

Supplementary Information for “Role of Natural Organic Matter and Hardness on Lead Release from Galvanic Corrosion”

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1.0. Materials and Methods



Figure S1. Lead and copper test pieces utilized in the experiment. The lead and copper pipes were attached with flexible polyvinylchloride tubing and separated using a rubber hose washer. The galvanic connection was provided using bronze ground clamps and copper ground wire.

1.1. Pretesting of Test Pieces

Cleaning and pretesting of the test pieces were completed using a similar protocol as Parks et al.

(1). After the test pieces were assembled, they were flushed with tap water then rinsed three times with ultrapure MilliQ Water (18.2 M Ω -cm, 5 μ g/L or less total organic carbon [TOC]) and three times with National Sanitation Foundation (NSF) extraction water (pH = 8.0 \pm 0.5, DIC = 122 \pm 5 mg/L, free chlorine = 2 \pm 0.5 mg/L), as outlined in NSF/American National Standards Institute (ANSI) Standard 61 Section B.9.1. (2). Afterwards, the test pieces were stagnated with NSF extraction water for a period of 24 hours, followed by three subsequent 12-hour stagnations with fresh NSF extraction water. The stagnated water from the three 12-hour stagnations was combined to form a composite sample that was analyzed for total lead and copper using ICP-MS. In total, 14 test pieces were constructed and the 10 test pieces that had the most consistent total lead release were used in the subsequent experimental work.

1.2. Synthetic Water Preparation

Ultrapure MilliQ water (18.2 M Ω -cm, 5 μ g/L or less TOC) was used as a starting matrix for all 10 of the synthetic drinking waters. The pH was controlled by adding either sodium hydroxide (Sigma-Aldrich) from a 0.1 M stock solution or sulfuric acid (Sigma-Aldrich) from a 0.1 M stock solution. The DIC was added as sodium bicarbonate (BDH) from a 0.4 M stock solution. 70% of the hardness was added from calcium chloride dihydrate (EMD Millipore) from a 0.2 M stock solution and 30% of the hardness was added from magnesium sulfate heptahydrate (EMD) from a 0.1 M stock solution. Both calcium and magnesium were added to better simulate real drinking water in Southern Ontario (3-6). Sodium chloride (Sigma-Aldrich) was added from a 0.3 M stock solution and sodium sulfate (EMD) was added from a 0.1 M stock solution to maintain a consistent chloride to sulfate mass ratio of 1.25 (measured range: 1.17–1.29), a chloride concentration of 223 mg/L (measured range: 233-239 mg/L), and a sulfate concentration of 186 mg/L (measured range: 182-200 mg/L). The doses of the chemicals added to the synthetic waters are listed in Table S1.

NOM was added as reference SRNOM (International Humic Substances Society [IHSS], 2R101N) (7) from a stock solution with a concentration of approximately 165 mg DOC/L (8). The SRNOM was composed of 50.70 wt% carbon, 3.97 wt% hydrogen, 41.48 wt% oxygen, 1.27 wt% nitrogen, 1.78 wt% sulfur and an ash content of 4.01 wt% (7). The SRNOM stock solution was prepared by dissolving 200 mg of SRNOM in a 500 mL solution with 20 mL of 0.1 M sodium hydroxide to neutralize the pH (8). The sodium hydroxide, sulfuric acid, and SRNOM were prepared on an as needed basis while the other stock solutions were prepared fresh weekly.

Table S1
Chemical Doses (in mg/L) for Synthetic Waters

Test Piece	NaHCO ₃	CaCl ₂	MgSO ₄	NaCl	Na ₂ SO ₄	NOM ¹	H ₂ SO ₄ ²	NaOH ²
TP1[pH-/DIC-/Hard-/NOM-]	70	39	18	327	199	0	6.8	0
TP2[pH+/DIC-/Hard-/NOM+]	70	39	18	327	199	17.0	0	0.44
TP3[pH-/DIC+/Hard-/NOM+]	560	39	18	327	199	17.0	54	0
TP4[pH+/DIC+/Hard-/NOM-]	560	39	18	327	199	0	0	2.6
TP5[pH-/DIC-/Hard+/NOM+]	70	349	162	0	28	17.0	6.8	0
TP6[pH+/DIC-/Hard+/NOM-]	70	349	162	0	28	0	0	0.44
TP7[pH-/DIC+/Hard+/NOM-]	560	349	162	0	28	0	54.2	0
TP8[pH+/DIC+/Hard+/NOM+]	560	349	162	0	28	17.0	0	2.6
TP9[pH0/DIC0/Hard0/NOM0]	315	194	90	164	114	8.5	5.8	0
TP10[pH0/DIC0/Hard0/NOM0]	315	194	90	164	114	8.5	5.8	0

¹SRNOM concentration added to achieve the DOC target

²Concentrations of H₂SO₄ and NaOH varied to achieve the pH target

Table S2
Schedule of Water Quality Analyses

Parameter	Weeks Measured
Alkalinity	1,5,9,13,17
FEEM*	12,16,20
Galvanic Current	1-20
Ion Chromatography (Chloride and Sulfate)	2,6,11,19
LC-OCD*	3,7,12,16,20
Lead and Copper - Total (Composite)	1-20
Lead and Copper - Total (48 Hour Stagnation)	4,8,10,12,14,16,19,20
Lead and Copper - Dissolved (48 Hour Stagnation)	4,8,10,12,14,16,19,20
Metals (Total)	11
pH	1-20
Specific Conductance	1,5,9,13,17
Turbidity	1,5,9,10,12,14,16,18-20

*The weeks listed are for the synthetic waters with NOM, only one synthetic water without NOM was analyzed per week as a control

Table S3

Average Measured Water Quality Before and After Stagnation

Parameter		TP1[pH-/DIC-/Hard-/NOM-]	TP2[pH+/DIC-/Hard-/NOM+]	TP3[pH-/DIC+/Hard-/NOM-]	TP4[pH+/DIC+/Hard-/NOM-]	TP5[pH-/DIC-/Hard+/NOM+]	TP6[pH+/DIC-/Hard+/NOM-]	TP7[pH-/DIC+/Hard+/NOM-]	TP8[pH+/DIC+/Hard+/NOM+]	TP9[pH0/DIC0/Hard0/NOM0]	TP10[pH0/DIC0/Hard0/NOM0]
pH	Initial*	7.08	8.49	7.08	8.44	7.07	8.44	7.08	8.40	7.81	7.84
	Final*	8.75	8.91	7.71	8.55	8.01	8.22	7.61	8.27	8.26	8.25
DIC (mg C/L)	Initial	9.7	11.3	77.5	77.0	11.1	11.0	75.3	78.8	42.9	43.1
	Final	8.0	12.3	65.9	75.1	10.4	10.5	62.0	74.1	41.2	42.1
Hardness (mg CaCO ₃)	Initial	48	47	45	48	404	403	415	409	199	232
	Final	51	50	48	43	426	419	440	416	207	242
DOC (mg C/L)	Initial	n.m.**	7.68	8.09	n.m.	7.28	n.m.	n.m.	7.50	3.74	3.66
	Final	n.m.	7.36	7.07	n.m.	5.46	n.m.	n.m.	6.79	3.56	3.58
Specific Conductance (μS/cm)	Initial	1,220	1,230	1,610	1,670	1,070	1,260	1,520	1,480	1,380	1,320
	Final	1,230	1,230	1,600	1,520	1,140	1,250	1,520	1,570	1,380	1,200
Turbidity (NTU)	Initial	0.08	0.15	0.15	0.09	0.16	0.07	0.08	0.16	0.12	0.11
	Final	0.56	1.14	5.36	0.38	8.17	0.29	1.16	0.52	0.53	0.33

*Initial measurements were taken prior to stagnation in test pieces, final measurements were taken after select 48-hour stagnation periods in the test pieces

**DOC not measured regularly for samples without SRNOM added

1.3. Sample Collection

Samples from the test pieces were collected in 500 mL low-density polyethylene bottles that were pre-washed in a 7% nitric acid bath (Fisher Scientific trace-metal grade) for a minimum of 16 hours. Samples were taken periodically for a variety of water quality analyses (Table S2) including total and dissolved lead and copper using ICP-MS following the final 48-hour stagnation event of the week (Monday-Wednesday). The total and dissolved lead and copper samples from the 48-hour stagnation events were measured on a biweekly basis, while the other water quality parameters were measured monthly (Table S2). Samples for dissolved lead and copper analysis were filtered through a 0.45 μm cellulose nitrate membrane filter (General Electric Healthcare Life Sciences WhatmanTM) prior to acidification in polypropylene (Caplug) vials with 7% nitric acid to pH 2 or less (Fisher Scientific trace-metal grade). It is noted that this is an operational definition of dissolved lead and copper, as some colloidal lead and copper will pass through the filter and will be included in this fraction as well. Additionally, a composite sample was taken weekly that combined 3.5 mL of acidified effluent from the week's three "dump and fill" events for total lead and copper analysis using ICP-MS (Table S2). Samples for total lead and copper analysis were acidified with 70% nitric acid (Fisher Scientific trace-metal grade) to pH 2 or less and were held for a minimum of 16 hours before being transferred to polypropylene vials (Caplug). The acidification for collection of samples for total lead and copper analysis was completed immediately after samples were collected for other water quality analyses, if any.

1.4. Changes to Water Quality Following Stagnation in the Test Pieces

Water quality parameters were routinely analyzed before and after the final 48-hour stagnation

event in the test pieces as summarized in Table S3. The pH increased following stagnation in most of the synthetic waters, and this was the most noticeable in TP1[pH-/DIC-/Hard-/NOM-] which increased from 7.08 to 8.75 on average. However, in TP6[pH+/DIC-/Hard+/NOM-] and TP8[pH+/DIC+/Hard+/NOM+] the pH decreased slightly. It is thought that these changes are likely due to an equilibrium being reached between the water and the corrosion scales. The DIC concentration decreased slightly following stagnation in all the waters except for TP2[pH+/DIC-/Hard-/NOM+], which increased slightly. This could be due to the incorporation of DIC into the corrosion scales or carbon dioxide exchange with the atmosphere. Hardness was only measured once before and after stagnation in week 11, but it did increase in nine out of the 10 test pieces. This indicates that the incorporation of hardness into the corrosion scales was not substantial. As discussed in the LC-OCD results section in the main manuscript, the DOC decreased in all the synthetic waters with SRNOM. This may have been due to the incorporation of humic substances into the corrosion scales. The specific conductance was virtually unchanged during stagnation, suggesting that dissolved substances were not incorporated into the corrosion scales in appreciable amounts. The turbidity increased in all the synthetic waters and was more notable when NOM was present. It is thought that this is due to particulate and colloidal lead containing constituents.

