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Impact Area Groundwater Study Program

Joint Base Cape Cod Small-Arms Ranges

Lead and Antimony Batch-Reaction Study

Amanda J. Barker, Samuel A. Beal, and Jay L. Clausen

September 2019



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Abstract

The results and documentation in this report summarize work supporting the Joint Base Cape Cod Camp Edwards Small-Arms Ranges (JBCC SARs) assessment of the potential for lead (Pb) and antimony (Sb) to migrate in soil and soil solutions. Batch studies were performed in an effort to interpret field results for metal concentrations at Camp Edwards SARs and to determine potential remediation strategies that limit both Pb and Sb.

The batch study showed phosphate and iron treatments had minimal to no impact on Pb and Sb dissolution. However, the lime treatment significantly decreased the dissolution of Sb while increasing the dissolution of Pb. Dissolved metal concentrations decreased over the course of the 48-hour experiment regardless of treatment.

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Preface

This study was conducted for the Massachusetts Army National Guard (MAARNG) under the Impact Area Groundwater Study Program. It was funded via Labor Charge Code. The technical monitor was Dr. Michael Ciaranca, MAARNG.

The work was performed by the Biogeochemical Sciences Branch (CEERD-RRN) of the Research and Engineering Division (CEERD-RR), U.S. Army Engineer Research and Development Center, Cold Regions Research and Engineering Laboratory (ERDC-CRREL). At the time publication, Dr. Justin Berman was Chief, CEERD-RRN; and Mr. Jared Oren was Acting Chief, CEERD-RR. The Deputy Director of ERDC-CRREL was Mr. David B. Ringelberg, and the Director was Dr. Joseph L. Corriveau.

COL Teresa A. Schlosser was Commander of ERDC, and Dr. David W. Pittman was the Director.

Acronyms and Abbreviations

$\alpha\text{-Fe(OH)}_3$	Iron (III) Hydroxide Alpha-Phase
Ca(OH)_2	Calcium Hydroxide
$\text{Ca}_3(\text{PO}_4)_2$	Calcium Phosphate Tribasic
CRREL	Cold Regions Research and Engineering Laboratory
DOC	Dissolved Organic Carbon
EMC	Environmental Management Commission
ERDC	U.S. Army Engineer Research and Development Center
JBCC	Joint Base Cape Cod
LOI ₃₇₅	Loss on Ignition at 375°C
MAARNG	Massachusetts Army National Guard
MMR	Massachusetts Military Reservation
OMMP	Operation, Maintenance, and Monitoring Plan
Pb	Lead
$\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$	Hydrocerussite
PbCO_3	Cerussite
PTFE	Polytetrafluoroethylene
SAR	Small-Arms Range
Sb	Antimony
TOC	Total Organic Carbon
USEPA	U.S. Environmental Protection Agency

1 Introduction

1.1 Background

Mobilization of toxic metals from small-arms ranges (SARs) on Joint Base Cape Cod (JBCC) is a concern of the Massachusetts Army National Guard. The metals lead (Pb) and antimony (Sb), which make up the bulk of conventional bullets, are of particular concern due to their potential toxicity and mobility into groundwater. Many environmental monitoring studies have been conducted at Camp Edwards, located on JBCC, including analyzing for metal(loid) concentrations that have been reported in the soil and groundwater at multiple SARs (particularly the Juliet, Kilo, and Tango Ranges) from 2010 to 2016. Additionally, near-surface waters are monitored through sampling via lysimeters located near target berms; and best management practices have been implemented on-site with the installation of STAPP bullet catch systems. A number of remediation efforts have occurred at Camp Edwards, including the addition of phosphate (Maec-tite) to soils in 1998, removal of contaminated soils and regrading surface soils in 2008, and the addition of lime to soils in 2011.

