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

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Supporting Information

SI. A1 Quality Assurance/Quality Control

Precautions were taken to avoid cross-contamination with plastic additives and other plastic-related chemicals in pilot and laboratory equipment and consumables. Pure cotton lab coats were used to avoid potential contamination associated with polyester (PET) or other plastic fiber lab coats. Nitrile gloves were always used to manipulate the samples. All plastic laboratory containers were avoided wherever possible, and only glassware (bottles, vials, tubes) was used during experiments. When not protected from external contamination with caps or aluminum foil, clean glassware, bottles containing aging waters and rinsed PET liner coupons were always handled under a vertical laminar flow hood. Similarly, the preparation of vials and tubes for the aging experiment and sampling for analysis, were always carried out under a vertical laminar flow hood. Disposable graduated pipettes and plastic pipette tips used for sampling were taken directly from their plastic bags, visually inspected, and used only once, to avoid using plastic materials for long periods. Atmospheric blanks for laboratory experiments were performed to attest to the controllability of external contamination as well as organic and metallic leaching of glassware. Also, pilots were fully built in copper and brass. For sealing the copper pipes, Teflon tape was avoided in favor of plumbing flax. For waterproofing the pilot yellow and pink Teflon tape was used as it is easy not to mistaken with PET in terms of color and FTIR-ATR spectra. Before every use, the homemade filtration system was sonicated to remove any attached particles. During the laboratory experiments, controls made of Type I water only followed the same aging process and were analyzed like the sample to evaluate the contamination occurring during the manipulations.

SI. A2 Protocol to assess the composition of the PET liner studied

The additive/polymer ratio in the PET liner was evaluated using a thermogravimetric analyzer TGA Q500 (TA Instruments, DE, USA). 2–5 mg of a sample were placed in a platinum pan (100 μ L) and burned at a temperature ramp of 20 $^{\circ}$ C/min, first under a nitrogen flow up to 600 $^{\circ}$ C and then under an oxygen flow up to 800 $^{\circ}$ C to pyrolyze the residue. This procedure was repeated on three samples. The mass loss curves were normalized and analyzed using TRIOS Software (version 5.0.0.44608, TA Instruments). Inorganic additives formulated in the PET liner were identified after TGA analysis without pyrolysis of the residue. This was analyzed in triplicates by a JSM-7600F MEB-EDX (JEOL Ltd, Tokyo, Japon) coupled to an energy-dispersive X-ray spectrometer (Tmax, Oxford Instruments, UK). Spot analyses and mapping were carried out to identify the relative abundance of elements. The composition of the organic fraction of the PET liner was conducted by SGS Canada Inc. (Mississauga, Ontario, Canada) using liquid and gas chromatography coupled with mass spectrometry and proton nuclear magnetic resonance spectroscopy.

SI. A3 Coupons preparation

The PET liner followed the same preparation as that applied for NSF61 certification. The liner was first thermoformed onto a PEX pipe (Internal diameter = 2-inch; length = 60 cm), rinsed and then extracted from the pipe. The liner was then cut into coupons measuring approximately 1cm*5cm along the length of the pipe. Coupons were identified, weighed, photographed and their dimensions precisely measured. The surface area of the coupons was determined automatically from photos of the coupons analyzed with ImageJ software. The average thickness of the coupons was determined on 80 randomly selected coupons with a caliper. Coupons were then rinsed with Type I water on both sides for 5 seconds and dried in a vertical laminar flow hood.

SI. A4 Aging waters preparation protocols

The laboratory and pilot buffer solutions at pH = 6.5 or 8 were prepared from two 500 mL stock buffer solutions with a ionic strength of 200 mM obtained by successively adding 2.8248 g or 4.6504 g of anhydrous Na_2HPO_4 to 200 mL of Type I water, then exactly 5.4247 g or 0.3048 g of anhydrous KH_2PO_4 , under agitation. The volume was then adjusted to 500 mL in a volumetric flask with Type I water. The stock solutions were then diluted 10-fold with Type I water to obtain the targeted ionic strength of 20 mM, chosen to mimic tap water. The pH of the resulting solutions was monitored and adjusted as necessary with HCl or NaOH (0.1/1 M). All

laboratory and pilot aging waters were autoclaved at 120 °C for 20 min to eliminate any risk of bacterial growth. Chlorine was added from a NaOCl stock solution to buffers to achieve a concentration of 25 mgCl₂/L.