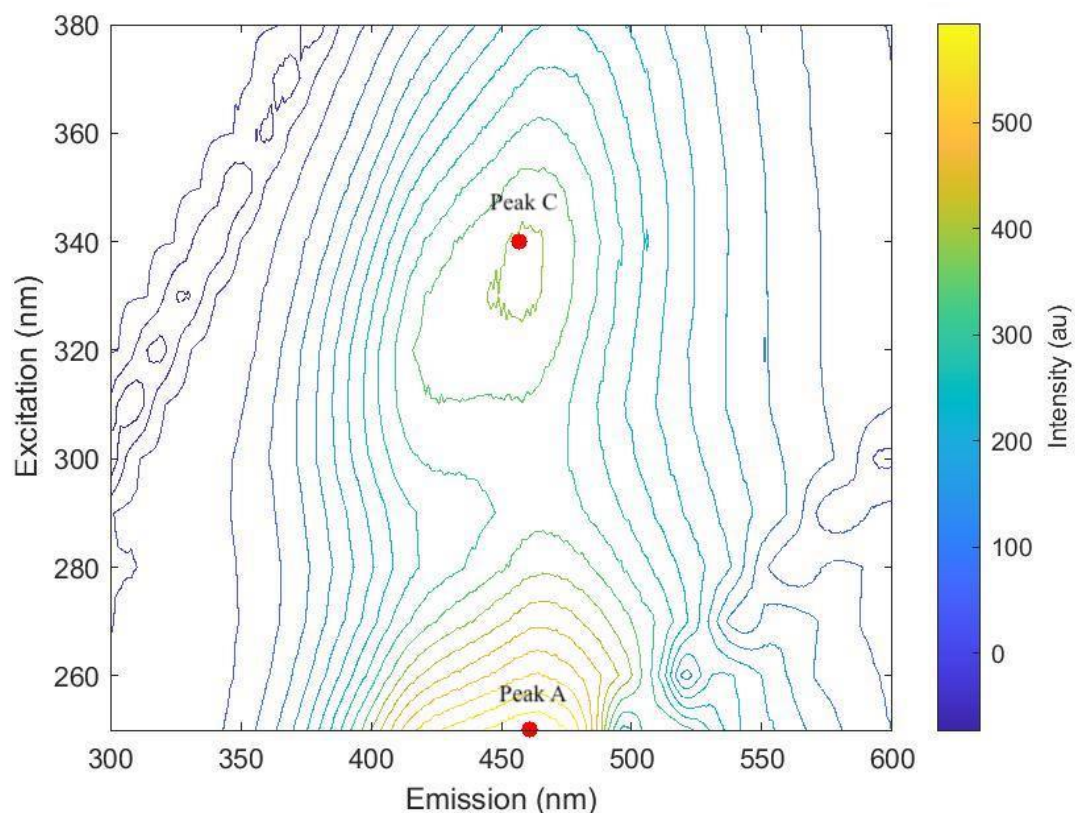


Figure S2. FEEM contour plot for TP3[pH-/DIC+/Hard-/NOM+] prior to stagnation on week 12. Peak picking was used to estimate humic-like peak A (Ex 250nm/Em 461nm) and peak C (Ex 340nm/Em 457nm) maxima. The excitation (Ex) wavelengths ranged from 250 to 380 nm with 10 nm increments and the emission (Em) wavelengths were monitored from 300 to 600 nm at 1 nm increments.

1.5. Scale Analysis

The scale analysis involved the examination of scales inside the lead and copper pipes using similar methods that were employed previously (9, 10). A section of the pipe was cut open into two half cylinders and another was embedded in epoxy to preserve the scale morphology. The embedded section was cut and polished using fine sandpaper. Each cross-section was imaged using a Thermofisher Quattro S E-SEM, and energy dispersive X-ray spectroscopy (Oxford AzTec Energy Dispersive X-ray Spectrometer) was used to semi-quantitatively determine the spatial distribution of elements within the scale. The elemental weight percentage (semi-quantitative) recorded in Table S11 for all pipes were taken from the measurements made by EDS. The SEM images with spot analysis were done at a magnification of 8,000 times up to 15,000 times, whichever combination gave the highest resolution. Scale removed from the half cylinders was analyzed by X-Ray powder diffraction (XRD) on a Bruker d8 Advance X-ray diffractometer with Cu Ka radiation at 40 kV and 40 mA. The database of the International Center for Diffraction Data (ICDD) was used for reference pattern matching. Portions of the scales were also digested in a mixture of nitric acid and hydrochloric acid in a ratio of 1:3 by volume and were then analyzed for their elemental compositions using ICP -MS (PerkinElmer Elan DRCII). The ICP-MS was used for a broader set of elements but Table S10 and S13 only report the elements that were detected in the pipes: Pb, Cu, Ca, Na, Mg and Zn.

1.6. Statistical Analysis

ANOVA assumes that the variance is similar among conditions, the observations are independent of time, and the data is normally distributed (11). As the fractional factorial design included one test piece under each condition (other than the mid-point replicates) it was not

possible to reject the null hypothesis that the variance was the same among the test pieces. The observations were independent of time as all 10 test pieces had dump and fill experiments completed during the same time period and the statistical analysis was completed on the average response (i.e., galvanic current, lead release, and copper release) over the 20-week experiment. It was difficult to determine if the data was normally distributed as only seven effects were calculated. Therefore, the Box Cox method was utilized to optimize normality to meet this assumption as best as possible as described below.

The Box Cox method involved determining the optimal value of λ for a power transformation ($y^* = y^\lambda$) by using the method of maximum likelihood (11). In accordance with Montgomery (11), ANOVA was completed for various values of λ using

$$y^{(\lambda)} = \begin{cases} \frac{y^\lambda - 1}{\lambda \dot{y}^{\lambda-1}} & \lambda \neq 0 \\ \dot{y} \ln y & \lambda = 0 \end{cases} \quad (S1)$$

where \dot{y} is the geometric mean of the observations calculated as

$$\dot{y} = \ln^{-1}[(1/n)\Sigma \ln y] \quad (S2)$$

The optimal value of λ is the value that minimizes the error (*SSE*). An approximate $100(1 - \alpha)$ confidence interval for λ was calculated as

$$SS^* = SSE(\lambda) \left(1 + \frac{t_{\alpha/2, v}^2}{v}\right) \quad (S3)$$

where α is the significance (0.05) and v is the number of degrees of freedom (2). The value of SS^* represents the upper bound of the error in the confidence interval. If $\lambda = 1$ was included in the 95% confidence interval no transformation was required. This was the case for the galvanic current and lead release data, but not the copper release data. As the total, dissolved, and particulate copper data all included $\lambda = 0.5$ in the 95% confidence interval, that value was

selected, which is equivalent to a square root transformation. Normal probability plots were generated for each test piece to check the normality of the dataset once an optimal λ value was identified.

After the data was transformed, if necessary, the ANOVA analysis was completed. Since two mid-point replicates were included, it was possible to calculate both the pure error and lack of fit error (difference between the total error and pure error). The pure error was used to assess the significance of the effects. The lack of fit error provided an indication of whether the linear model was appropriate. For all data sets the lack of fit error was not significant ($p > 0.10$) except for particulate lead ($p = 0.089$), which was potentially significant. This suggested that making statistical inferences using ANOVA was not appropriate for the particulate lead data.

1.7. Solubility Modelling Methodology

Lead solubility was modeled using tidyphreeqc (12), an R interface for the USGS' PHREEQC (13), and pbcusol (14), an extension of tidyphreeqc for lead and copper solubility modeling. Several other contributed R packages were used (15-17). The default thermodynamic database in pbcusol (used here) is the *minteq* database available in PHREEQC, with modifications for consistency with the data provided in Schock et al. (18) Table 4-14 (LEADSOL values). The relevant reactions for lead are shown in Table S4 (13, 18). Activity coefficients to compensate for the ionic strength were considered using the Davies equation for charged species;

$$\log \gamma = -Az^2 \left(\frac{\sqrt{\mu}}{1 + \mu} - 0.3\mu \right) \quad (\text{S4})$$

where γ is the activity coefficient, μ is the ionic strength, A is a constant at a given temperature, and z is the number of equivalents of exchanger in the exchange species (13). Activity coefficients for uncharged species were determined using (13);

$$\log \gamma = 0.1\mu \quad (\text{S5})$$

The effect of NOM on lead solubility was modeled using the approach outlined in Example 19 of Parkhurst and Appelo (13). Complexation constants for protons and cations on mono- and bi-dentate sites were sourced from the *Tipping_Hurley* database available in PHREEQC. The solubilities of cerussite and hydrocerussite were calculated separately at each experimental condition by fixing the saturation index of each phase at zero. The least soluble phase was considered to control the solubility (hydrocerussite for TP2[pH+/DIC-/Hard-/NOM+] and TP6[pH+/DIC-/Hard+/NOM-], and cerussite for all the other synthetic waters). R code to reproduce the model results is included below, and source code for the relevant functions in *pbculsol* is available on GitHub (14).

```
library("tidyverse")
# remotes::install_github("paleolimbot/chemr")
library("chemr")
# remotes::install_github("bentrueman/pbcusol")
library("pbcusol")

# data:

wqdata <- tibble::tribble(
  ~pipe, ~pH, ~DIC, ~Ca, ~Mg, ~Na, ~Cl, ~SO4, ~DOC,
    1, 7, 10, 14, 3.6, 226.7, 223.2, 186, 0,
    2, 8.5, 10, 14, 3.6, 230.1, 223.2, 186, 7,
    3, 7, 80, 14, 3.6, 338.5, 223.2, 186, 7,
    4, 8.5, 80, 14, 3.6, 365.4, 223.2, 186, 0,
    5, 7, 10, 126.1, 32.8, 43, 223.2, 186, 7,
    6, 8.5, 10, 126.1, 32.8, 46.4, 223.2, 186, 0,
    7, 7, 80, 126.1, 32.8, 154.7, 223.2, 186, 0,
    8, 8.5, 80, 126.1, 32.8, 181.6, 223.2, 186, 7,
    9, 7.75, 45, 70.1, 18.2, 202.3, 223.2, 186, 3.5,
    10, 7.75, 45, 70.1, 18.2, 202.3, 223.2, 186, 3.5
)
```



```

# Lead solubility:

wqout <- wqdata %>%
  crossing(phase_in = c("Cerussite", "Hydcerussite")) %>%
  rowwise() %>%
  mutate(
    model = list(pbcusol::pb_sol_wham(
      ph = pH,
      dic = DIC,
      phase = phase_in,
      Ca = Ca / chemr::mass("Ca"),
      Mg = Mg / chemr::mass("Mg"),
      Na = Na / chemr::mass("Na"),
      Cl = Cl / chemr::mass("Cl"),
      `S(6)` = SO4 / chemr::mass("SO4"),
      mass_ha = DOC * 1e-3 * .86 # NOM is 86% humic
    ))
  ) %>%
  unnest(model, names_repair = tidyr_legacy) %>%
  ungroup() %>%
  mutate(
    # convert moles of phase dissolved to µg Pb/L:
    pb_tot = coalesce(mol_Cerussite, mol_Hydcerussite * 3),
    pb_tot = pb_tot * 1e6 * chemr::mass("Pb")
  ) %>%
  # phase with minimum solubility:
  group_by(pipe, pH, DIC, Ca, DOC) %>%
  summarize(
    phase = phase[which.min(pb_tot)],
    pb_tot = min(pb_tot)
  ) %>%
  ungroup()