Both Pb and Sb are sensitive to oxidation-reduction (redox) conditions and changes in soil chemistry and are known to form different metal species that control their mobility in soil (Johnson et al. 2005; Clausen et al. 2011). Studies have shown that pH, redox environment, presence of organic matter and iron/manganese oxides in soils, and speciation significantly affect the overall solubility and mobility of Pb and Sb (Vantelon et al. 2005; Scheinost et al. 2006; Ackermann et al. 2009; Okkenhaug et al. 2016). Understanding the behavior of Pb and Sb in soil and soil solution as a function of site parameters and soil remediation is integral to assessing the overall fate and transport of Pb and Sb on-site and estimating the potential for off-site migration and infiltration into groundwater.

Prior to this study, there was no record at Camp Edwards of Sb ever exceeding the Operation, Maintenance, and Monitoring Plan (OMMP) Action Levels of 6 ppb established by the Commonwealth of Massachusetts, Environmental Management commission (EMC) for JBCC. The U.S. Environmental Protection Agency (USEPA) ceded oversight authority of the

SARs at JBCC to the EMC with prescribed monitoring. JBCC wants to develop a scientific-supported position for long-term environmental monitoring of the SARs to prevent environmental degradation.

The USEPA issued an Administrative Order in 1997 ceasing weapons training with leaded ammunition at the Massachusetts Military Reservation (MMR), the predecessor to JBCC. In 2009, MMR petitioned the USEPA to allow for the resumption of training with leaded ammunition. The petition consisted of MMR site-specific data and an understanding of the fate and transport of Pb based on an exhaustive literature review, which indicated Pb transport from MMR SARs would be minimal and have a negligible impact on the environment. At that time, the Massachusetts Department of Environmental Protection and EMC concurred with MMR's findings but required a monitoring program for the SARs. In 2017, the USEPA ceded the oversight responsibility of JBCC SARs to the EMC based on a new set of findings of fact.

During the years we studied (2010–2016), the Best Management Practices and OMMP Action Levels of Sb in the soil pore-water samples remained below 6 ppb until 2012, 2013, and 2016 when the Sb levels at the Juliet, Kilo, and Tango Ranges increased, respectively. In 2014 and 2015, only the Juliet Range and Kilo Range soil pore-water samples has Sb exceeding the OMMP Action Levels. The recent increase in Sb concentrations in lysimeters from samples collected at the Juliet, Kilo, and Tango Ranges occurred the year following the lime amendment application, which raised the question: Was this the cause of the Sb increase? In March 2010 and 2015, Pb exceeded the OMMP Action Levels set for groundwater at the Tango Range.

Juliet, Kilo, and Tango Ranges are 25 m SARs used for marksmanship training, and each range has a STAPP bullet containment system installed. The three ranges are undergoing a pilot period to determine if it is possible to use Pb bullets on the ranges while protecting the soils and groundwater from contamination. The ranges are annually assessed for Pb, Sb, and copper contamination in soils, soil pore water, and groundwater.

Juliet, Kilo, and Tango Ranges were established as SARs at Camp Edwards in the 1980s. During 2006–2008, the Impact Area Groundwater Study Program conducted an environmental impact assessment and reported elevated concentrations of Pb in soils but not in groundwater for all three ranges. In 2008 at the Juliet and Kilo Ranges, the MAARNG removed all

surface soils, regraded the ranges, and installed a STAPP systems. In 2009, the OMMP pilot period commenced for Juliet and Kilo Ranges. In 2010, Juliet and Kilo Ranges were regraded to improve drainage, and pan lysimeters were installed to monitor metals and metalloids in pore water.

In 2006, the STAPP system was installed at the Tango Range. MAARNG regraded Tango Range surface soils in 2007 at the mounded firing line to improve the firing angle to the STAPP; the area between the firing line and the impact berm were not regraded. In 2008, the OMMP pilot period commenced for Tango Range. In 2010, pan lysimeters were installed to monitor heavy metals and metalloids in pore water.

