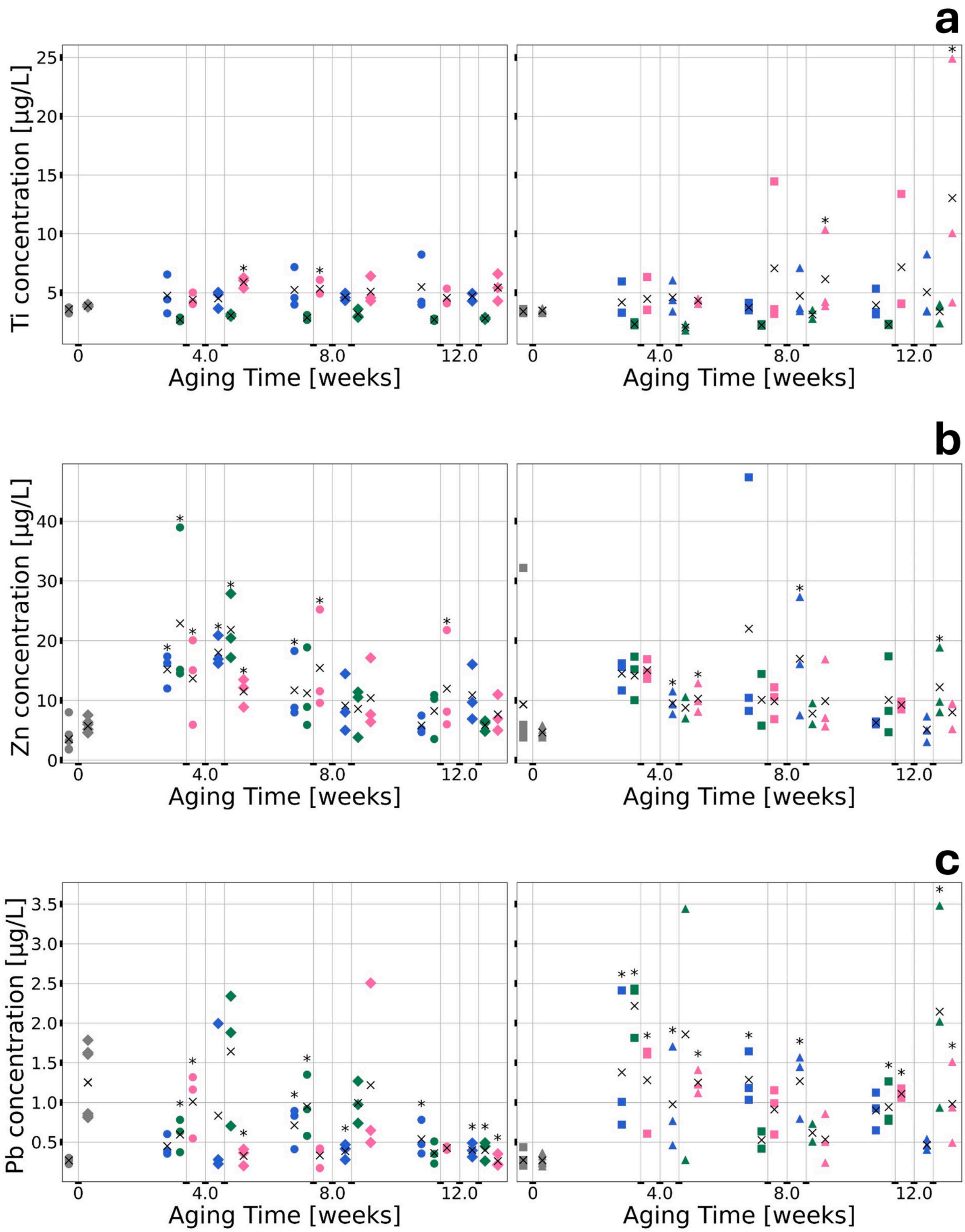

Fig. 2. Aging time against titanium (**a**), zinc (**b**) and lead (**c**) concentrations in the aging water after the laboratory experiment. Legend: Triplicate values for buffer solutions at pH 6.5 (left panel) unchlorinated (dot) and chlorinated (diamond), and for buffer solutions at pH 8 (right panel) unchlorinated (square) and chlorinated (triangle). Data provided for pre-aging (grey) and post-aging at 20 °C (blue), 20 °C and 2 FTC per month (green) and 40 °C (pink). Significative results to the blank (*) and replicate mean values (x).

particle sizes being below the equipment's 1 µm spatial resolution. These metals exhibit distinct leaching trends that are difficult to compare with each other or with other studies due to their varying behaviors in response to water quality, aging and operating conditions, as well as the wide variability of material formulations and potential aging states (Chapa-Martinez et al., 2016; Gonzalez et al., 2013).