```

Table S4

Summary of Thermodynamic Data Used in Equilibrium Solubility Modeling

Reaction Type	Equation	Log K*
cerussite dissolution	$\text{PbCO}_3 \leftrightarrow \text{Pb}^{2+} + \text{CO}_3^{2-}$	-13.11
hydrocerussite dissolution	$\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2 + 2\text{H}^+ \leftrightarrow 3\text{Pb}^{2+} + 2\text{CO}_3^{2-} + 2\text{H}_2\text{O}$	-18.00
complexation with H_2O	$\text{Pb}^{2+} + \text{H}_2\text{O} \leftrightarrow \text{PbOH}^+ + \text{H}^+$	-7.22
	$\text{Pb}^{2+} + 2\text{H}_2\text{O} \leftrightarrow \text{Pb}(\text{OH})_2 + 2\text{H}^+$	-16.91
	$\text{Pb}^{2+} + 3\text{H}_2\text{O} \leftrightarrow \text{Pb}(\text{OH})_3^- + 3\text{H}^+$	-28.08
	$\text{Pb}^{2+} + 4\text{H}_2\text{O} \leftrightarrow \text{Pb}(\text{OH})_4^{2-} + 4\text{H}^+$	-39.72
	$2\text{Pb}^{2+} + \text{H}_2\text{O} \leftrightarrow \text{Pb}_2\text{OH}^{3+} + \text{H}^+$	-6.36
	$3\text{Pb}^{2+} + 4\text{H}_2\text{O} \leftrightarrow \text{Pb}_3(\text{OH})_4^{2+} + 4\text{H}^+$	-23.86
	$4\text{Pb}^{2+} + 4\text{H}_2\text{O} \leftrightarrow \text{Pb}_4(\text{OH})_4^{4+} + 4\text{H}^+$	-20.88
	$6\text{Pb}^{2+} + 8\text{H}_2\text{O} \leftrightarrow \text{Pb}_6(\text{OH})_8^{4+} + 8\text{H}^+$	-43.62
complexation with CO_3^{2-}	$\text{Pb}^{2+} + \text{CO}_3^{2-} \leftrightarrow \text{PbCO}_3^0$	7.10
	$\text{Pb}^{2+} + 2\text{CO}_3^{2-} \leftrightarrow \text{Pb}(\text{CO}_3)_2^{2-}$	10.33
	$\text{Pb}^{2+} + \text{CO}_3^{2-} + \text{H}^+ \leftrightarrow \text{PbHCO}_3^+$	12.59
complexation with SO_4^{2-}	$\text{Pb}^{2+} + \text{SO}_4^{2-} \leftrightarrow \text{PbSO}_4^0$	2.73
	$\text{Pb}^{2+} + 2\text{SO}_4^{2-} \leftrightarrow \text{Pb}(\text{SO}_4)_2^{2-}$	3.50
complexation with Cl^-	$\text{Pb}^{2+} + \text{Cl}^- \leftrightarrow \text{PbCl}^+$	1.59
	$\text{Pb}^{2+} + 2\text{Cl}^- \leftrightarrow \text{PbCl}_2^0$	1.80
	$\text{Pb}^{2+} + 3\text{Cl}^- \leftrightarrow \text{PbCl}_3^-$	1.71
	$\text{Pb}^{2+} + 4\text{Cl}^- \leftrightarrow \text{PbCl}_4^{2-}$	1.43

*Log K values are from Schock et al. (18)

2.0. Results and Discussion

Table S5

Galvanic Current, Lead Release, and Copper Release from the Test Pieces

Parameter		TP1[pH-/DIC-/Hard-/NOM-]	TP2[pH+/DIC-/Hard-/NOM+]	TP3[pH-/DIC+/Hard-/NOM+]	TP4[pH+/DIC+/Hard-/NOM-]	TP5[pH-/DIC-/Hard+/NOM+]	TP6[pH+/DIC-/Hard+/NOM-]	TP7[pH-/DIC+/Hard+/NOM-]	TP8[pH+/DIC+/Hard+/NOM+]	TP9[pH0/DIC0/Hard0/NOM0]	TP10[pH0/DIC0/Hard0/NOM0]
Galvanic Current ¹ (μ A)	Mean	25.0	26.5	71.9	33.2	25.0	21.6	51.4	55.2	45.8	41.3
	SD ²	4.4	1.4	7.0	3.8	3.5	4.9	4.3	3.1	4.7	3.6
	CV ²	17.5	5.2	9.8	11.4	14.2	22.7	8.4	5.5	10.1	8.8
Total Lead ³ (μ g/L)	Mean	693	3,700	12,600	4,480	7,110	1,900	8,390	1,130	2,620	1,380
	SD	343	569	9,060	5,500	3,690	1,330	6,530	386	925	126
	CV	49.4	15.4	72.0	123	51.9	69.8	77.9	34.3	35.3	9.1
Dissolved Lead ⁴ (μ g/L)	Mean	166	2,310	3,890	132	3,050	122	140	578	1,580	1,140
	SD	42.0	230	503	28.9	620	30.0	48.1	145	319	81.6
	CV	25.3	10.0	12.9	21.9	20.4	24.6	34.4	25.2	20.2	7.1
	% of Total	29.4	56.2	17.7	3.3	39.9	8.0	1.5	69.8	54.5	81.4
Particulate Lead ⁴ (μ g/L)	Mean	399	1,800	18,100	3,840	4,590	1,400	9,490	250	1,320	260
	SD	244	1,290	22,200	4,680	2,470	1,280	9,940	171	718	100
	CV	61.3	71.4	123	122	53.8	91.3	105	68.5	54.4	38.6
	% of Total	70.6	43.8	82.3	96.7	60.1	92.0	98.5	30.2	45.5	18.6
Total Copper ³ (μ g/L)	Mean	10.9	91.4	438	25.8	80.7	7.5	55.1	98.4	84.5	69.7
	SD	5.5	75.9	152	8.4	24.1	5.5	32.4	23.0	26.6	16.7
	CV	50.1	83.0	34.7	32.7	29.9	72.7	58.8	23.3	31.5	24.0

Table S5 (continued)

Galvanic Current, Lead Release, and Copper Release from the Test Pieces

Parameter		TP1 [pH-/DIC-/Hard-/NOM-]	TP2 [pH+/DIC-/Hard-/NOM+]	TP3 [pH-/DIC+/Hard-/NOM+]	TP4 [pH+/DIC+/Hard-/NOM-]	TP5 [pH-/DIC-/Hard+/NOM+]	TP6 [pH+/DIC-/Hard+/NOM-]	TP7 [pH-/DIC+/Hard+/NOM-]	TP8 [pH+/DIC+/Hard+/NOM+]	TP9 [pH0/DIC0/Hard0/NOM0]	TP10 [pH0/DIC0/Hard0/NOM0]
Dissolved Copper ⁴ (µg/L)	Mean	2.9	47.2	326	12.1	29.5	1.4	39.8	71.8	55.4	44.9
	SD	2.6	9.1	89.3	3.4	6.0	1.2	16.4	16.2	7.8	5.3
	CV	88.5	19.2	27.4	27.9	20.4	84.9	41.3	22.6	14.1	11.8
	% of Total	29.9	61.2	83.0	56.1	39.1	13.2	55.7	77.2	72.6	72.1
Particulate Copper ⁴ (µg/L)	Mean	6.9	29.9	66.7	9.5	46.0	9.0	31.7	21.2	20.9	17.4
	SD	6.6	53.4	25.8	5.1	20.3	9.1	34.3	8.9	10.1	4.0
	CV	95.5	178	38.7	53.6	44.2	101	108	42.1	48.3	22.9
	% of Total	70.1	38.8	17.0	43.9	60.9	86.8	44.3	22.8	27.4	27.9

¹Galvanic current was measured between the lead and copper pipes at the end of the final 48-hour stagnation event of the week

²SD = standard deviation, CV = coefficient of variation

³Total lead and copper were measured from the weekly composite samples

⁴Dissolved and particulate lead and copper were measured from the samples collected following the final 48-hour stagnation event on weeks 4,8,10,12,14,16,19, and 20

Table S6
Effects and Significance of Water Quality Factors Determined Using ANOVA

Parameter		Main Effects				Interaction Effects ^{1,2}		
		pH (P)	DIC (D)	Hardness (H)	NOM (N)	PD + HN	PH + DN	PN + DH
Galvanic Current (μ A)	Low (-) ³	43.3	24.5	39.2	32.8	42.8	34.0	37.9
	Mid-point (0) ³	43.6	43.6	43.6	43.6	43.6	43.6	43.6
	High (+) ³	34.1	52.9	38.3	44.6	34.6	43.4	39.5
	Effect ⁴	-9.2	28.4	-0.9	11.8	-8.2	9.4	1.6
	p-value	0.15	0.051⁵	0.76	0.12	0.17	0.15	0.61
Total Lead (μ g/L)	Low (-)	7,190	3,350	5,360	3,870	6,640	5,920	6,520
	Mid-point (0)	2,000	2,000	2,000	2,000	2,000	2,000	2,000
	High (+)	2,800	6,640	4,630	6,130	3,350	4,080	3,480
	Effect	-4,390	3,290	-730	2,260	-3,290	-1,840	-3,040
	p-value	0.089	0.12	0.45	0.17	0.12	0.21	0.13
Dissolved Lead (μ g/L)	Low (-)	1,810	1,410	1,630	140	1,620	1,410	1,800
	Mid-point (0)	1,360	1,360	1,360	1,360	1,360	1,360	1,360
	High (+)	785	1,190	970	2,460	980	1,190	800
	Effect	-1,030	-230	-650	2,320	-640	-220	-1,000
	p-value	0.13	0.49	0.21	0.060	0.21	0.50	0.14
Particulate Lead (μ g/L) ⁶	Low (-)	8,140	2,050	6,030	3,780	7,690	4,930	6,980
	Mid-point (0)	790	790	790	790	790	790	790
	High (+)	1,820	7,920	3,930	6,180	2,270	5,030	2,980
	Effect	-6,320	5,870	-2,100	2,400	-5,420	100	-4,000
	p-value	0.053	0.057	0.16	0.14	0.062	0.88	0.084
Total Copper (μ g/L)	Low (-)	146	47.6	141	24.8	148	63.3	138
	Mid-point (0)	77.1	77.1	77.1	77.1	77.1	77.1	77.1
	High (+)	55.8	154	60.4	177	54.0	139	64.0
	Effect	-90	107	-81	152	-94	75	-74
	p-value	0.076	0.054	0.11	0.034	0.080	0.16	0.13
Dissolved Copper (μ g/L)	Low (-)	99.5	20.2	97.0	14.0	104	32.1	92.2
	Mid-point (0)	50.1	50.1	50.1	50.1	50.1	50.1	50.1
	High (+)	33.1	112	35.6	119	29.1	101	40.4
	Effect	-66.4	92	-61.4	105	-74	68	-51.8
	p-value	0.083	0.045	0.11	0.036	0.071	0.13	0.19
Particulate Copper (μ g/L)	Low (-)	37.8	22.9	28.2	14.2	34.3	29.3	32.8
	Mid-point (0)	19.1	19.1	19.1	19.1	19.1	19.1	19.1
	High (+)	17.4	32.3	27.0	41.0	20.9	25.9	22.4
	Effect	-20.4	9.3	-1.3	26.7	-13.4	-3.3	-10.4
	p-value	0.056	0.096	0.35	0.042	0.12	0.31	0.10