Prior to the STAPP installation, the firing berms for these ranges were bare ground, making their soils and groundwater highly susceptible to Pb and Sb contamination. Lead phosphates are the most stable form of Pb in the environment, and they form rapidly when phosphate amendments are added to Pb-contaminated soils. Therefore, the MAARNG applied phosphate amendments to these ranges in 1998 to reduce groundwater contamination. From the Impact Area Groundwater Study Program reports from 2006–2008, Pb and Sb were not found in groundwater samples, indicating that the 1998 phosphate additions may have been successful at immobilizing the Pb. In 2011, the MAARNG sought to immobilize Pb in range soils by raising the pH to about 8 using a lime amendment. The lime successfully increased soil pH to 7.6–8.7, and Pb concentrations were moderately reduced in pore water for the Juliet and Tango Ranges.

The effectiveness of liming to immobilize Sb varies across soils types and condition. Liming soils with high sand content, similar to the ones found at Camp Edwards, can increase the concentration of Sb in pore water of soils. Hale et al. (2012) found that Sb was least soluble around pH 5 and most soluble around pH 7–8. The soils of the Juliet, Kilo, and Tango Ranges naturally range from 5.9 to 6.6; and in 2010, prior to the lime amendment, Sb in pore water was below the OMMP action levels. In 2012, after the lime amendment and the increase in soil pH, Juliet and Kilo Ranges concurrently had Sb concentration above OMMP pore-water action levels in lysimeters located near the STAPP firing berm while Sb concentration in the soil samples remained low. Tango Range began showing the same trend in Sb around 2014. Because of the increased Sb levels, the MAARNG ceased adding lime amendments to range soils in 2012; however, Sb levels continued to increase until 2015. In 2016, Sb levels in pore

water decreased but remained above the OMMP action level. In August 2015, Pb in the groundwater for the Tango Range spiked well above the action level. Groundwater was resampled in October 2015, and Pb levels were below the OMMP action level. The Camp Edwards Environmental and Readiness Center attributed this spike in Pb concentration to lower water levels yielding a turbid, sediment laden sample (Environmental and Readiness Center, pers. comm. with J. Clausen, 2019).

The U.S. Army Engineer Research and Development Center's Cold Regions Research and Engineering Laboratory (ERDC-CRREL) proposed a series of experiments to understand the mobility potential of Pb and Sb on JBCC SARs in response elevated Sb levels and the application of different soil amendments. This report discusses batch-reaction experiment results and the collection of supplemental field data from the JBCC SARs.

1.2 Objectives

We conducted batch studies to investigate why Sb initially mobilized in the summer of 2012 at the Juliet, Kilo, and Tango Ranges at JBCC and continues to be elevated. The objective of the batch studies is to better understand the mechanisms responsible for the release of Sb from soil in the soil pore water at several small-arms ranges. In addition, we explored materials to immobilize Sb.

1.3 Approach

To understanding the mobilization of Sb at JBCC SARs, we obtained site soils and water to conduct a series of batch experiments. The following sections detail the experimental approach.

2 Methods

2.1 Sample collection

ERDC-CRREL collected samples from JBCC SARs on 13 August 2018. Soil samples were collected using the CRREL Multi-Increment Sampling Tool, which consists of a stainless steel 3 cm diameter cylindrical tip set to a depth of 3 cm. Each soil sample was composed of approximately 100 increments collected in a systematic-random pattern across the berm face and range floor using the Incremental Sampling Methodology for each range. Soil samples were collected from five locations (Old Kilo Range Berm, Juliet Range Berm Face, Tango Range Berm, India Range Berm Face, and Juliet Range Floor).

Water samples were extracted from operational lysimeters located adjacent to the berm face and to the port of STAPP systems by peristaltic pump into new 125 mL low-density polyethylene bottles.