SI. A5 Pilot operation conditions

Before the test phase, the pilot was purged with tap water for 15 minutes and then, for 15 minutes with filtered tap water on 28 µm through a home-designed filtration system (**Fig. A3**). The pilots operated in closed-circuit and filtered tap water circulated for 5 days at 7.6 L/min, maintaining a pressure of 40-30 psi and a temperature of 20±5 °C using a glycol cooling system. Between conditions, the pilot underwent a 1-hour rinse with filtered tap water.

SI. A6 PET liners additives identification

Chemical analysis revealed no isophthalic acid inclusion or presence of semi-volatile additives. The sole presence of a structural isomer or proanthocyanidin A1 was detected (results not shown). C and O were abundant at all analyzed sites. Na, Zn, S, Cu, Pb and Ca were present over the entire surface, with a few sites showing higher concentrations. All are additives commonly formulated in plastics and are present in relatively low concentrations compared to other commercial plastics, making it a relatively clean plastic (INRS, 2019) [1].

SI. A7 FTIR analysis

For each sample, spectra were acquired in the 4000–600 cm⁻¹ region using 16 scans and a spectral resolution of 4 cm⁻¹. For each aging condition, three replicate coupons were analyzed once in the middle of its internal layer. All spectra were pre-processed with a rubberband correction baseline using Orange DATA MINING software (version 3.35.0) and its Spectroscopy add-on (version 0.6.12). To attest chemical modifications, an oxidation index (OI) representing a combination of carbonyl (CI) and unsaturated bond (UBI) index was calculated and defined as the area under the peaks between [1535;1785] cm⁻¹ representing the C=O and C=C bonds, on the area of a reference peak [2795;3150] cm⁻¹ representing a methylene peak (Fernandez-Gonzalez et al., 2021) [2].

SI. A8 DSC analysis

Each coupon was analyzed once with 3-6 mg of the sample placed in an aluminum hermetic pan (DSC Consumables, MN, USA). Under a nitrogen flow (50 mL/min), the samples were first maintained at 20 °C for 5 min, then heated to 300 °C at a rate of 10 °C/min. This final temperature was kept constant for 5 min before being reduced to 20 °C at the same rate. This

first cycle provides the crystallinity history and depicts the weathering of the polymer (Ter Halle et al., 2017) [3]. Melting peaks were integrated between 187.5 °C and 287.5 °C using TRIOS Software (version 5.0.0.44608, TA Instruments) to obtain the melting enthalpy (ΔH_f).

SI. A9 Determination of leached and ingested metal concentrations

The ingested metal concentrations considered in calculating impacts on human health were determined for the PET liner and copper pipe and compared to a reference scenario of an unreplaced LSL. Lead concentration in the LSL, and therefore at the tap, vary with water stagnation and consumption habits. Therefore, a linear relationship (**Eq. A1**) between the maximum dissolved lead concentration in the LSL (C_{LSL}) and the average (mean) or 90th percentile (high-90%) of lead concentration at the kitchen tap during one week of usage (C_{ING}) was established employing hydraulic and water quality modeling. For this purpose, a simple domestic water supply system was selected, utilizing the integration of EPANET (Rossman, 1994) [4] with the stochastic water demand model SIMDEUM®, as described in Hatam et al. (2023) [5]. This combined model considers house plumbing, water quality of the partner utility, consumption habits, and lead dissolution kinetics.

$$C_{ING} = F * C_{LSL} \quad (\text{Eq. A1})$$

Mean and high-90% were determined for each LSL length: with mean representing average consumers, and high-90%, highly exposed consumers (90th percentile). They were based on modeling 2.8 m and 14 m LSL (diameter 25.6 mm) with respective maximum lead concentrations of 84.21 µg/L and 94.20 µg/L in the LSL and a daily water demand of 184.5 LCD. The same factors, mean and high-90%, were considered for all metal studied (copper, titanium and zinc). Copper concentration leached from the copper pipe was set at 1000 µg/L (Doré et al., 2019) [6]. Titanium, zinc, and lead leached concentrations from the PET liner were extracted from the leaching data at 20 °C of this current study normalized by PET coupon surface area and experimental water volume (0.03 L). Maximum concentrations obtained with each water type over three months were averaged to represent municipal tap water. This concentration, assumed to occur after one night's stagnation, is multiplied by the liner's internal surface area and water volume ($S/V = 3149.96 \text{ cm}^2/\text{L}$), independent of LSL length. We suppose that the metal concentration calculated in the LSL remains constant over 50 years. For all scenarios considered, mean and high-90% were then used to determine the metal concentration at the tap from the concentration in the service line for the three lengths considered. Metal