¹PD+HN represents the sum of the confounded pH/DIC and hardness/NOM interaction effects.
²PH+DN represents the sum of the confounded pH/hardness and DIC/NOM interaction effects.
³PN+DH represents the sum of the confounded pH/NOM and DIC/hardness interaction effects.

²The low level for interaction effects indicates that factors were at different levels (i.e. one at the low level and the other at the high level). The high level for interaction effects indicates that the factors were at the same level (i.e. both at the low level or both at the high level).

³Low (n=4), mid-point (n=2), and high (n=4) indicate the average value when the water quality factor was at that level.

⁴Effects were calculated from untransformed data while significance was calculated from untransformed data for galvanic current and lead release and Box Cox transformed data with $\lambda=0.5$ for copper.

⁵Bolded values were found to be significant at 10% and bolded and italicized values were found to be significant at 5%.

⁶The effects for particulate lead release could not be statistically determined as the ANOVA model had a potentially significant lack of fit error ($p=0.089$).

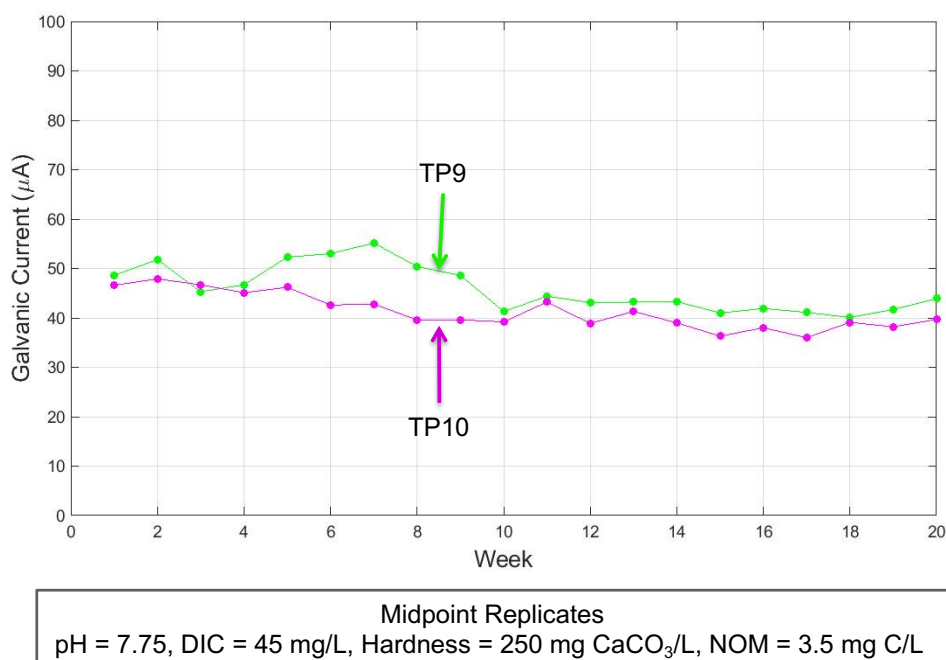


Figure S4. Panel plot of time series of galvanic current between the lead and copper pipes of the test pieces during the 20-week study for TP9 and 10. The galvanic current was measured following a 48-hour stagnation period.

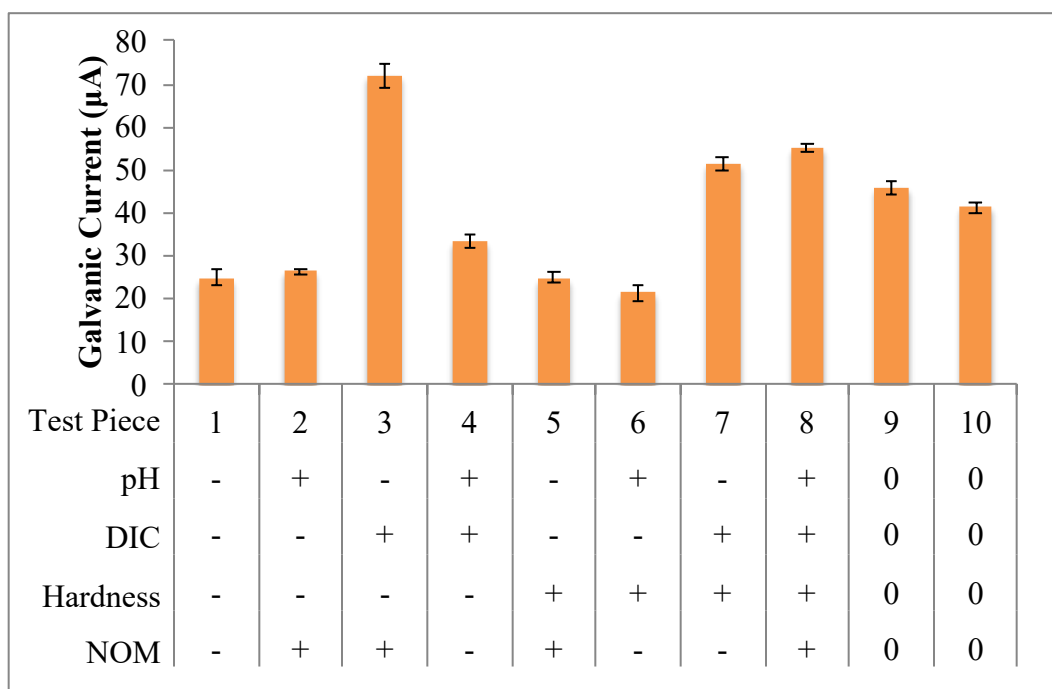


Figure S5. Galvanic current measured between lead and copper pipes of the test pieces. The galvanic current was measured at the end of the final 48-hour stagnation event of the week. Bars represent the average values and error bars represent the 90% confidence interval.

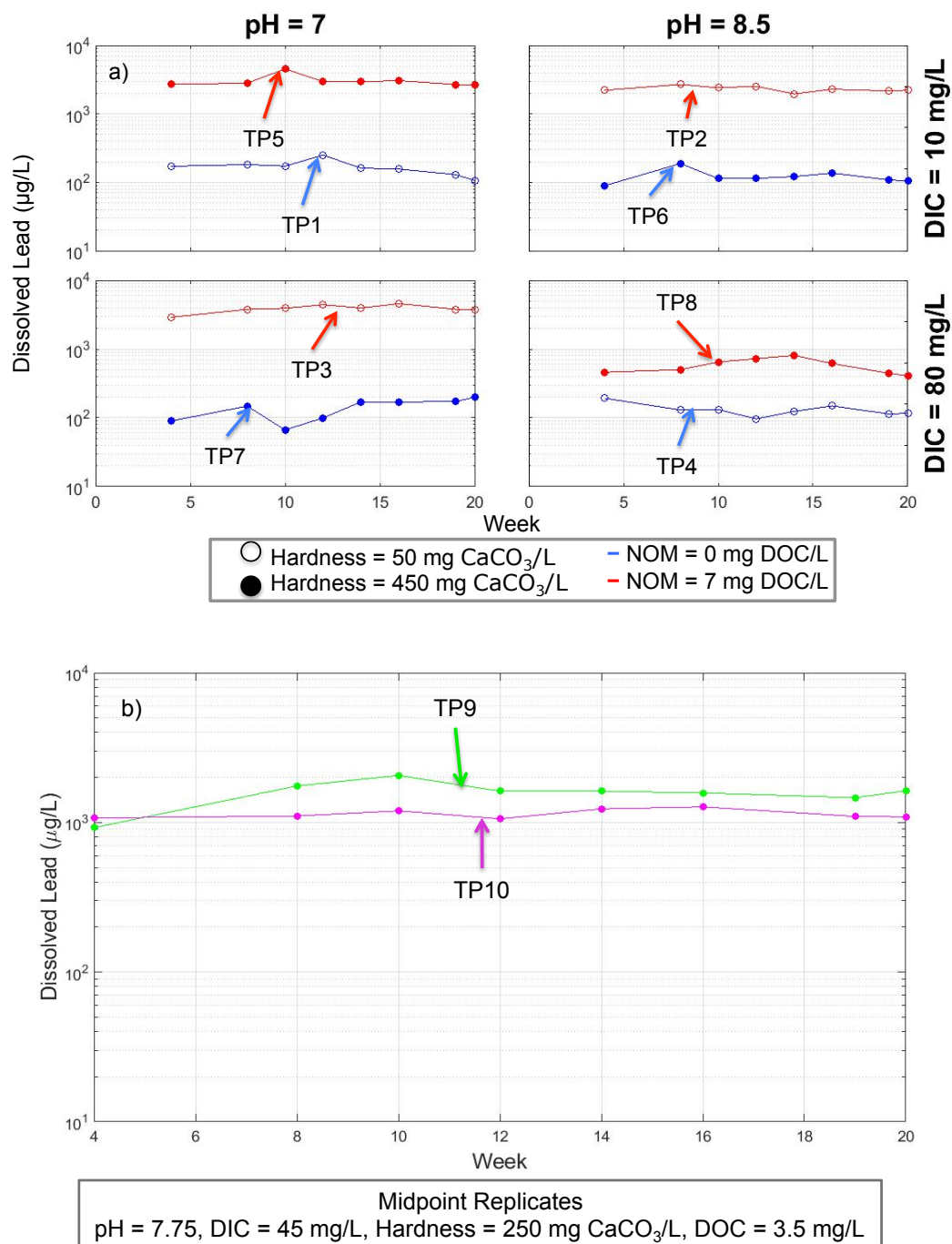


Figure S6. Panel plot of dissolved lead concentrations in the stagnated water from a) TP1-8 and b) TP9-10. Dissolved lead was measured from water samples collected following a 48-hour stagnation period on select weeks.

