Supplemental information for effectiveness of sodium silicates for lead corrosion control: a critical review of current data

Bofu Li*, Benjamin F. Trueman, Evelyne Doré and Graham A. Gagnon

Centre for Water Resources Studies, Department of Civil & Resource Engineering, Dalhousie University, 1360 Barrington St., Halifax, Nova Scotia, Canada B3H 4R2

*Corresponding author:
E-mail: bofu.li@dal.ca
Tel: 902.494.6070
Fax: 902.494.3105

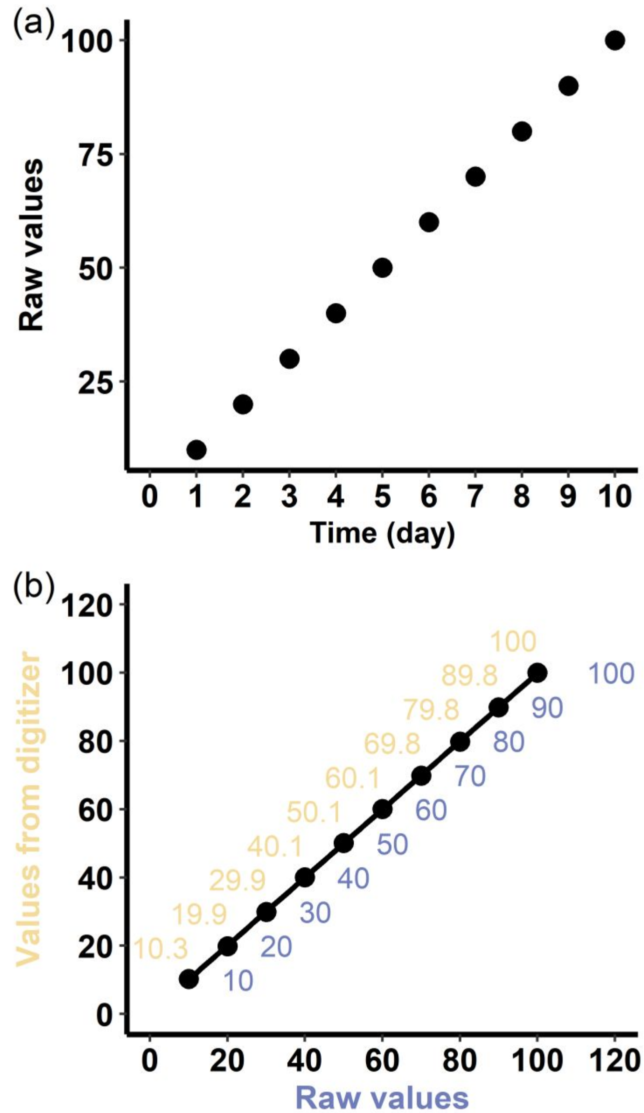

Figure S1. a) Simulated data and b) comparison of the data and values returned by the digitizer (WebPlotDigitizer, version 4.4);[1] R^2 was 1.

Table S1. Raw data for Figure 1 (units are consistent with the corresponding paper).[2,3]

author	pH	Pb (μg/L)	SiO2 (mg/L)
Lintereur et al. (2010)	8.1	1.0	12.8
Lintereur et al. (2010)	8.2	1.4	16.5
Lintereur et al. (2010)	8.4	1.0	21.6
Lintereur et al. (2010)	7.8	8.3	10.2
Lintereur et al. (2010)	8.0	2.4	10.1
Lintereur et al. (2010)	8.0	4.1	7.8
Lintereur et al. (2010)	8.2	1.1	11.7
Lintereur et al. (2010)	8.4	1.0	17.2
Lintereur et al. (2010)	7.9	11.6	4.9
Lintereur et al. (2010)	7.9	8.4	5.2
Lintereur et al. (2010)	8.2	10.1	13.3
Lintereur et al. (2010)	8.3	1.4	17.0
Lintereur et al. (2010)	8.4	1.1	22.6
Lintereur et al. (2010)	7.7	32.1	10.2
Lintereur et al. (2010)	8.1	10.2	10.5
Lintereur et al. (2010)	8.0	8.9	9.2
Lintereur et al. (2010)	8.2	1.2	13.1
Lintereur et al. (2010)	8.4	1.1	17.7
Lintereur et al. (2010)	7.6	4.3	6.3
Lintereur et al. (2010)	7.9	3.4	6.3
Schock et al. (2005)	6.0	34.1	0.0
Schock et al. (2005)	6.2	37.3	0.0
Schock et al. (2005)	6.0	31.0	0.0
Schock et al. (2005)	6.7	31.1	30.0
Schock et al. (2005)	7.2	38.4	30.0
Schock et al. (2005)	7.3	23.2	30.0
Schock et al. (2005)	7.1	21.3	30.0
Schock et al. (2005)	6.9	9.8	30.0

Schock et al. (2005)	7.0	11.0	30.0
Schock et al. (2005)	7.2	20.1	55.0
Schock et al. (2005)	7.6	8.0	55.0
Schock et al. (2005)	7.7	5.8	55.0
Schock et al. (2005)	7.6	8.7	55.0