quantity ingested over the 50 years of use corresponding to the metal exposure (E_c [kg_{intake}]) was quantified as follows in **Eq. A2**:

$$E_c = C_{ING} * V_{ingested} * 365 [days] * 50 [years] \quad (\text{Eq. A2})$$

With : $V_{ingested} = 1.242 \text{ LCD}$ (Phillips et al., 2021) [7].

SI. A10 Human health characterization factors determination

Toxicity impacts arising from metal leaching during the use phase were calculated based on the exposure to chemicals (E_c [kg_{intake}]) and characterization factors to quantify direct exposure to those chemicals (CF). CF to assess human toxicity account for fate, exposure and effect mechanisms. In the case of the use phase, EF [cases/kg_{intake}] align with a direct exposure of chemicals [kg_{intake}]. Hence, in this study, EF is equal to CF for assessing the impacts of chemicals present in drinking water. The USEtox model, a consensus multimedia model by UNEP/SETAC designed for characterizing the (eco)toxicological impacts of chemicals in LCA (Fantke et al., 2017) [8], was employed to derive EF for the direct exposure to chemicals. Furthermore, the severity factors from USEtox were used to convert cancer [11.5 DALY/case] and non-cancer cases [2.7 DALY/non-cancer cases] to endpoint level. **Table A5** contains the EF that were derived from USEtox.

SI. A11 LCA endpoint indicators

HH covered cancer and non-cancer human toxicity, particulate matter formation, photochemical oxidant formation, ozone depletion, ionizing radiation and water availability. EQ included freshwater ecotoxicity, freshwater acidification, marine acidification, terrestrial acidification, freshwater eutrophication, marine eutrophication, ionizing radiation, land transformation, land occupation and water availability (Bulle et al., 2019) [9]. Contributions from CC and WS were excluded from HH and EQ to avoid double counting.

Supporting figures

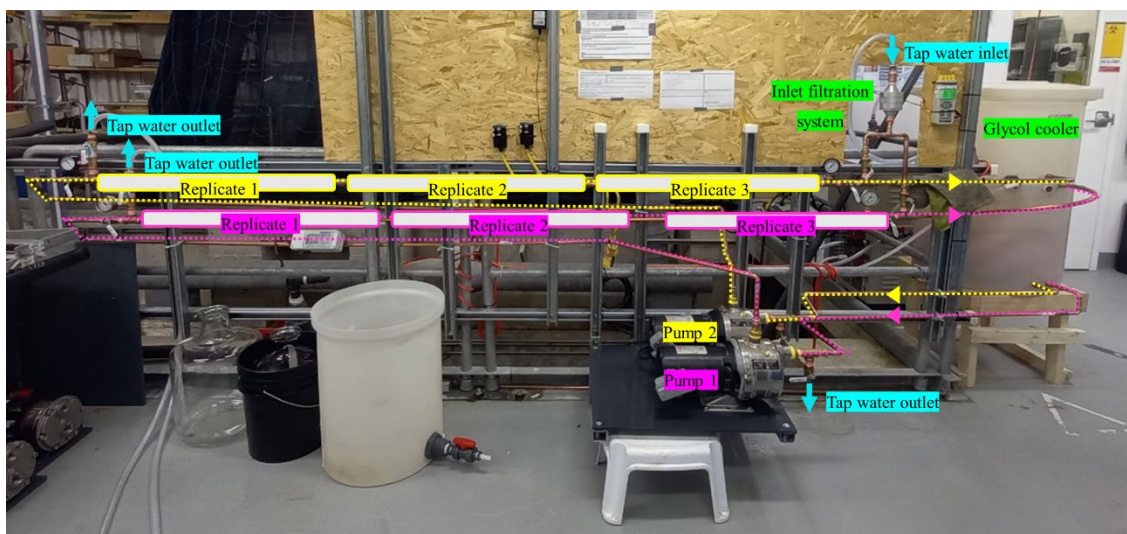


Fig. A1 Photo of the two independent pilots operating in parallel.