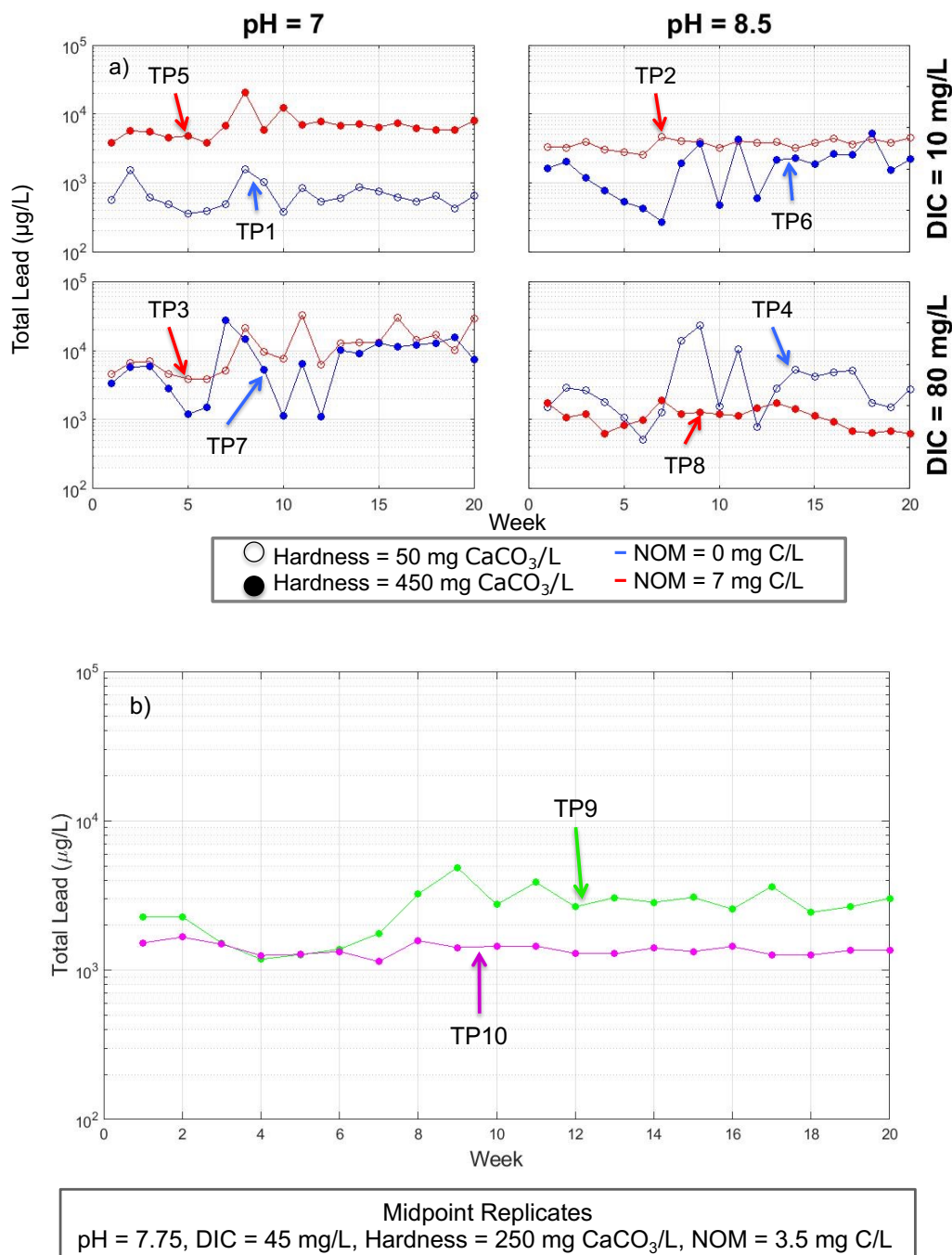


Figure S7. Panel plot of total lead concentrations in the stagnated water from a) TP1-8 and b) TP9-10. Total lead was measured from composite samples combining stagnated water from the week's three dump and fill events.

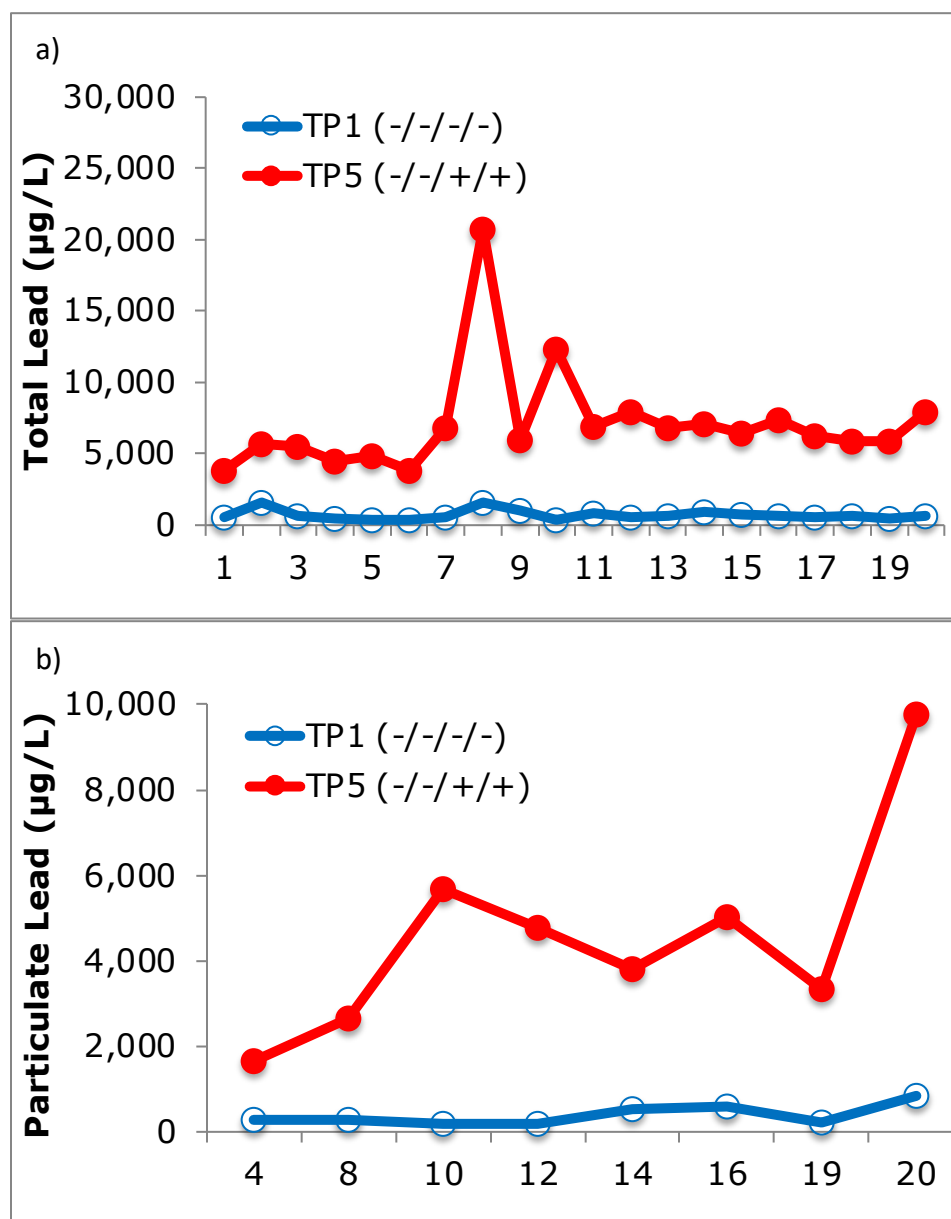


Figure S8. a) Total lead release from TP1[pH-/DIC-/Hard-/NOM-] and TP5 [pH-/DIC-/Hard+/NOM+]. Total lead was measured from composite samples combining stagnated water from the week's three dump and fill events. b) Particulate lead release from from TP1[pH-/DIC-/Hard-/NOM-] and TP5 [pH-/DIC-/Hard+/NOM+]. Particulate lead was measured from water samples collected following a 48-hour stagnation period on select weeks.

2.1. Comparison of Mass of Oxidized Lead Predicted by Faraday's Law and Mass of Lead Released

Faraday's Law was used to estimate the production of oxidized lead using the below equation;

$$m_F = \frac{ItM}{2F} \quad (S6)$$

where m_F is the mass of oxidized lead (g), I is the galvanic current (A, 1 A = 1 C/s), t is the time (s), M is the molar mass (g/mol), and F is Faraday's constant (96,500 C/mol). Assuming the galvanic current was constant throughout the entire week, it was possible to estimate the mass of oxidized lead that was produced. In addition, the mass of lead that was actually released per week (m_R) was calculated as

$$m_R = C \times 1000 \left(\frac{\pi d_L^2 L}{4} + \frac{\pi d_C^2 L}{4} \right) \times 3 \quad (S7)$$

which can be further simplified to;

$$m_R = 750CL\pi(d_L^2 + d_C^2) \quad (S8)$$

where C is the concentration of lead in the weekly composite sample in $\mu\text{g/L}$, L is the length of the lead and copper pipes (0.50 m), d_L is the inner diameter of the lead pipe (0.0191 m), and d_C is the inner diameter of the copper pipe (0.0206 m). Equation S7 represents the concentration of lead in the weekly composite sample multiplied by three (three dump and fill events per week) times the volume of water in the test pieces ($0.309 \text{ L} \times 3 = 0.927 \text{ L}$). The total mass of oxidized lead and released lead over the entire 20-week experiment were then computed and compared to estimate the percentage of oxidized lead that was released as indicated in Table S7.