Titanium is released in significant quantities at 40 °C, especially in chlorinated buffers reaching 9.57 µgTi/L in chlorinated buffer pH 8 after 12 weeks. An increasing leaching trend with exposure time is observed both at 20 °C and 40 °C, which is consistent with the literature for antimony leaching (Al-Malack, 2001; Breault, 2014). However, no titanium leaching is observed with FTC regardless of the aging time. Titanium is either rapidly released and then precipitates within the first weeks due to static aging conditions and the extended measurement period (every 1 month), or it may be resorbed to the PET, considering the FTC in this study involved thawing for 5h at high temperature (80 °C) after freezing (Chapa-Martinez et al., 2016). Indeed, higher temperatures generally accelerate metal leaching, even over such short thermal aging periods (Chapa-Martinez et al., 2016; Westerhoff et al., 2008).

Increasing zinc leaching occurs in all buffers for all thermal conditions tested after 4, 8 or 12 weeks of aging and is significant for all buffers pH 6.5 and in chlorinated buffer pH 8. Similarly, significant lead leaching is observed for almost all buffers tested, i.e. unchlorinated buffer pH 6.5 and pH 8 and chlorinated buffer pH 8 under all thermal conditions. For both lead and zinc, significant leaching occurs after 4 to 8 weeks depending on water and thermal conditions, followed by a decrease after 8 to 12 weeks. These results contrast with our findings for titanium and with previous literature, which generally indicates that longer exposure times typically increase metal leaching (Al-Malack, 2001; Breault, 2014; Westerhoff et al., 2008). Again, this may be attributed to the slow precipitation or resorption of metals on PET during the month-long stagnation periods between measurements, unlike other studies that employed shorter measurement intervals (hours to days), with static or dynamic conditions (Al-Malack, 2001; Breault, 2014). Highest zinc and lead release occurs under FTC conditions: zinc at pH 6.5 and lead at pH 8. Zinc leaching reaches 19.37 µg/L without chlorine and 16.09 µg/L with chlorine after 4 weeks, while lead release reaches 1.95 µgPb/L without chlorine after 4 weeks and 1.87 µg/L with chlorine after 12 weeks. Westerhoff et al. (2008) showed that freezing does not impact antimony leaching compared to 22 °C, but the heating phase (80 °C) carried out in this study during FTC can significantly increase Pb and Zn leaching (Chapa-Martinez et al., 2016; Westerhoff et al., 2008). However, similarly to Al-Malack (2001), we did not observe a significant Pb increase between 20 °C and 40 °C likely due to the low temperatures tested. At 20 °C, pH does not significantly impact Ti, Pb and Zn leaching, contrary to findings from Al-Malack (2001), who reported that acidic conditions increase lead leaching. However, our results align with Westerhoff et al. (2008), which found that municipal tap water pH does not affect antimony leaching. No antimony leaching was detected, despite Breault (2014) reporting Sb presence in PET liners and leaching up to 0.33±0.17 µg/L after 4 days at room temperature, likely due to ICP-MS detection limits (< 0.47 µg/L) and the 20-fold required dilution for buffers.

These metals released could pose a significant risk for human health if ingested, which will be further evaluated through LCA and IEUBK analyses. Moreover, additives like plasticizers, flame retardants, antioxidants, acid scavengers, stabilizers and pigments are in general present to provide PET with thermal, chemical and mechanical protection, enhancing performance and extending service life. Their depletion over time can therefore increase plastics vulnerability to operating conditions and affect directly the long-term durability and service life of the PET liner (Castillo Montes et al., 2012; Hahladakis et al., 2018; Hassinen, 2004). As the PET liner ages, the risk of lead diffusion from the supporting LSL through the liner's wall may increase, potentially elevating lead leaching risks (Randtke et al., 2017). Finally, it is also worth noting that on a ½-inch pipe scale, the higher polymer surface area to water volume ratio (x19) suggests that metal concentrations resulting from leaching into drinking water could be much greater (Keresztes et al., 2013).