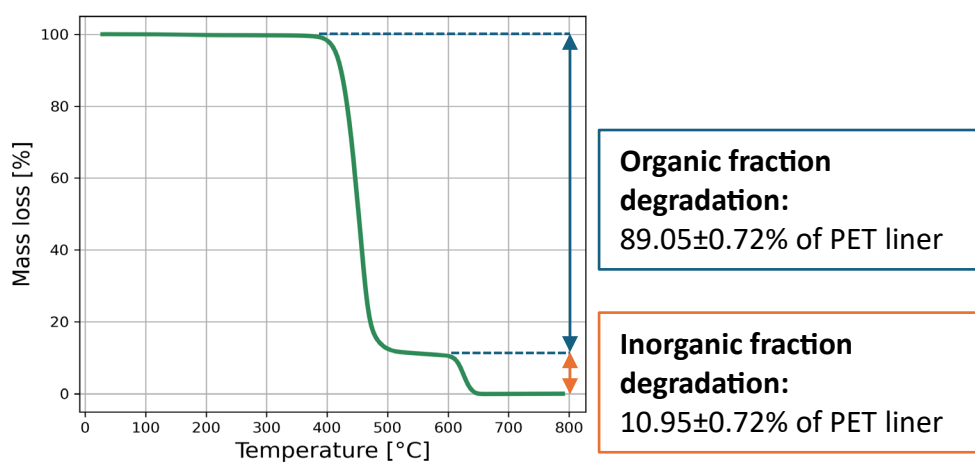


Fig. A2 Polymer/Additives ratio. PET liner thermogram averaged on three unaged PET liner samples. From 0 to 600 °C organic degradation occurs followed by inorganic degradation above 600 °C.

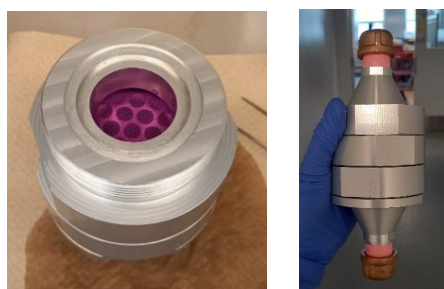


Fig. A3 Photo of the home-made filtration system containing two nylon filters cut from 300 and 28 μm mesh nylon film and dyed with Jacquart iDye Poly Pink (Karakolis et al., 2019) [10] and 304 stainless steel filters as support for the nylon filters.