2.2 Batch reactions

Batch reactors were set up using 250 mL high-density polyethylene sample bottles and filled with each of the soil types collected from JBCC SARs. Figure 1 shows the batch-reactor experimental setup. A mass of 25 g of soil in duplicate (for five different types of soil) was mixed with 200 mL of ultrapure Milli-Q deionized water (resistivity of 18.2 M Ω -cm) and placed on a shaker table and rotated at a speed of 150 revolutions per minute. The batch reactions were allowed to equilibrate at room temperature for 24 hours at 20°C. After 24 hours of equilibration time, the reaction time was referred to as “zero.” The liquid in the batch reactions was sampled at three different reaction times (0, 24, and 48 hours). For each of the three sampling events, the shaker table was stopped; the samples were allowed to settle for 5 minutes; and then 120 mL of aqueous sample was removed, filtered to less than 0.45 μ m with nylon syringe-fitted filters, acidified with ultrapure nitric acid, and stored at 4°C until analysis.

Initial pH measurements were taken within 30 minutes after all the components were combined in the reactors, and then pH was measured immediately before each sampling event. After each sampling event, 120 mL of ultrapure Milli-Q deionized water was added into each of the batch reac-

tions in preparation for the next sampling event. Batch reactions were simulated in duplicate and allowed to run a maximum of 48 hours (plus 24 hours equilibration time). There were five different batch-reaction systems as follows: (1) soil collected from the JBCC ranges with no addition, (2) soil collected from the ranges and spiked with Pb and Sb powder, (3) soil collected from the ranges and spiked with Pb and Sb powder and a lime amendment, (4) soil collected from the ranges and spiked with Pb and Sb powder, and (5) soil collected from the ranges and spiked with a phosphate amendment. Table 1 outlines the batch-reaction setup. The batch reactions (2–5) were spiked with 0.1 g elemental Sb and 0.5 g elemental Pb powder (200 mesh, 99.9% pure) to compare to control soils collected from ranges (batch reactor 1). The additions used were 0.5 g calcium hydroxide ($\text{Ca}(\text{OH})_2$), 0.5 g calcium phosphate tribasic ($\text{Ca}_3(\text{PO}_4)_2$), and 0.25 g iron (III) hydroxide alpha phase ($\alpha\text{-Fe}(\text{OH})_3$) per batch reaction.

Figure 1. Batch-reaction shaker-table setup, showing reactions 41–50 (iron amendment).



Table 1. Summary of batch-reaction contents, spike identification, and amendments added for sampling intervals of 0, 24, and 48 hours.

Batch	Contents	Amendment	Amendment Mass (g)
1	SAR soil (control)	-	-
2	SAR soil with spike (unamended)	-	-
3	SAR soil with spike and lime	$\text{Ca}(\text{OH})_2$	0.5
4	SAR soil with spike and phosphate	$\text{Ca}_3(\text{PO}_4)_2$	0.5
5	SAR soil with spike and iron	$\alpha\text{-Fe}(\text{OH})_3$	0.25

2.3 Soil analysis

Multi-increment soil samples, each weighing approximately 3 kg, were spread out on clean aluminum sheets to air dry until achieving a constant mass. Whole, dry soil samples were disaggregated with a clean-gloved hand and sieved through a #10 sieve to remove particles greater than 2 mm in diameter. Triplicate subsamples of the less than 2 mm fraction were collected by spreading the sample out evenly on the aluminum sheet and collecting 25 increments across the surface, totaling approximately 10 g. These subsamples were further subsampled for subsequent tests. Conductivity and pH were measured on 1:10 soil extracted with deionized water, shaken overnight, and filtered through 0.45 μm polytetrafluoroethylene filters by using Fisher Accumet probes with National Institute of Standards and Technology–traceable standards. Extractable anions (chloride, nitrite, sulfate, nitrate, and phosphate) and cations (sodium, ammonium, potassium, magnesium, and calcium) were measured in these extracts by ion chromatography (Thermo Integrion). Particle size was measured in soil disaggregated in sodium hexametaphosphate solutions (38 g/L) by using laser-diffraction particle-size analysis (Horiba LA-960). Organic matter was measured by proxy through mass loss on ignition at 375°C (LOI₃₇₅) in a muffle furnace for 4 hours. Soils were digested for metals analysis by mixing 1 g of soil with 2.5 mL of concentrated nitric acid and 10.0 mL of concentrated hydrochloric acid in polypropylene tubes, heating on a hot block at about 90°C for 4 hours, and diluting to 50 mL.