3.2. Impact of pH, temperature and chlorine on PET liner aging – pilot scale

3.2.1. Plastic surface oxidation

OI for pilot aging results (Fig. 3) follows a similar increasing trend as for laboratory results, although not always statistically significant. However, no chlorine effect or dynamic aging effect can be observed. Short-term testing (13 weeks) cannot assess long-term effects of real service line conditions affecting liner degradation, such as varied flow rates and pressure, water quality, biofilm formation and high chlorine concentrations (Dear and Mason, 2001; Fadel, 2022). As compared to static laboratory testing, greater variability is observed between samples aged under similar conditions. This reflects the more dynamic operations of the flowing pilot as well as the fact that pilot liners were independently thermoformed on different ½-inch pipes. This certainly led to variability in material homogeneity and crystallinity, resulting in partial oxidation and slightly different surface modifications even for unaged samples (Panowicz et al., 2021).

3.2.2. PET liner fragmentation

No plastic fragmentation (> 28 µm) was detected under pilot conditions, most likely due to the short durations tested. PET surface oxidation after pilot aging was minimal, resulting in fewer oxygen-containing functional groups that are an important factor influencing microplastic release, particularly in small particles (Zhang et al., 2022). Only one fragment attributable to the liner was recovered after dynamic aging of pipes aged at 20 °C in chlorinated buffer at pH 8 and matched at 85% with PET. Also, when fragmentation occurs, smaller particles (< 30 µm) are more prevalent, but these could not be recovered in our pilot set-up (Zhang et al., 2022).

3.3. LCA results

3.3.1. Production, installation and end of life phases

LCA results of the production, installation and end of life phases for

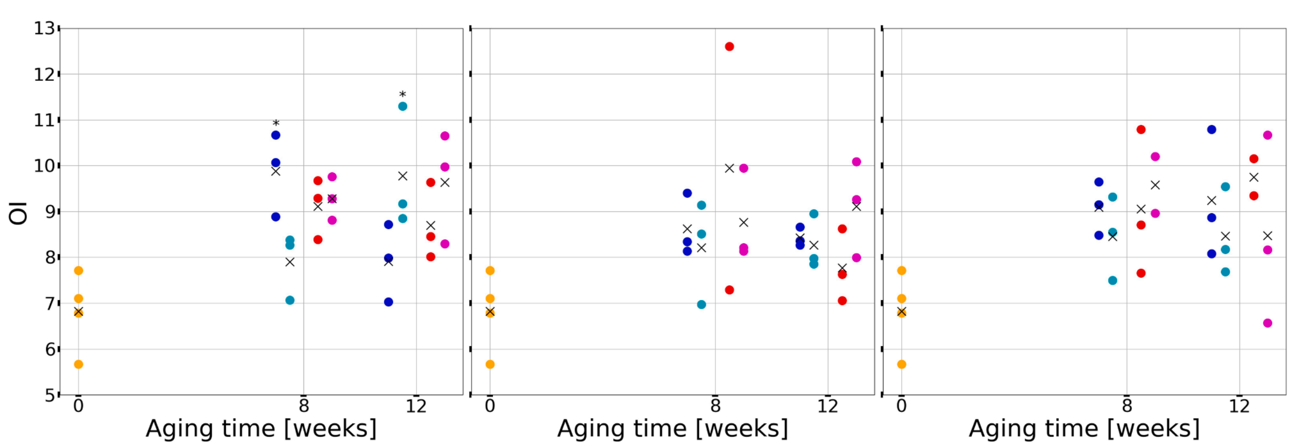

Fig. 3. Aging time against OI for pilot experiment conducted under static aging at 20 °C (left), 20 °C and 2 FTC per month (center) and 40 °C (right) and then under dynamic aging. Legend: unaged (orange), and aged PET liner in buffer solution at pH 8 unchlorinated and chlorinated after static aging (dark and light blue) and after dynamic aging (dark and light red). Significative results to the unaged PET liner (*) and replicate mean values (x).

EQ, HH, CC, WS and FNEU are displayed in Fig. 4.

Results show that the production of PET liners and PEX pipes has a negligible impact (< 0.1%) across all impact categories and areas of protection. However, copper production phase contributes by up to 24.3% (excavation) and 40.3% (torpedoing) for HH and to 10.3% (excavation) and 19.6% (torpedoing) to EQ impacts associated with copper pipes, due to copper processing. Indeed, literature shows that copper pipe production impacts far exceed those of plastic pipes (Asadi et al., 2016; Hajibabaei et al., 2018).