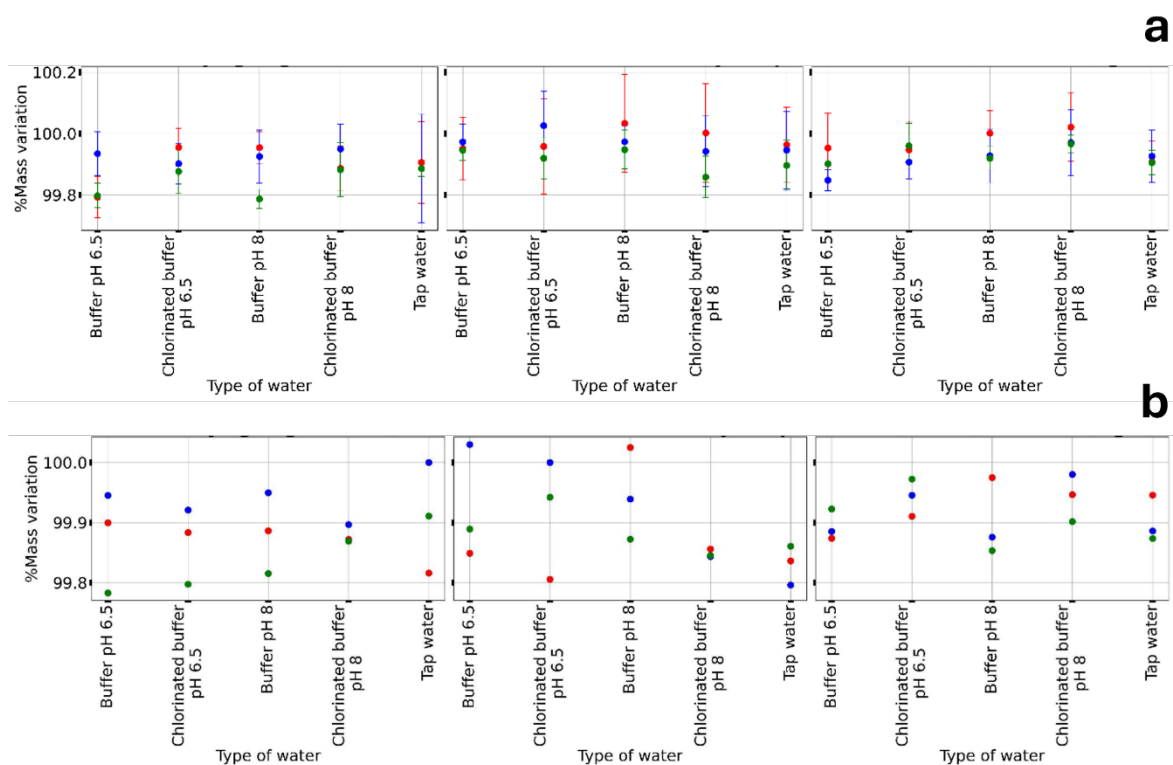


Fig. A4 Mass variations monitoring of the quadruplicates of coupons aged under the same conditions (**a**) and one dried coupon replicate (**b**) at 20 °C (left), 20 °C and 2 FTC per month (center) and 40 °C (right). Legend: Mass variation after 4 (red), 8 (blue) and 12 (green) weeks of aging.

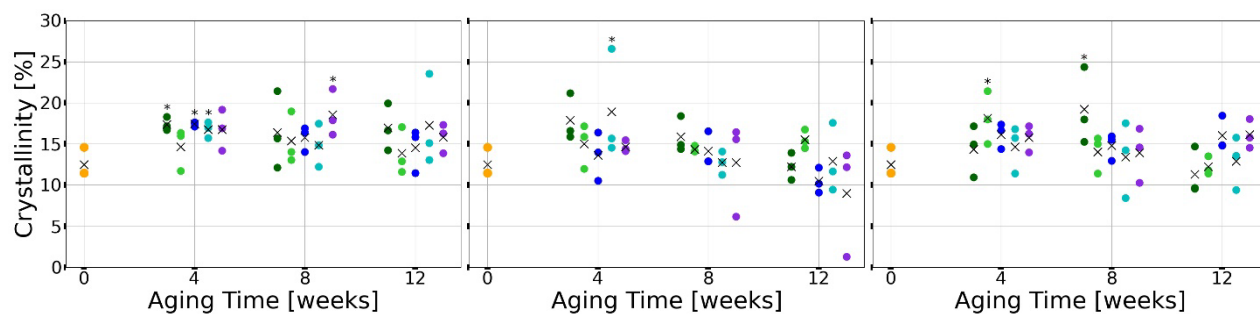


Fig. A5 Aging time against crystallinity of unaged and aged PET liner after 4, 8 and 12 weeks of aging at 20 °C (left), 20 °C and 2 FTC per month (center) and 40 °C (right). Legend: Triplicates of unaged (orange), and static aged coupons/lined pipe in contact with buffer solution at pH 6.5 unchlorinated (dark green) and chlorinated (light green) and at pH 8 unchlorinated (dark blue) and chlorinated (light blue) and tap water (violet). Significance are relative to the unaged PET liner (*) and mean on triplicates (x).