Table S7

Percent of Oxidized Lead in the Test Pieces Released into the Water and Stored as a Corrosion Scale Based on Faraday's Law

Test Piece	% of Oxidized Lead Released	% of Oxidized Lead Stored as a Corrosion Scale
TP1[pH-/DIC-/Hard-/NOM-]	4.0	96.0
TP2[pH+/DIC-/Hard-/NOM+]	19.9	80.1
TP3[pH-/DIC+/Hard-/NOM+]	25.0	75.0
TP4[pH+/DIC+/Hard-/NOM-]	19.3	80.7
TP5[pH-/DIC-/Hard+/NOM+]	40.7	59.3
TP6[pH+/DIC-/Hard+/NOM-]	12.6	87.4
TP7[pH-/DIC+/Hard+/NOM-]	23.3	76.7
TP8[pH+/DIC+/Hard+/NOM+]	2.9	97.1
TP9[pH0/DIC0/Hard0/NOM0]	8.2	91.8
TP10[pH0/DIC0/Hard0/NOM0]	4.8	95.2

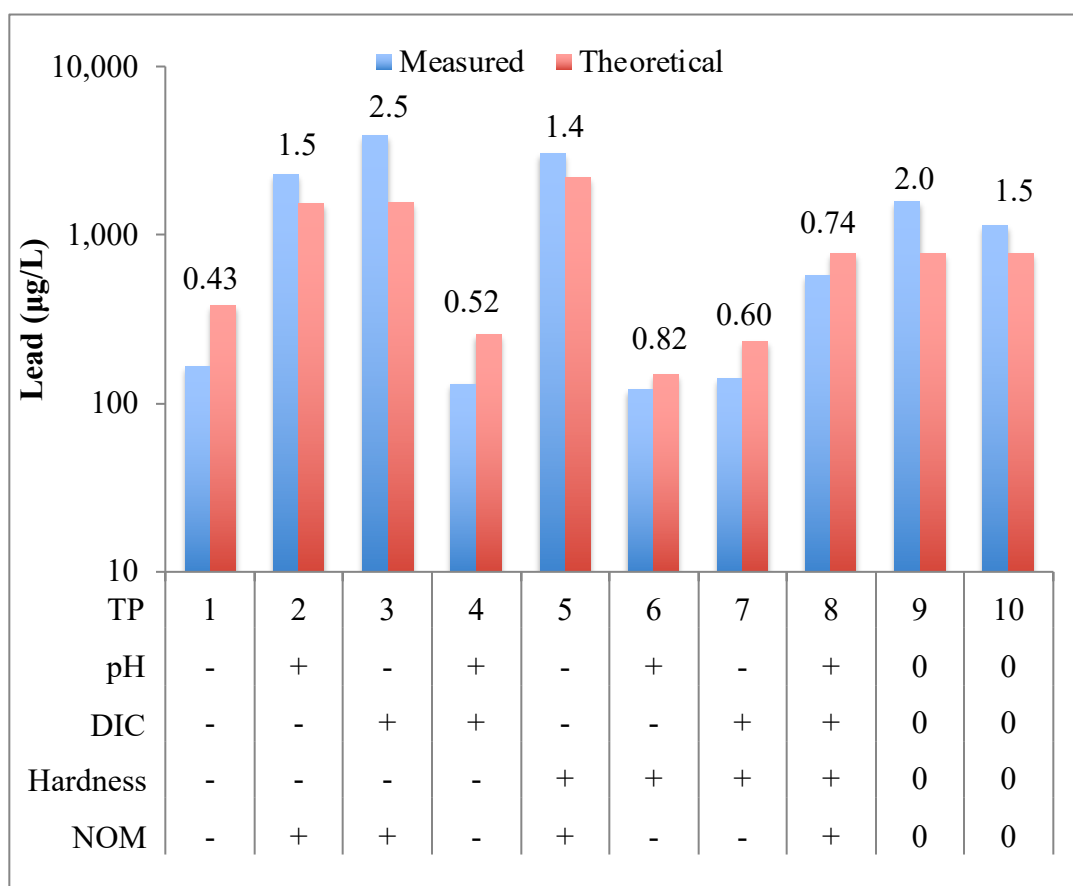


Figure S9. Comparison of average measured dissolved lead concentrations to theoretical lead solubility predicted by tidyphreeqc. Numbers on top of the bars represent the ratio of measured dissolved lead concentrations to theoretical solubility. Hydrocerussite was the solubility-controlling solid for TP2[pH+/DIC-/Hard-/NOM+] and TP6[pH+/DIC-/Hard+/NOM-], while cerussite was the solubility-controlling solid for the other synthetic waters.

Table S8

Measured Average Dissolved Lead Concentrations Versus Solubility Predicted by Tidyphreeqc

	Measured Dissolved Lead (µg/L)	Theoretical Lead Solubility (µg/L)*	Ratio (Measured:Dissolved)
TP1[pH-/DIC-/Hard-/NOM-]	166	383 (C)	0.43
TP2[pH+/DIC-/Hard-/NOM+]	2,310	1,540 (H)	1.5
TP3[pH-/DIC+/Hard-/NOM+]	3,890	1,554 (C)	2.5
TP4[pH+/DIC+/Hard-/NOM-]	132	256 (C)	0.52
TP5[pH-/DIC-/Hard+/NOM+]	3,050	2,221 (C)	1.4
TP6[pH+/DIC-/Hard+/NOM-]	122	148 (H)	0.82
TP7[pH-/DIC+/Hard+/NOM-]	140	233 (C)	0.60
TP8[pH+/DIC+/Hard+/NOM+]	578	785 (C)	0.74
TP9[pH0/DIC0/Hard0/NOM0]	1,580	779 (C)	2.0
TP10[pH0/DIC0/Hard0/NOM0]	1,140	779 (C)	1.5

*C indicates cerussite was predicted to form, H indicates hydrocerussite was predicted to form.

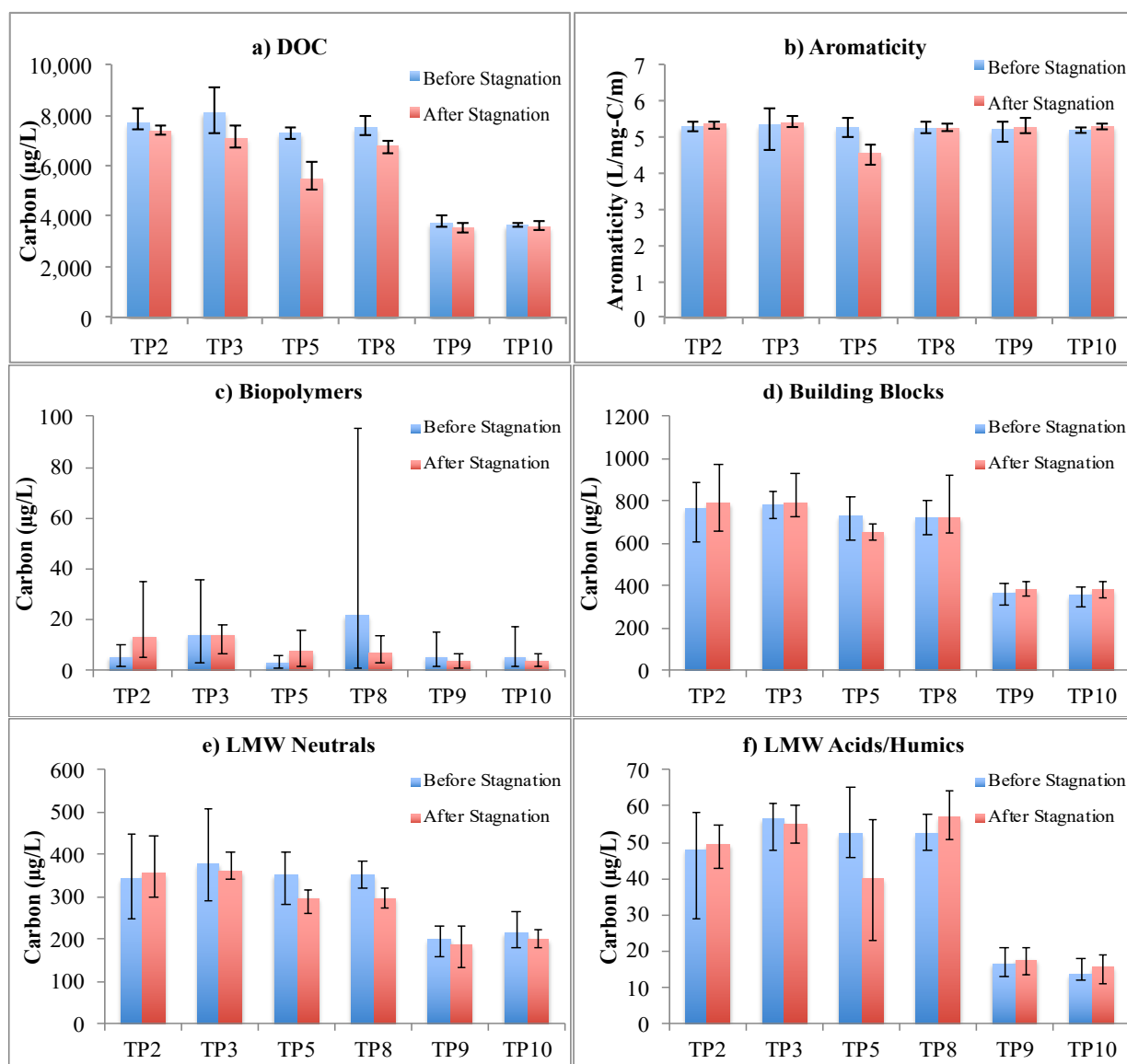


Figure S10. Average values for LC-OCD NOM fractions in weeks 3, 7, 12, 16 and 20 for a) DOC, b) aromaticity, c) biopolymers, d) building blocks, e) LMW neutrals, f) LMW acids/humics. Error bars represent minimum and maximum values. TP2[pH+/DIC-/Hard-/NOM+], TP3[pH-/DIC+/Hard-/NOM+], TP5[pH-/DIC-/Hard+/NOM+], and TP8[pH+/DIC+/Hard+/NOM+] were dosed with 7 mg DOC/L of SRNOM while TP9[pH0/DIC0/Hard0/NOM0] and TP10[pH0/DIC0/Hard0/NOM0] were dosed with 3.5 mg DOC/L of SRNOM.