Soil samples were analyzed by inductively coupled plasma optical emission spectroscopy (Thermo iCAP 6500). Metal concentrations were quantitated using a six- or seven-point external calibration curve that was matrix-matched with the sample acid composition and normalized to an internal standard containing bismuth, indium, scandium, terbium, and yttrium introduced through a tubing Y junction.

2.4 Water analysis

Water samples were aliquoted for subsequent tests. An aliquot for pH, conductivity, metals analysis, and alkalinity was filtered through 0.45 μm PTFE (polytetrafluoroethylene) filters and measured using the same procedures as above. Alkalinity was determined by Gran titration with concentrated hydrochloric acid. Two fractions of metals were prepared: (1) a dissolved fraction by acidifying 25 g filtered aliquots to 1% nitric acid and (2) a total extractable fraction by adding 1.0 mL nitric acid and

2.5 mL hydrochloric acid to 50 g unfiltered aliquots and heating at 85°C for four hours.

Water samples were analyzed by inductively coupled plasma optical emission spectroscopy (Thermo iCAP 6500). Metal concentrations were quantitated using a six- or seven-point external calibration curve that was matrix-matched with the sample acid composition and normalized to an internal standard containing bismuth, indium, scandium, tellurium, and yttrium introduced through a tubing Y junction.

3 Results and Discussion

3.1 Soil samples

The soils collected from JBCC SARs exhibited textures reflecting silt loam to sandy loam and were particularly low in clay. Organic content in these soils was low, and soil pH values were circumneutral (Table 2). Overall soil composition and pH were relatively consistent across the sampled ranges. Soluble major anion and cation concentrations were also relatively consistent between the sampled ranges, except for phosphate concentrations that were one order of magnitude (Tango Range) and three orders of magnitude (Kilo Range) higher than in the other soils (Table 3).

Table 2. Soil particle size, pH, and conductivity results (mean \pm one standard deviation) of samples collected from Camp Edwards.

Site Name	Soil Classification (Volume % < 2 mm)			pH	Conductivity (μ S/cm)	LOI375 (wt. %)
	Clay	Silt	Sand			
Juliet Range Berm Face	0.8 \pm 0.2	42 \pm 2	57 \pm 2	6.66 \pm 0.02	33 \pm 2	2.3 \pm 0.1
Juliet Range Floor	0.6 \pm 0.1	37 \pm 6	63 \pm 6	6.65 \pm 0.01	61 \pm 3	4.2 \pm 0.2
Old Kilo Range Berm	0.5 \pm 0.0	54 \pm 4	46 \pm 4	7.19 \pm 0.03	126 \pm 2	5.4 \pm 0.3
India Range Berm Face	1.5 \pm 0.1	44 \pm 1	55 \pm 1	6.8 \pm 0.1	19 \pm 3	1.6 \pm 0.1
Tango Range Berm	0.7 \pm 0.1	48 \pm 2	52 \pm 2	6.97 \pm 0.05	81 \pm 3	5.9 \pm 0.4

Table 3. Soluble cation and anion concentrations (mean \pm one standard deviation) in soil samples collected from Camp Edwards.