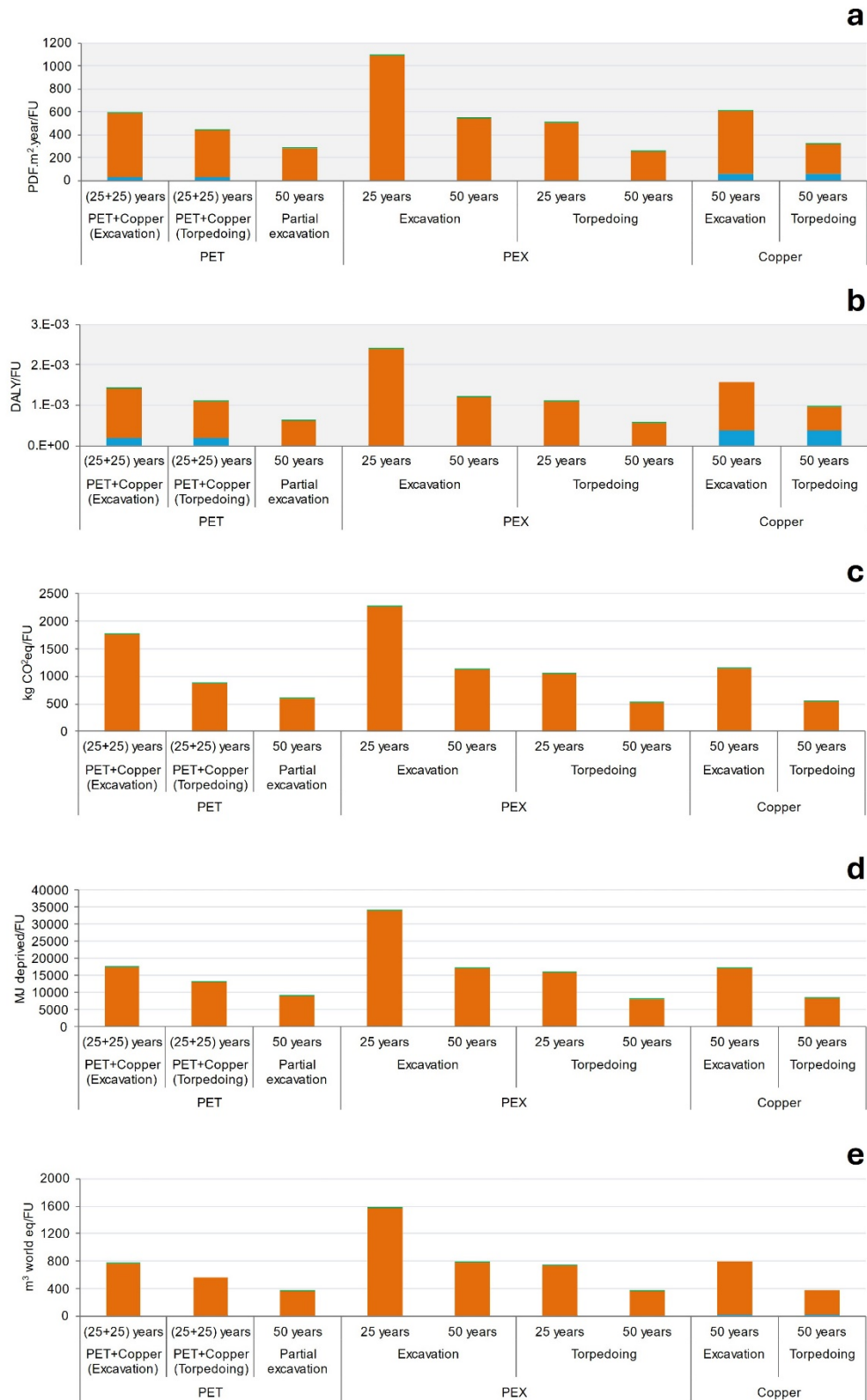


Fig. A6 Sensitivity analysis on service life of plastic technologies varying between 25 and 50 years for the protection areas: EQ (**a**) and HH (**b**) and the impact categories: CC (short-term) (**c**), FNEU (**d**) and WS (**e**). **Legend:** The production (blue), installation (orange), and end of life (green) for the three LSL upgrade scenarios.