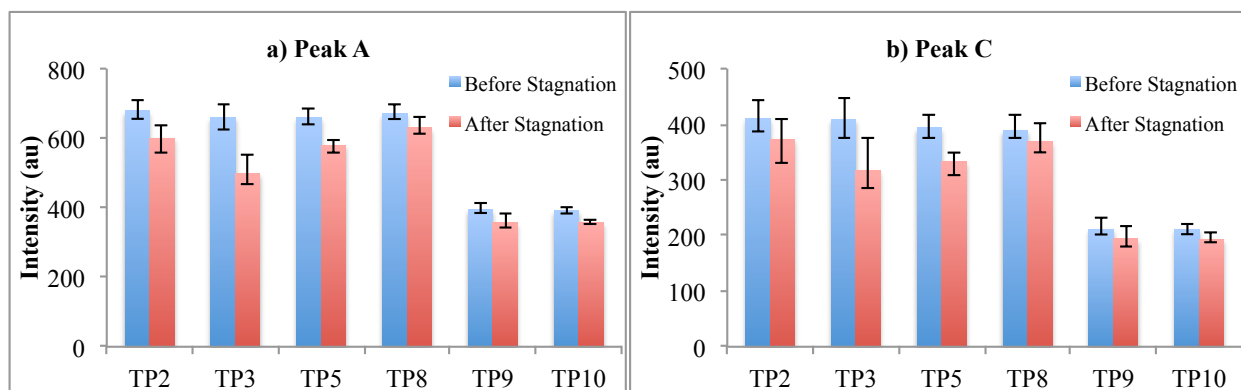


Figure S11. Average values for FEEM NOM humic-like peaks in weeks 12, 16, and 20 for a) peak A (Ex/Em = 250nm/461nm), and b) peak C (Ex/Em = 340nm/457nm). Error bars represent minimum and maximum values. TP2[pH+/DIC-/Hard-/NOM+], TP3[pH-/DIC+/Hard-/NOM+], TP5[pH-/DIC-/Hard+/NOM+], and TP8[pH+/DIC+/Hard+/NOM+] were dosed with 7 mg DOC/L of SRNOM while TP9[pH0/DIC0/Hard0/NOM0] and TP10[pH0/DIC0/Hard0/NOM0] were dosed with 3.5 mg DOC/L of SRNOM.

Table S9

Summary of Results from XRD Conducted on the Powdered Samples from the Lead Pipe Surface

	Cerussite (PbCO ₃)	Hydrocerussite (Pb ₃ (CO ₃) ₂ (OH) ₂)	Lead (Pb)	Litharge (PbO)
TP1[pH-/DIC-/Hard-/NOM-]	+		+	+
TP2[pH+/DIC-/Hard-/NOM+]			+	
TP3[pH-/DIC+/Hard-/NOM+]	+	+	+	+
TP4[pH+/DIC+/Hard-/NOM-]	+	+		
TP5[pH-/DIC-/Hard+/NOM+]	+	+	+	+
TP6[pH+/DIC-/Hard+/NOM-]	+	+	+	+
TP7[pH-/DIC+/Hard+/NOM-]	+	+	+	+
TP8[pH+/DIC+/Hard+/NOM+]	+		+	+
TP9[pH0/DIC0/Hard0/NOM0]	+		+	+
TP10[pH0/DIC0/Hard0/NOM0]	+	+	+	+

*‘+’ indicates the presence of certain mineral.

Table S10

Mass Percentage of Elements in the Lead Pipe Pit and Pit-Containing Scales Determined by Acid Digestion of Solids Followed by Analysis with ICP-MS

	Pb	Cu	Ca	Mg	Na	Zn
TP1[pH-/DIC-/Hard-/NOM-]	58.4	T*	T	.*	1.2	T
TP2[pH+/DIC-/Hard-/NOM+]	86.9	T	-	-	-	-
TP3[pH-/DIC+/Hard-/NOM+]	72.4	T	-	-	-	T
TP4[pH+/DIC+/Hard-/NOM-]	55.8	T	-	T	-	T
TP5[pH-/DIC-/Hard+/NOM+]	57.9	T	T	-	1.4	T
TP5[pH-/DIC-/Hard+/NOM+]	64.7	T	-	-	T	-
TP7[pH-/DIC+/Hard+/NOM-]	58.0	T	T	T	-	T
TP9[pH0/DIC0/Hard0/NOM0]	57.4	T	-	-	T	T
TP10[pH0/DIC0/Hard0/NOM0]	57.2	T	T	-	1.7	T
Scales on the side of connection						
TP1[pH-/DIC-/Hard-/NOM-]	48.1	T	T	-	1.3	T
TP3[pH-/DIC+/Hard-/NOM+]	76.5	T	T	-	-	-
TP5[pH-/DIC-/Hard+/NOM+]	59.5	T	T	-	1.8	T
TP6[pH+/DIC-/Hard+/NOM-]	78.8	T	-	-	-	-
TP7[pH-/DIC+/Hard+/NOM-]	65.6	T	-	-	T	-
TP8[pH+/DIC+/Hard+/NOM+]	35.5	T	T	-	1.3	T
TP9[pH0/DIC0/Hard0/NOM0]	29.3	0.1	1.9	T	T	-
TP10[pH0/DIC0/Hard0/NOM0]	49.5	T	T	-	2.0	T

*‘T’ is trace quantity (<0.1%) and ‘-’ is not detected

Table S11

Semi-Quantitative Analysis of Elements by Weight Percentage on the Scale Surface of the Lead Pipes Analyzed using SEM-EDS

	Pb	C	O	Si	Cl	Ca	Al	F	K	Na
TP1[pH-/DIC-/Hard-/NOM-]	70.8	24.6	4.3	0.2	0.0	0.1	0.1	0.0	0.0	0.0
TP2[pH+/DIC-/Hard-/NOM+]	69.1	27.5	2.2	0.4	0.6	0.0	0.0	0.2	0.0	0.0
TP3[pH-/DIC+/Hard-/NOM+]	85.2	7.7	7.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP4[pH+/DIC+/Hard-/NOM-]	54.6	35.3	9.1	0.4	0.0	0.2	0.1	0.0	0.2	0.2
TP5[pH-/DIC-/Hard+/NOM+]	82.2	10.5	7.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP6[pH+/DIC-/Hard+/NOM-]	67.7	22.8	9.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP7[pH-/DIC+/Hard+/NOM-]	72.1	21.1	6.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP8[pH+/DIC+/Hard+/NOM+]	84.8	7.9	7.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP9[pH0/DIC0/Hard0/NOM0]	75.8	17.3	7.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
TP10[pH0/DIC0/Hard0/NOM0]	75.2	18.7	6.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0

2.2. Scale Analysis for Copper Pipes

XRD results for the copper pipes of the test pieces are displayed in Figure S12 and Table S12.

The XRD results indicated that the copper solids cuprite (Cu_2O ; TP3, TP7, TP9), tenorite (CuO ; TP1, TP5, TP10), elemental copper (TP2, TP4, TP7, TP8), and malachite ($\text{Cu}_2(\text{CO}_3)(\text{OH})_2$; TP5, TP9) were present on the copper pipes. In addition, lead carbonate solids cerussite (PbCO_3 ; TP1, TP3, TP5, TP7, TP9, TP10) and hydrocerussite ($\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$; TP2, TP5, TP9) were present on many of the copper pipes. This can be attributed to dissolved lead precipitating on the copper pipes or particulate lead from the lead pipes being transported and incorporated into the corrosion scales on the copper pipes. Interestingly, the hardness scale calcite magnesian (CaO.MgO ; TP4, TP6-8, TP10) was detected on some of the copper pipes. More research is needed to determine if the hardness films that formed on the copper pipes can mitigate the

release of copper. The ICP-MS analysis on the copper corrosion scales (Table S13) indicated that the corrosion scales on the copper pipes contained large amounts of copper, calcium, and lead with lesser amounts of magnesium, sodium and zinc.

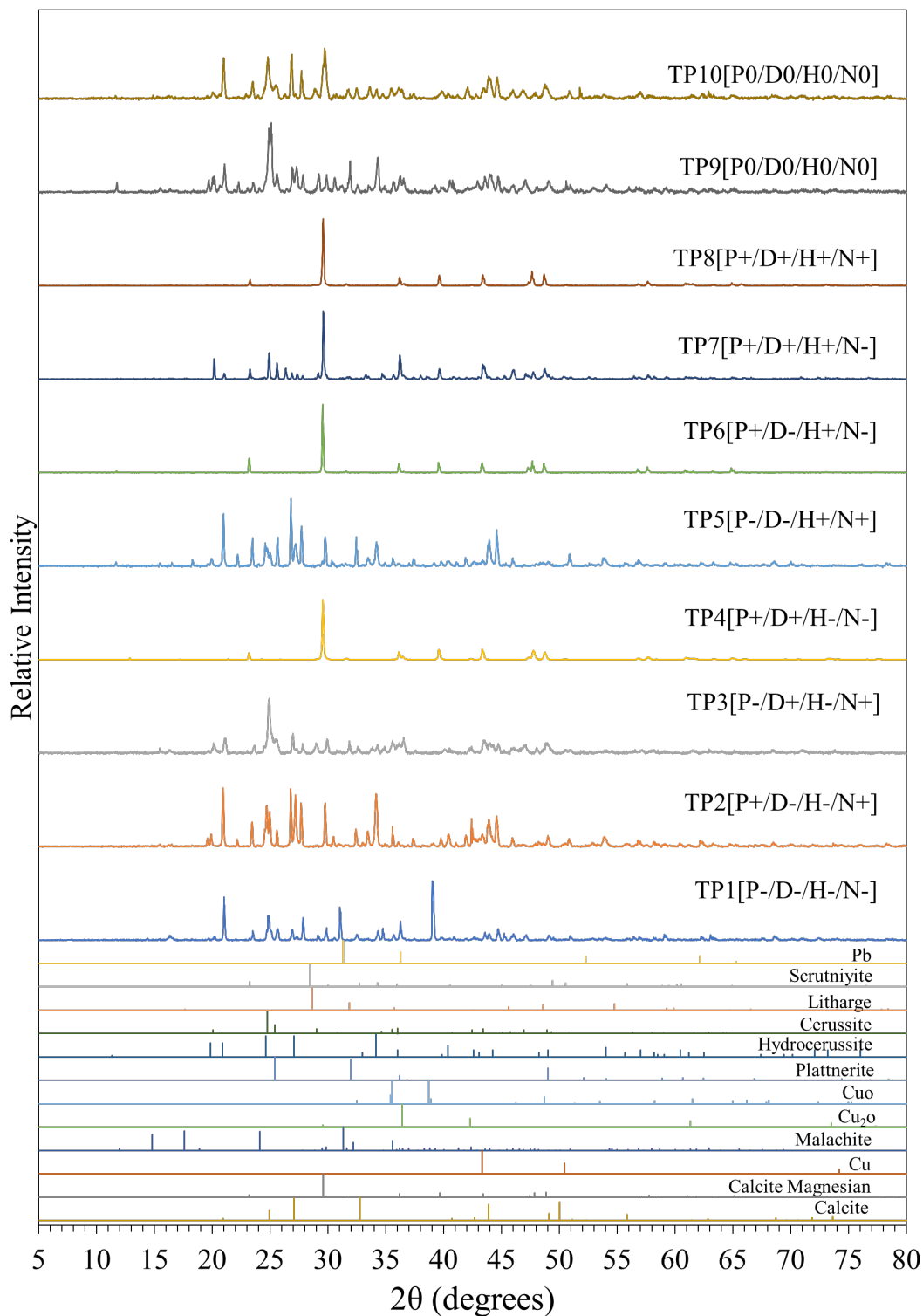


Figure S12. XRD patterns obtained from the surface of the copper pipes for a range of 5° to 80° 2θ. The patterns at the bottom are the reference patterns of the solids that had peaks identified in the samples.