Table S2. Percent lead in scale analysis reported in literature. [4–9]

author	year	tool	% Pb before treatment	% Pb after treatment	unit	position
CDM Smith (2019)	2019	XRF	Not reported	<0.5-30.15	% weight	cross-section
Mishrra et al. (2020)	2020	SEM-EDS	0.3-0.5	1-2	% weight	cross-section
Aghasadeghi et al. (2021)	2021	SEM-EDS	Not reported	2.2	% atomic	cross-section
Li et al. (2021a) ref [7]	2021	XPS	Not reported	5.7	% atomic	inner-surface
Li et al. (2021b) ref [9]	2021	SEM-EDS	Not reported	3.4	% weight	inner-surface
Li et al. (2021c) ref [8]	2021	SEM-EDS	Not reported	0.42- 0.58	% weight	inner-surface

Table S3. Summary of thermodynamic data used in equilibrium solubility modeling.[10,11]

Phase	Equation	log K
.	$Pb^{+2} + H_2O = PbOH^{+} + H^{+}$	-7.22
.	$Pb^{+2} + 3H_2O = Pb(OH)_3^{-} + 3H^{+}$	-28.08
.	$Pb^{+2} + 4H_2O = Pb(OH)_4^{-2} + 4H^{+}$	-39.72
.	$Pb^{+2} + 2H_2O = Pb(OH)_2 + 2H^{+}$	-16.91
.	$2Pb^{+2} + H_2O = Pb_2OH^{+3} + H^{+}$	-6.36
.	$3Pb^{+2} + 4H_2O = Pb_3(OH)_4^{+2} + 4H^{+}$	-23.86
.	$4Pb^{+2} + 4H_2O = Pb_4(OH)_4^{+4} + 4H^{+}$	-20.88
.	$6Pb^{+2} + 8H_2O = Pb_6(OH)_8^{+4} + 8H^{+}$	-43.62
.	$Pb^{+2} + CO_3^{-2} = PbCO_3$	7.10
.	$Pb^{+2} + 2CO_3^{-2} = Pb(CO_3)_2^{-2}$	10.33
.	$Pb^{+2} + CO_3^{-2} + H^{+} = PbHCO_3^{+}$	12.59
.	$Pb^{+2} + PO_4^{-3} + H^{+} = PbHPO_4$	15.41
.	$Pb^{+2} + PO_4^{-3} + 2H^{+} = PbH_2PO_4^{+}$	21.05
.	$Pb^{+2} + SO_4^{-2} = PbSO_4$	2.73
.	$Pb^{+2} + 2SO_4^{-2} = Pb(SO_4)_2^{-2}$	3.50
.	$Pb^{+2} + Cl^{-} = PbCl^{+}$	1.59
.	$Pb^{+2} + 2Cl^{-} = PbCl_2$	1.80
.	$Pb^{+2} + 3Cl^{-} = PbCl_3^{-}$	1.71
.	$Pb^{+2} + 4Cl^{-} = PbCl_4^{-2}$	1.43
Cerussite	$PbCO_3 = Pb^{+2} + CO_3^{-2}$	-13.11
Hydrocerussite	$Pb_3(CO_3)_2(OH)_2 + 2H^{+} = 3Pb^{+2} + 2CO_3^{-2} + 2H_2O$	-18.00
Hydroxylpyromorphite	$Pb_5(PO_4)_3OH + H^{+} = 5Pb^{+2} + 3PO_4^{-3} + H_2O$	-62.83
PbSiO3	$PbSiO_3 + H_2O + 2H^{+} = Pb^{+2} + H_4SiO_4$	7.32

Table S4. Lead concentration/lead release per unit surface area used to calculate the ratios in Figure 4 (rows represent comparable water quality conditions; units are consistent with the corresponding paper).[4–9,12–14]

reference	silicate treated	control	phosphate	silicate treated filtered	control filtered	phosphate filtered
Kogo et al. (2017)	18.0	21.0	6.0	14.0	18.0	4.0
Kogo et al. (2017)	50.0	57.0	11.0	45.0	54.0	7.0
Kogo et al. (2017)	96.0	110.0	15.0	83.0	101.0	9.0
Kogo et al. (2017)	19.0	16.0	3.0	17.0	14.0	2.0
Kogo et al. (2017)	55.0	51.0	8.0	53.0	47.0	6.0
Kogo et al. (2017)	109.0	103.0	14.0	98.0	96.0	11.0
Kogo et al. (2017)	14.0	19.0	4.0	11.0	15.0	3.0
Kogo et al. (2017)	37.0	47.0	10.0	33.0	40.0	8.0
Kogo et al. (2017)	63.0	102.0	22.0	54.0	90.0	15.0
Kogo et al. (2017)	23.0	19.0	4.0	10.0	11.0	2.0
Kogo et al. (2017)	46.0	46.0	11.0	23.0	27.0	5.0
Kogo et al. (2017)	65.0	62.0	39.0	42.0	51.0	29.0
Pinto et al. (1997)	2538.0	1145.0	NA	NA	NA	NA
Pinto et al. (1997)	1086.0	876.0	NA	NA	NA	NA
Pinto et al. (1997)	1345.0	611.0	NA	NA	NA	NA
Pinto et al. (1997)	1470.0	470.0	NA	NA	NA	NA
Pinto et al. (1997)	1080.0	1350.0	NA	NA	NA	NA
Pinto et al. (1997)	1480.0	1105.0	NA	NA	NA	NA
Woszczynski et al. (2015)	46.0	NA	12.0	33.1	NA	6.1
Woszczynski et al. (2015)	6.7	NA	2.0	5.6	NA	1.2
Mishrra et al. (2021)	17.8	37.1	NA	9.6	23.7	NA
Mishrra et al. (2021)	17.3	35.5	NA	14.4	21.7	NA
Mishrra et al. (2021)	23.3	23.1	NA	13.0	17.3	NA
Aghasadeghi et al. (2021)	85.0	53.0	20.0	77.0	45.0	17.0
Aghasadeghi	102.0	65.0	23.0	87.0	59.0	16.0