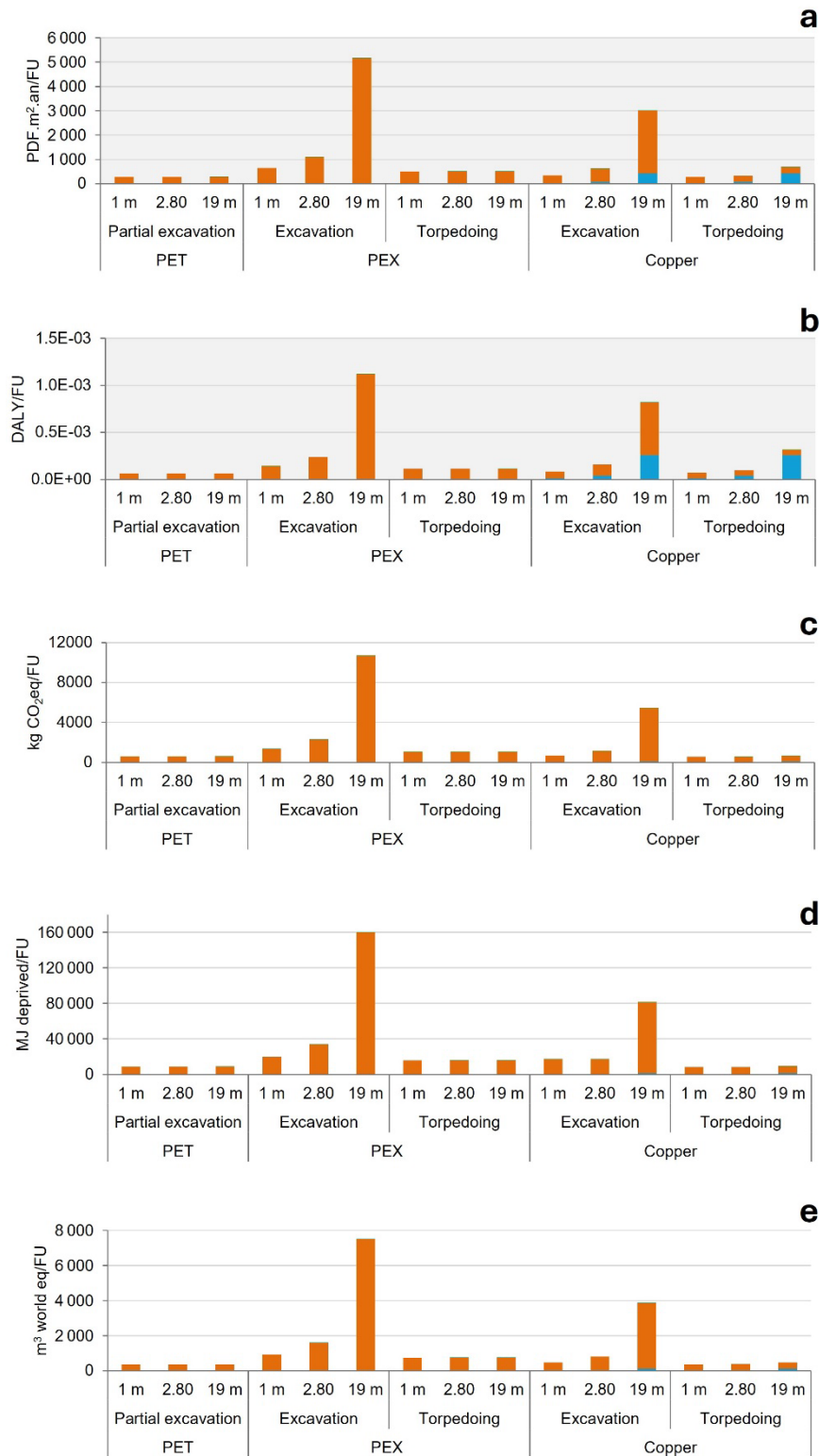


Fig. A7 Sensitivity analysis on LSL length varying between 1 and 19 m for the protection areas: EQ (**a**) and HH (**b**) and the impact categories: CC (short-term) (**c**), FNEU (**d**) and WS (**e**). Legend: The production (blue), installation (orange), and end of life (green). Lifespan of 25 years (PEX pipe) and 50 years (PET liner and copper pipe).

Supporting tables

Table A1 Chlorine decay monitoring over time (t) [weeks] during laboratory aging experiment.

		t=0	t=4	t=8	t=12
Chlorinated buffer solution pH 6.5	20 °C	23.7	19.5	18.8	18.6
	20 °C + 2FTC/month	23.7	3.9	1.1	0.0
	40 °C	23.7	15.8	12.5	1.1
Chlorinated buffer solution pH 8	20 °C	23.8	18.5	17.7	20.5
	20 °C + 2FTC/month	23.8	8.5	4.8	1.0
	40 °C	23.8	16.9	11.5	2.7
Tap water	20 °C	0.03	0.1	0.0	0.0
	20 °C + 2FTC/month	0.03	0.2	0.0	0.0
	40 °C	0.03	0.1	0.0	0.0

Table A2 Chlorine decay monitoring over time (t) [weeks] during pilot static aging experiment.

		t=0	t=8	t=12
Chlorinated buffer solution pH 8	20 °C	23.7	5.9	2.6
	20 °C + 2FTC/month	22.3	0.0	0.0
	40 °C	22.3	0.0	0.0

Table A3 Metals concentration at the tap and ingested over 50 years for lead and copper pipes and the PET liner.