For the three technologies studied, the installation phase is the main contributor to impact scores across all indicators. Total or partial excavation processes contribute the most to the installation phase, with over 85% attributed to gravel production and transport, as well as excavated soil transport. Moreover, full excavation for PEX and copper pipe replacement proves to be more impactful due to longer excavation lengths needed, leading to higher mass of gravel and excavated soil, and increasing CO_2eq emissions by over 110% compared to torpedoing. This observation aligns with findings by Alsadi (2019) and Hajibabaei et al. (2018), highlighting that trenchless installation methods (pulling, torpedoing, resin coating) have lower environmental impacts compared to full excavation. Alsadi (2019) underscored that coatings generate fewer CO_2 emissions than pulling, torpedoing and full excavation due to reduced equipment, materials, excavation dimensions, and shorter installation times. In this study, the copper pipe has a lower CC impact, owing to equivalent excavation dimensions considered for liner rehabilitation and replacement by torpedoing. Conversely, the PEX pipe has the greatest impact due to the required replacements (x2) to meet the FU. The end-of-life phase for all technologies is negligible, contributing less than 0.01% to total impacts across all categories.

Finally, global results (excluding the use phase) showed that copper pipes installed via torpedoing stand out for CC, WS and FNEU for a LSL upgrade in Quebec in 2024. PET liners scored lower in HH and EQ due to the less impacting production phase compared to copper. However, if complete excavation is necessary, PET liners emerge as the less impactful choice across all categories and areas of protection.

3.3.2. Use phase

The HH impacts (carcinogenic and non-carcinogenic) depending on the LSL length and associated to the use phase are presented in Fig. 5.

Health impacts of PET liners evaluated by the LCA are mainly due to zinc (86.4%) and lead (13.6%) for non-carcinogenic effects, and exclusively to lead for carcinogenic effects. Indeed, the United States Environmental Protection Agency report no health effects associated with titanium ingestion (United States Environmental Protection Agency (USEPA), 2005a) or carcinogenic effects for zinc (United States Environmental Protection Agency (USEPA), 2005b). However, in 2022, the European Commission banned the use of titanium dioxide as a food additive (European Commission, 2022). Recent studies on titanium even suggest that early exposure during pregnancy might cause language development delays (Jiang et al., 2023). Titanium could also accumulate in key organs (kidney, brain, reproductive organs), requiring further research to understand its toxicity and health risks (Li and Tang, 2024). For zinc, the associated noncarcinogenic DALY were unexpected, given that the concentrations calculated in our study were significantly below drinking water standards (< 5 mg/L, aesthetic) (**Table A3**) (Health Canada, 1987). HH impacts related to lead are clearly the most significant and vary significantly between average and highly exposed consumers (high-90%). For 1, 2.8 and 19 m service lines, highly exposed consumers health hazards are 5.3, 3.4 and 2.4 times greater than average consumers exposed to mean Pb concentrations. The DALY of an unreplaced LSL of 2.8 m is 14 times higher than that associated with a PET liner rehabilitation and 80 times higher than that with a copper pipe replacement. As for zinc, noncarcinogenic impacts of the copper pipe exceed 10^{-6} DALY although copper concentrations considered to assess the impact of copper exposure used in the analysis were well below the current drinking water standards (**Table A3**) (Health Canada, 2019b). Our results show similar trends than reported by Asadi et al. (2016) comparing PEX and copper piping for buildings for all LCA stages (HH impacts associated with water quality not considered). They report that use of PEX could also significantly reduce carcinogenic and non-carcinogenic health impacts by 99% and 42%, respectively, reflecting the contribution of the production phase.

3.3.3. LCA sensitivity analyses

Impact on LCA results of two key variables parameters were assessed: the lifespans of the plastic-based alternatives (PET liner and PEX pipe) (**Fig. A6**) and the LSL lengths (1, 2.8 and 19 m) (**Fig. A7**).

Considering a 50-year lifespan, PEX pipes are more advantageous over copper pipes across all impact indicators, mainly due to the less impactful production phase. For equal lifespan and if a complete excavation is necessary, the PET liner is a more advantageous solution than the PEX pipe regarding all indicators. Prior reports have stated that trenchless installation methods, such as torpedoing and liners, offer environmental benefits over traditional trenching, as they use less equipment, require shorter installation times, and involve minimal excavation (Alsadi, 2019). However, if torpedoing can be used, replacement with a PEX pipe is less impactful than a PET liner. While fixed-trench methods (PET liner or torpedoing) show minimal impact variation with LSL length, impacts associated with complete excavation may vary widely due to increased material extraction and longer excavation dimensions. Depending on the actual lifespan of the PEX pipe and the PET liner, the choice of the least impactful replacement option will vary.