Table S12

Summary of Results from XRD Conducted on the Powdered Samples from the Copper Pipe Surface

	Cerussite (PbCO_3)	Magnesian Calcite (Ca,MgCO_3)	Cuprite (Cu_2O)	Tenorite (CuO)	Copper (Cu)	Malachite ($\text{Cu}_2(\text{CO}_3)(\text{OH})_2$)
TP1[pH-/DIC-/Hard-/NOM-]	++ ¹			+ ¹		
TP2[pH+/DIC-/Hard-/NOM+]	HC ²				++	
TP3[pH-/DIC+/Hard-/NOM+]	++		++			
TP4[pH+/DIC+/Hard-/NOM-]		++			++	
TP5[pH-/DIC-/Hard+/NOM+]	++ HC			++		++
TP6[pH+/DIC-/Hard+/NOM-]		++				
TP7[pH-/DIC+/Hard+/NOM-]	++	++	++		+	
TP8[pH+/DIC+/Hard+/NOM+]		++			+	
TP9[pH0/DIC0/Hard0/NOM0]	++ HC		++			++
TP10[pH0/DIC0/Hard0/NOM0]	++	++		+		

¹‘+’ indicates the presence of the mineral at low intensity, ‘++’ indicates the presence of the mineral at moderate intensity.

²HC stands for hydrocerussite, it is present in minor quantities.

Table S13

Mass Percentage of Elements in the Copper Pipe Pit and Pit-Containing Scales Determined by Acid Digestion of Solids Followed by Analysis with ICP-MS

	Cu	Ca	Pb	Mg	Na	Zn
TP1[pH-/DIC-/Hard-/NOM-]	14.3	-*	6.2	T*	0.1	T
TP2[pH+/DIC-/Hard-/NOM+]	4.2	T	0.5	T	0.1	T
TP3[pH-/DIC+/Hard-/NOM+]	27.8	T	5.7	T	0.1	T
TP4[pH+/DIC+/Hard-/NOM-]	29.8	7.4	0.3	0.1	T	0.1
TP5[pH-/DIC-/Hard+/NOM+]	25.2	37.5	0.2	0.3	4.8	T
TP6[pH+/DIC-/Hard+/NOM-]	51.0	8.3	5.6	0.1	T	T
TP7[pH-/DIC+/Hard+/NOM-]	50.0	18.5	0.4	0.1	0.1	T
TP8[pH+/DIC+/Hard+/NOM+]	29.5	0.6	6.4	0.1	-	T
TP9[pH0/DIC0/Hard0/NOM0]	8.1	0.8	6.4	T	0.4	T
TP10[pH0/DIC0/Hard0/NOM0]	T	T	7.0	-	-	-

*‘T’ is trace quantity (<0.01%) and ‘-’ is not detected.

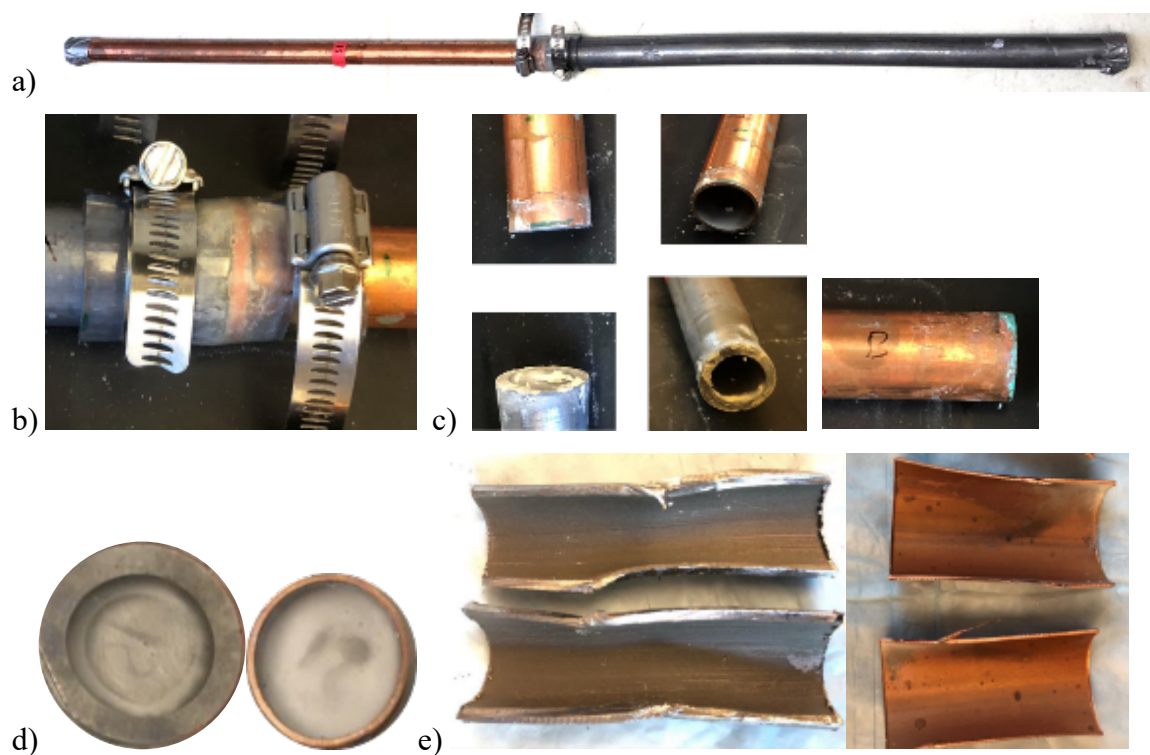


Figure S13. Example photographs of scales on a test piece exposed to synthetic water without SRNOM (TP1[pH-/DIC-/Hard-/NOM-]): a) entire length of the lead and copper pipes at end of the experiment; b) magnified photograph of the lead and copper connection; c) deposits on the ends of the lead and copper pipes near the connection and on the far end of the copper pipe; d) cross-sections of the lead and copper pipes embedded in epoxy; e) cross-sections of the lead and copper pipes.



Figure S14. Example photographs of scales on a test piece exposed to synthetic water with SRNOM (TP8[pH+/DIC+/Hard+/NOM+]): a) entire length of the lead and copper pipes at end of the experiment; b) magnified photograph of the lead and copper connection; c) deposits on the ends of the lead and copper pipes near the connection and on the far end of the copper pipe; d) cross-sections of the lead and copper pipes embedded in epoxy; e) cross-sections of the lead and copper pipes.

References

- 1 J. Parks, M. Edwards and A. Atassi, *Non-Intrusive Methodology for Assessing Lead and Copper Corrosion* [PDF]. Am. Water Works Assoc. Res. Foundation, Denver, CO, 2014.
- 2 National Sanitation Foundation and American National Standards Institute, *NSF/ANSI Standard 61 – 2016 Drinking Water System Components – Health Effects* [PDF]. The NSF Joint Committee on Drinking Water Additives. Ann Arbor, MI, 2016.
- 3 City of Guelph, *Annual and Summary Report 2017* [PDF]. Water Services, Environmental Services Department, Guelph, ON, 2018.
- 4 City of Ottawa, *Lemieux Water Purification Plant - 2017 Drinking Water Quality* [PDF]. Ottawa, ON, 2018.
- 5 City of Toronto, *Drinking Water Analysis Summary 2017* [PDF]. Toronto, ON, 2018.
- 6 City of Waterloo, *2017 Annual Report* [PDF]. Waterloo, ON, 2018.
- 7 International Humic Substances Society, *Elemental Compositions and Stable Isotopic Ratios of IHSS Samples*. Retrieved January 11, 2020, from <http://humic-substances.org/elemental-compositions-and-stable-isotopic-ratios-of-ihss-samples/>
- 8 E. O. Zhou, S. J. Payne, R. O. Hofmann and R. C. Andrews, Factors Affecting Lead Release in Sodium Silicate-Treated Partial Lead Service Line Replacements, *J. Environ. Sci. Health A*, 2015, **50**(9), 922–930. doi: 10.1080/10934529.2015.1030283
- 9 K. Aghasadeghi, S. Peldszus, B. F. Trueman, A. Mishrra, M. G. Cooke, R. M. Slawson, D. E. Giammar, G. A. Gagnon and P. M. Huck, Pilot-Scale Comparison of Sodium Silicates, Orthophosphate, and pH Adjustment to Reduce Lead Release from Lead Service Lines, *Water Res.*, 2021, 195, Article 116955, 1-11. doi: 10.1016/j.watres.2021.116955
- 10 A. Mishrra, Z. Wang, V. Sidorkiewicz and D. E. Giammar, Effect of Sodium Silicate on Lead Release from Lead Service Lines, *Water Res.*, 2021, **188**, 116485. doi: 10.1016/j.watres.2020.116485
- 11 D. C. Montgomery, *Design and Analysis of Experiments*. John Wiley & Sons Inc., Hoboken, NJ, 2013.
- 12 D. Dunnington, *Tidyphreeqc: Tidy Geochemical Modeling Using Phreeqc*. 2019. Retrieved on February 13, 2020, from <https://github.com/paleolimbot/tidyphreeqc>.
- 13 D. L. Parkhurst and C. A. J. Appelo, *Description of Input and Examples for Phreeqc Version 3—a Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations*. Techniques and Methods. U.S. Geological Survey vol. 6, 2013. Retrieved on February 13, 2020, from <http://pubs.usgs.gov/tm/06/a43>
- 14 B. F. Trueman, *pbculsol: Predict Lead and Copper Solubility*. 2021. Retrieved on October 22, 2021, from <https://github.com/bentrueman/pbcusol>.
- 15 S. Garnier, *Viridis: Default Color Maps from 'Matplotlib'*. 2018. Retrieved on February 13, 2020, from <https://CRAN.R-project.org/package=viridis>.

- 16 H. Wickham, *Tidyverse: Easily Install and Load the 'Tidyverse'*. 2017. Retrieved on February 13, 2020, from <https://CRAN.R-project.org/package=tidyverse>.
- 17 H. Wickham and J. Bryan, *Readxl: Read Excel Files*. 2019. Retrieved on February 13, 2020, from <https://CRAN.R-project.org/package=readxl>.
- 18 M. R. Schock, I. Wagner and R. J. Oliphant, *Corrosion and Solubility of Lead in Drinking Water*. In *Internal Corrosion of Water Distribution Systems*, 2nd ed., 131–230. Am. Water Works Assoc. Res. Foundation, Denver, CO, 1996.