Scenarios	LSL Length	Mean		High-90%	
		Concentration at the tap [µg/L]	Quantity ingested [kg/50years]	Concentration at the tap [µg/L]	Quantity ingested [kg/50years]
Lead service line					
	1	4.63E-01	1.05E-05	2.48E+00	5.61E-05
	2.8	1.30E+00	2.94E-05	4.37E+00	9.91E-05
	19	8.80E+00	1.99E-04	2.14E+01	4.86E-04
Copper service line					
	1	5.50E-00	1.25E-04	2.94E+01	6.66E-04
	2.8	1.54E+01	3.49E-04	5.19E+01	1.18E-03
	19	1.05E+02	2.37E-03	2.54E+02	5.77E-03
PET liner					
Titanium	1	6.16E-02	1.40E-06	3.29E-01	7.46E-06
	2.8	1.72E-01	3.91E-06	5.81E-01	1.32E-05
	19	1.17E+00	2.65E-05	2.85E+00	6.46E-05
Zinc	1	5.82E-01	1.32E-05	3.11E+00	7.05E-05
	2.8	1.63E+00	3.69E-05	5.49E+00	1.24E-04
	19	1.11E+01	2.51E-04	2.69E+01	6.10E-04
Lead	1	2.98E-02	6.77E-07	1.60E-01	3.62E-06
	2.8	8.36E-02	1.89E-06	2.82E-01	6.38E-06
	19	5.67E-01	1.29E-05	1.38E+00	3.13E-05

Table A4 Mean and maximum leached concentrations of titanium, zinc and lead over 4, 8 and 12 weeks of aging depending on water type and thermal conditions.

Leached concentrations [$\mu\text{g/L}$] ¹								
Aging Water	Conditions	Total number of replicate	Titanium		Zinc		Lead	
			Mean ²	Max ³	Mean ²	Max ³	Mean ²	Max ³
Buffer solution pH 6.5	20 °C	n=9	1.57	1.90	7.39	11.69	0.30	0.44
	20 °C + FTC	n=9	*	*	10.60	19.37	0.37	0.68
	40 °C	n=9	1.18	1.74	10.19	11.94	0.32	0.74
Chlorinated buffer solution pH 6.5	20 °C	n=9	0.72	0.82	6.96	12.28	*	*
	20 °C + FTC	n=9	*	*	6.37	16.09	*	0.39
	40 °C	n=9	1.58	2.01	4.13	5.78	*	*
Buffer solution pH 8	20 °C	n=9	0.55	0.77	4.90	12.64	0.92	1.11
	20 °C + FTC	n=8	*	*	2.26	4.82	1.04	1.95
	40 °C	n=9	2.82	3.75	2.05	5.71	0.83	1.01
Chlorinated buffer solution pH 8	20 °C	n=9	1.32	1.58	5.88	12.32	0.63	1.00
	20 °C + FTC	n=7	*	*	5.32	7.57	1.36	1.87
	40 °C	n=9	4.36	9.57	4.74	5.64	0.65	0.98
Atmospheric blank (Type I)	20 °C	n=9	0.01	0.02	0.69	3.18	0.05	0.15
	20 °C + FTC	n=8	0.08	0.11	0.98	2.45	0.30	0.44
	40 °C	n=9	≈0	≈0	0.06	0.28	≈0	0.01

¹ Leached concentrations are obtained by subtracting pre-aging water averages (n=6) from post-aging water averages of replicate (n=2; 3).

² Mean concentrations are calculated by averaging the replicate values leached at 4, 8 and 12 weeks (n=7; 8; 9).

³ Maximum concentrations are replicate averages (n=2; 3) of highest leaching after 4, 8, or 12 weeks.

* No leaching or negative values due to precipitation (Pb) or no apparent leaching (Ti).

Note: Metal release is attributable to the PET liner and not to the glassware or experimental setup, as atmospheric controls without coupons subjected to the same aging process leached a maximum of 0.11 $\mu\text{gTi/L}$, 3.18 $\mu\text{gZn/L}$, and 0.44 $\mu\text{gPb/L}$.

Table A5 Metal exposure (characterization) factors.

Element	EF _{ing,noncanc} [DALY/kg _{intake}]	EF _{ing,canc} [DALY/kg _{intake}]	Reference
Pb (II)	2.33E+01	2.83E-01	USEtox*
Zn (II)	9.22E-02	0.00E+00	USEtox*
Cu (II)	2.47E-02	0.00E+00	USEtox*
Ti (II)	0.00E+00	0.00E+00	REACH

Notes: *USEtox : (Fantke et al., 2017) [8]

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