However, in the case of earlier liner failure, full replacement is likely to be required as there is no demonstrated ability to replace a liner, incurring significant additional replacement impacts and additional costs not modeled in this study. The lifespan of the PET liner is also closely linked to that of the LSL supporting it, and the failure of the latter, which are already over 50 years old, would similarly necessitate a complete replacement. To investigate a PET liner lifespan of 25 years, we considered the impacts of adding a copper pipe replacement for the remaining 25 years. In all cases, adding a full replacement significantly reduces any environmental benefits associated with the use of PET liners. In that case, if installation by torpedoing is possible, then replacing with a copper pipe instead of a PET liner significantly improves the impacts associated with HH and CC by 16.0% and 58.8% respectively. On the other hand, when replacement requires complete excavation, copper pipes become preferable compared to PET liners with a 54.4% improvement for CC, but PET liners reduce the HH impacts by 11.4% compared to copper pipes.

LCA lifespan estimates for some plastic technologies range from 25 to 100 years (Alsadi, 2019; Asadi et al., 2016). However, lifespans field data for plastic piping and PET liners is lacking for informed upgrading decision-making.

Our LCA results are coherent with the few published LCA analysis comparing various plastic to copper pipes (Alsadi, 2019; Asadi et al., 2016; Hajibabaei et al., 2018), despite each LCA being tied to specific contexts, boundaries, indicators, methods and hypotheses stated. However, this LCA used more recent and comprehensive databases and methodology, including chemical leaching during use, which is essential for assessing human health impacts but often overlooked in other LCA (Phillips et al., 2021). For instance, Asadi et al. (2016) lack transparency in foreground inventory modeling, while Hajibabaei et al. (2018) focus on environmental impacts but have a limited scope, omitting use and disposal phases, and relying on outdated impact assessment methods (CML 2 baseline 2000 and EI 99). Additionally, Alsadi (2019) assesses only the carbon footprint of pipeline materials using ISO 14064, neglecting other impact categories like toxicity and ecotoxicity. Regardless, our LCA results should be interpreted considering limitations relative to the quality of primary and secondary data across installation, production and end-of-life phases. Commercially available LCA databases may not adequately reflect regional specifics. To enhance