Parameters (mg/kg)	Juliet Berm Face	Juliet Range Floor	Old Kilo Range Berm	India Berm Face	Tango Berm
Chloride	6.9 \pm 0.8	7.5 \pm 0.1	12.6 \pm 0.6	1.5 \pm 0.2	35 \pm 2
Nitrite	0.3 \pm 0	0.4 \pm 0	2.4 \pm 0.1	<0.2	1.1 \pm 0.1
Nitrate	7 \pm 0.7	8.3 \pm 0.4	12.6 \pm 0.3	19 \pm 3	13 \pm 2
Sulfate	3.3 \pm 0.6	8.5 \pm 0.8	6.7 \pm 0.7	8 \pm 1	16.2 \pm 0.4
Phosphate	0.6 \pm 0.1	0.7 \pm 0.1	119 \pm 4	0.2 \pm 0.0	13.3 \pm 0.5
Sodium	3.9 \pm 0.1	5.2 \pm 0.3	8.9 \pm 0.5	3.6 \pm 0.6	20.5 \pm 0.8
Ammonium	4.2 \pm 0	11.4 \pm 0.5	14.1 \pm 0.8	<1	3.3 \pm 0.4
Potassium	19.4 \pm 0.4	36.5 \pm 0.5	70 \pm 2	10 \pm 1	300 \pm 200
Magnesium	8.6 \pm 0.6	9.4 \pm 0.3	8.2 \pm 0.2	1.3 \pm 0.3	9.2 \pm 0.2
Calcium	19 \pm 1	37 \pm 2	178 \pm 4	17 \pm 4	47 \pm 4

Table 4 reports total extractable metal concentrations for the five SAR soils samples. Concentrations of Sb were relatively low and consistent between

all sampled soils (ranging from 7 ± 1 mg/kg at the Tango Range Berm to 11 ± 1 mg/kg at the Old Kilo Range Berm). However, Pb concentrations varied between the Juliet Range with the lowest concentration of approximately 30 mg/kg and the Kilo Range with the highest concentration of 430 mg/kg. Copper concentrations were greatest in the soil from India Range but also highly variable, potentially reflecting the sampling of Pb-free bullet fragments as historically these ranges were used for firing both leaded and Pb-free projectiles. Iron and manganese concentrations, which could affect Sb and Pb mobility through sorption reactions, were relatively consistent among all soil samples. The highest concentrations of iron and manganese were located at the Juliet Range Floor ($11,000 \pm 1000$ mg/kg iron and 140 ± 10 mg/kg manganese) and to a lesser extent the Tango Range Berm (9500 ± 800 mg/kg iron and 180 ± 10 mg/kg manganese).

Table 4. Extractable metals concentrations using aqua regia digestion (mean \pm one standard deviation) for the soils collected from Camp Edwards.

Parameters (mg/kg)	Juliet Berm Face	Juliet Range Floor	Old Kilo Range Berm	India Berm Face	Tango Berm
Arsenic	22 ± 2	40 ± 3	35 ± 2	23 ± 2	38 ± 3
Barium	8.8 ± 0.5	27 ± 3	11 ± 1	12 ± 1	43 ± 8
Calcium	750 ± 40	3000 ± 1000	17000 ± 1000	600 ± 100	3000 ± 300
Copper	6.1 ± 0.2	8.9 ± 0.7	40 ± 3	500 ± 500	68 ± 6
Iron	6300 ± 900	11000 ± 1000	8400 ± 200	7100 ± 400	9500 ± 800
Magnesium	580 ± 50	3200 ± 300	990 ± 80	700 ± 70	1410 ± 40
Manganese	58 ± 7	140 ± 10	72 ± 5	88 ± 7	180 ± 10
Sodium	16 ± 1	41 ± 9	24 ± 4	26 ± 4	75 ± 4
Nickel	2.9 ± 0.4	9 ± 1	4.6 ± 0.5	4.0 ± 0.4	7 ± 1
Lead	28 ± 1	35 ± 2	430 ± 20	151 ± 90	110 ± 10
Antimony	8 ± 2	8 ± 1	11 ± 1	7 ± 2	7 ± 1
Strontium	2.7 ± 0.2	6.6 ± 0.4	14 ± 1	5 ± 1	15 ± 2
Tungsten	6 ± 1	12 ± 2	50 ± 3	7 ± 1	10 ± 1
Zinc	29 ± 2	35 ± 1	32 ± 2	110 ± 40	80 ± 10

It is important to note that calcium concentrations were significantly higher in the sample collected at the Old Kilo Range Berm ($17,000 \pm 1000$ mg/kg calcium) in comparison to the other soil samples (Table 3). The Old Kilo Range Berm also contains a relatively high level of silt, a higher pH value, and a higher conductivity (Table 2) compared to the other soils. This is likely due to the presence of fine-grained calcium carbonate, which is easily extractable, a result of prior liming applications. Thus, the application of

lime to the soil is one possible theory explaining the increased mobility of Sb with higher calcium concentrations associated with higher Sb levels.