reference	silicate treated	control	phosphate	silicate treated filtered	control filtered	phosphate filtered
et al. (2021)						
Aghasadeghi et al. (2021)	143.0	92.0	18.0	121.0	85.0	17.0
Aghasadeghi et al. (2021)	155.0	77.0	20.0	146.0	69.0	16.0
Aghasadeghi et al. (2021)	193.0	53.0	20.0	176.0	45.0	17.0
Aghasadeghi et al. (2021)	193.0	65.0	23.0	176.0	59.0	16.0
Aghasadeghi et al. (2021)	158.0	92.0	18.0	153.0	85.0	17.0
Aghasadeghi et al. (2021)	158.0	77.0	20.0	153.0	69.0	16.0
Li et al. (2021b) ref [9]	398.0	NA	67.0	NA	NA	NA
Li et al. (2021b)	578.0	NA	69.0	NA	NA	NA
Li et al. (2021c) ref [8]	108.0	90.5	5.5	64.6	57.2	0.2
Li et al. (2021c)	92.4	73.7	17.2	41.5	31.6	0.9
Li et al. (2021c)	73.3	54.7	19.9	63.9	40.0	5.4
Li et al. (2021c)	66.7	45.6	36.9	56.5	36.1	22.5
Li et al. (2021c)	358.9	90.5	5.5	96.1	57.2	0.2
Li et al. (2021c)	36.5	73.7	17.2	25.2	31.6	0.9
Li et al. (2021c)	49.8	54.7	19.9	40.9	40.0	5.4
Li et al. (2021c)	42.0	45.6	36.9	38.4	36.1	22.5
Li et al. (2021c)	2656.8	2004.6	218.9	251.9	105.0	0.4
Li et al. (2021c)	599.8	440.2	594.2	57.9	84.5	3.4
Li et al. (2021c)	804.1	1431.0	293.0	103.6	72.8	7.6
Li et al. (2021c)	613.5	250.3	160.2	65.4	47.2	23.4
Li et al. (2021c)	10457.2	2004.6	218.9	157.0	105.0	0.4
Li et al. (2021c)	2247.9	440.2	594.2	114.7	84.5	3.4
Li et al. (2021c)	2363.2	1431.0	293.0	97.0	72.8	7.6
Li et al. (2021c)	5392.5	250.3	160.2	42.3	47.2	23.4

reference	silicate treated	control	phosphate	silicate treated filtered	control filtered	phosphate filtered
Li et al. (2021a) ref [7]	78.5	85.3	3.0	66.5	52.7	1.7
Li et al. (2021a)	52.6	29.2	7.9	42.6	26.0	5.4
Li et al. (2021a)	47.0	38.7	26.1	32.7	32.5	15.2
Li et al. (2021a)	82.5	104.2	64.1	60.0	91.5	57.5

R code to reproduce equilibrium lead solubility model

```r
library("tidyverse")
library("furrr") # speed things up with parallel iteration
# remotes::install_github("bentrueman/pbcusol")
library("pbcusol")

# make sure things keep running even if phreeqc throws an error:

possibly_pb_sol <- possibly(
  eq_sol_fixed,
  otherwise = tibble(pb_ppb = NA_real_, phase = NA_real_)
)

future::plan("multisession")

out <- crossing(
  # modify the `by` argument to increase the resolution (we use 0.1)
  ph = seq(6, 11, by = 1),
  dic = c(7, 114),
  phase = c("Cerussite", "Hydcerussite", "PbSiO3", "Hxypyromorphite")
) %>%
  mutate(po4 = if_else(phase == "Hxypyromorphite", 1, 0)) %>%
  rowid_to_column() %>%
  group_by(rowid) %>%
  nest() %>%
  ungroup() %>%
  mutate(
    output = furrr::future_map(
      data,
```

```
    ~ possibly_pb_sol(
      ph = .x$ph, dic = .x$dic, phosphate = .x$po4,
      phase = .x$phase, buffer = "HCl",
      Na = .5, Cl = .5,
      element = "Pb"
    ) %>%
      select(-phase)
  )
) %>%
unnest(c(data, output))
```

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