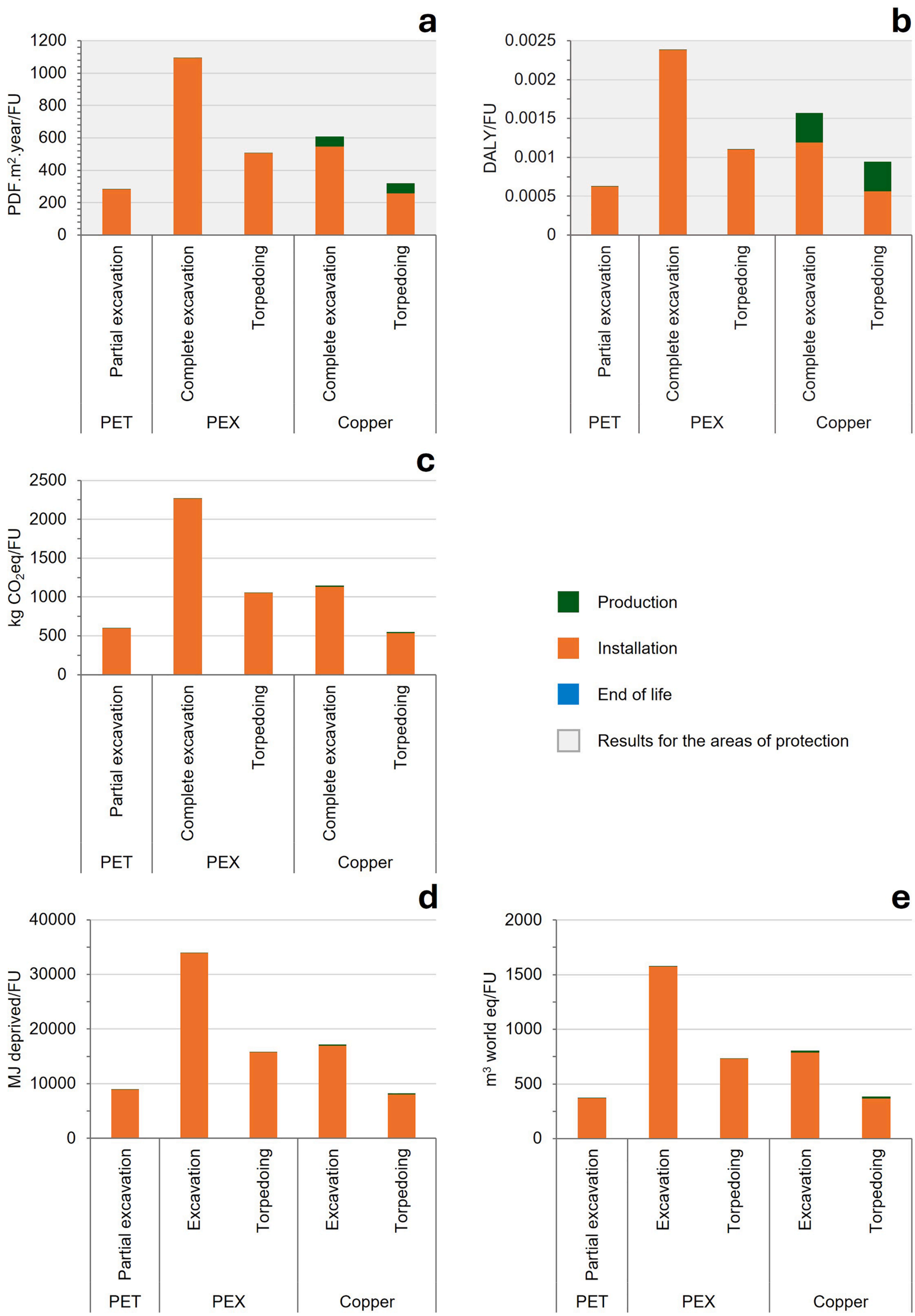

Fig. 4. Potential environmental impacts: EQ [PDF.m^2.an/FU] (**a**), HH [DALY/FU] (**b**), CC short-term [kg CO_2eq/FU] (**c**), FNEU [MJ deprived/FU] (**d**) and WS [m^3 world$_{eq}$/FU] (**e**). Legend: Contribution of production (green), installation (orange) and end of life (blue) phases for the three LSL upgrade scenarios.

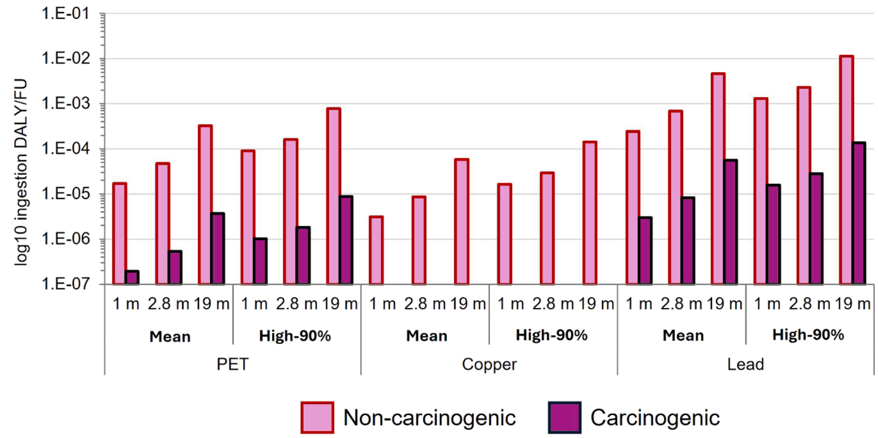

Fig. 5. HH impacts of metal leaching from PET liners, copper pipes and unreplaced LSL. Legend: Non-carcinogenic (light pink) and carcinogenic (dark pink) DALY for average (mean) and highly (high-90%) exposed consumers.

the impact assessment, additional excavation data (pipes, liners or gravel losses, additional material requirements, transportation) would be beneficial, given their significant influence compared to other phases. Results could be improved with more precise data on copper pipe production and recycling content considered (100% in this study), end-of-life management, chemical exposure, and conditions (flow dynamics, freezing and ground movements) leading to liner and pipe failures. Most importantly, the sensitivity study shows uncertainties surrounding the lifespan of plastic technologies directly affect study conclusions. Additionally, as mentioned by Randtke et al. (2017), the use of PET liners should consider the long-term effects of exposure to chlorine disinfectants, the leaching of antimony from aged PET and the long-term likelihood of copper and lead diffusion through the liner as it ages.

Some uncertainties in modeling concern the health impact of exposure to metals during the use phase. We assumed constant metal leaching from the pipes over 50 years derived from one reference water quality, which may not be extended to predict the long-term lead release, nor account for the impact of water quality or the use of corrosion inhibitors. Furthermore, LCA HH endpoints are set in USEtox characterization factors for carcinogenic and non-carcinogenic human health that are respectively derived from dated studies on rodents (Gold et al., 2011) and WHO data on fetal mental development (Fantke et al., 2017), while more recent studies could be considered. They also consider the overall environmental burden of lead exposure and are not specific to drinking water or age-related effects, precluding a focused assessment of the impacts on children, most vulnerable to lead exposure. Most strikingly, exposure to zinc and copper during the use phase, which remains well below international drinking water standards (> 5 mg/L for zinc as an aesthetic guideline and 0.3–2 mg/L for copper) and recommended daily intakes, led to significant DALY estimates by the LCA model. However, these estimates may not be fully supported by epidemiological or toxicology assessments (Health Canada, 1987; 2019b). These discrepancies reflect the differences in the endpoints considered in the LCA and drinking water ingestion biokinetic models. They also support the use of biokinetic models that account for populational vulnerabilities that serve as the basis for protective drinking water standards. Additionally, the omission of potential emissions like secondary MNP and additives leaching from PEX affects the accuracy of toxicity assessments.

These constraints underscore the need for more precise field data and continued improvements and methodological adjustments in LCA of LSL replacement technologies.

3.4. Impacts of LSL upgrade scenarios on children BLL

Drinking water standards are set to protect the most vulnerable populations and are therefore based on the setting levels to limit the impacts of exposure on the BLL of infants and young children. USEtox characterization factors (CF) are not aged-specific and are not directly relevant to manage lead exposure through drinking water. Fig. 6 shows IEUBK simulation results for geometric mean BLL and the proportion of children aged 0 to 84 months with BLL > 5 μg/dL. BLL rise from 5.6% (mean) and 29.9% (high-90%) to 98.1% (mean) and 207.8% (high-90%) for a respectively 1 m to 19 m unreplaced LSL, as compared to a rehabilitation with a PET liner. The limited lead leaching considered for the PET liner did not increase BLL, except for the 19 m LSL (< 3.7%-18.2%). All values stay below the 3.5 μg/dL health advisory for elevated BLL for young children (Ruckart et al., 2021), except for highly exposed consumers (high-90%) with a 19 m unreplaced LSL. However, the risk of exceeding the threshold value of 5 μg/dL is notably higher without a replacement (38.6%) than with PET liners rehabilitation (0.4%) even for a 19 m LSL (high-90%), while replacement with a copper pipe would not impact BLL.

As expected, longer LSL increase lead exposure severity, emphasizing the need for prompt replacement to mitigate health risks. Replacing a LSL with a copper or a PEX pipe or installing a PET liner will drastically reduce the risk of elevated BLL. Although these results show that PET liners significantly reduce the risk of exposure to lead, they also highlight the paradox of using a liner leaching small concentrations of lead, especially in light of the clear consensus that there is no safe lead concentration, especially for young children (Health Canada, 2013).

4. Conclusion

This study provides actionable evidence to assist utilities in their LSL management strategies by addressing key questions regarding the sustainability and risks of PET liners. Specifically, their short-term physicochemical durability was evaluated, and their sustainability and health benefits compared to complete replacement options with copper and PEX pipes against a non-replacement scenario without corrosion control.

Key findings include the observation of significant leaching of health-relevant metals (Pb, Ti and Zn) from the PET liner. Furthermore, rapid surface oxidation of the PET liner indicates a potential for more severe degradation over its projected service life and under real operation conditions possibly affecting its integrity and effectiveness as barriers against lead. No fragmentation of the liners was observed during a short-term pilot study. Investigations of field aged PET liners installed in European distribution systems could significantly aid in assessing their

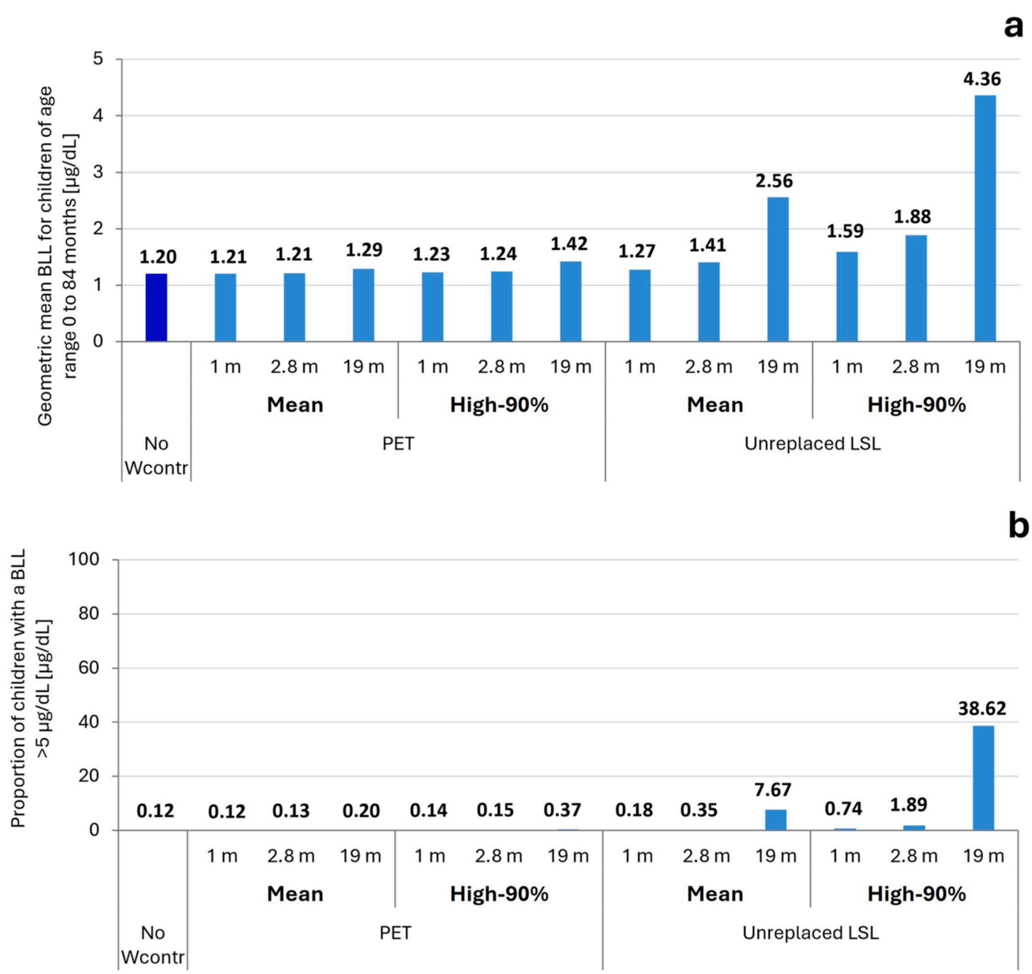

Fig. 6. Geometric mean BLL for children of age range 0 to 84 months using average (mean) and 90th percentile of lead concentration at kitchen tap (high-90%) during usage (**a**), and the proportion of children with a BLL > 5 µg/dL using average (mean) and 90th percentile (high-90%) (**b**).

long-term durability especially the frequency liner failures and concerns over leaching and migration of metals.

LCA results clearly point to the installation being the critical factor for all scenarios. Results indicate that a LSL upgrade using a copper pipe via torpedoing has lower impacts on CC, WS and FNEU, while PET liners minimize HH and EQ impacts. However, PET liners score better in all categories and areas of protection and total excavation is required, if a 50-year lifespan is confirmed. However, even if plastic pipes appear to bring substantial benefits, concerns persist regarding additives leaching, lifespan in field conditions and MNP fragmentation. These are often overlooked in current LCA studies (Phillips et al., 2021) despite being essential for assessing long-term potential health risks.

LCA and IEUBK modeling converge to show that copper pipes effectively eliminate lead and related health risks, especially for children. PET liners reduce carcinogenic and non-carcinogenic, albeit with remaining concerns regarding lead, zinc and titanium leaching. Health hazards associated with titanium may have been overlooked in this study due to the lack of regulations in drinking water and limited understanding of its risks, highlighting the need for further investigation.

Ultimately, the optimal upgrade solution should consider water quality, field constraints, LSL characteristics, and the remaining uncertainties regarding effective lifespan and potential metal leaching risks. Most importantly, additional improvement in health outcomes of the LCA model would be desirable and relative costs and feasibility of these technologies should be evaluated.

CRediT authorship contribution statement

Amélie Surmont: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Laura Rowenczyk:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Ivan Viveros Santos:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. **Fatemeh Hatam:** Writing – review & editing, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Anne-Marie Boulay:** Writing – review & editing, Supervision. **Michèle Prévost:** Writing – review & editing, Validation, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary materials associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.122686.

Data availability

I have shared my inventory file for LCA modeling as an additional document. Additional data obtained and used during this study will be provided upon request.

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