3.2 Water samples

Six water samples were collected from three different SARs at JBCC: four lysimeters at Juliet, Kilo, and Tango Ranges were sampled and two STAPP systems at Juliet and Kilo Ranges. Table 5 lists major water chemistry properties and soluble ion concentrations. The water samples collected from the STAPP systems tended to have the greatest conductivity and ionic composition, particularly of calcium, magnesium, chloride, and sulfate, whereas the lysimeters exhibited low concentrations consistently. Dissolved and total organic carbon (DOC and TOC) in STAPP samples were also elevated compared with the lysimeters.

Table 5. Chemical characterization and soluble ion composition of water samples collected from Camp Edwards ranges.

Analyte	Juliet Range Lysimeter	Juliet Range STAPP	Kilo Range Lysimeter 1	Kilo Range Lysimeter 2	Kilo Range STAPP	Tango Range Lysimeter	Blank
pH	6.98	6.89	7.66	7.16	4.93	5.5	5.84
Conductivity ¹	290.6	1609	54.41	121.7	1635	109.4	1.39
Alkalinity ²	140	155	52	96	56	30	-
DOC ³	14.7	127	9.44	21.7	89.4	11.1	-
TOC	12.4	225	11.6	21.2	90.1	7.64	-
Chloride	4.67	96.4	2.71	3.29	86.5	11.2	<0.02
Nitrite	<0.1	<10	<0.1	<0.1	<10	<0.1	<0.02
Nitrate	0.2	<10	0.9	<0.1	103	11.1	<0.02
Sulfate	4.33	640	1.75	0.27	727	17.1	<0.02
Phosphate	<0.1	<10	<0.1	<0.1	<10	<0.1	<0.02
Sodium	3.79	13.5	1.93	2.13	11.6	6.79	<0.02
Ammonium	<0.2	39	<0.2	<0.2	16.2	<0.2	<0.03
Potassium	0.58	<8	0.5	0.8	7.79	1.07	<0.02
Magnesium	3.36	69.5	0.68	0.36	57.1	2.5	<0.02
Calcium	47.7	148	6.65	21.1	160	6.49	<0.09

Note: All concentrations are in mg/kg except as below.

¹ $\mu\text{S}/\text{cm}$

² $\text{mg CaCO}_3/\text{L}$

³ mg/L

Metal concentrations in water samples are reported for the dissolved (<0.45 μm diameter) fraction in Table 6 and total extractable in Table 7. Figure 2 compares these two fractions for select metals. The water samples

collected from the STAPP systems consistently contained higher concentrations of analytes (arsenic, barium, calcium, iron, magnesium, manganese, sodium, nickel, Pb, Sb, strontium, tungsten, and zinc). The higher STAPP concentrations are attributed to the concentrated collection system whereby projectiles are entrained within the STAPP. Perforations of the STAPP membrane allow precipitation to collect within the STAPP and interact with the projectiles before collecting at the sump at the base. Consequently, the effluent in the sump is enriched with respect to the SAR metals of interest.

The Pb and Sb concentrations were relatively low in the lysimeters but high in the STAPP systems at Juliet Range and Kilo Range. The Juliet Range STAPP Pb and Sb concentrations are predominantly attributed to the total metal fraction, whereas the dissolved metal fraction comprises approximately all of the Pb and Sb in the Kilo Range STAPP. This difference may reflect the lower pH in the Kilo Range STAPP (4.93), which promotes metal dissolution, or the lower concentration of DOC, TOC, and iron, which would promote sorption and complexation.

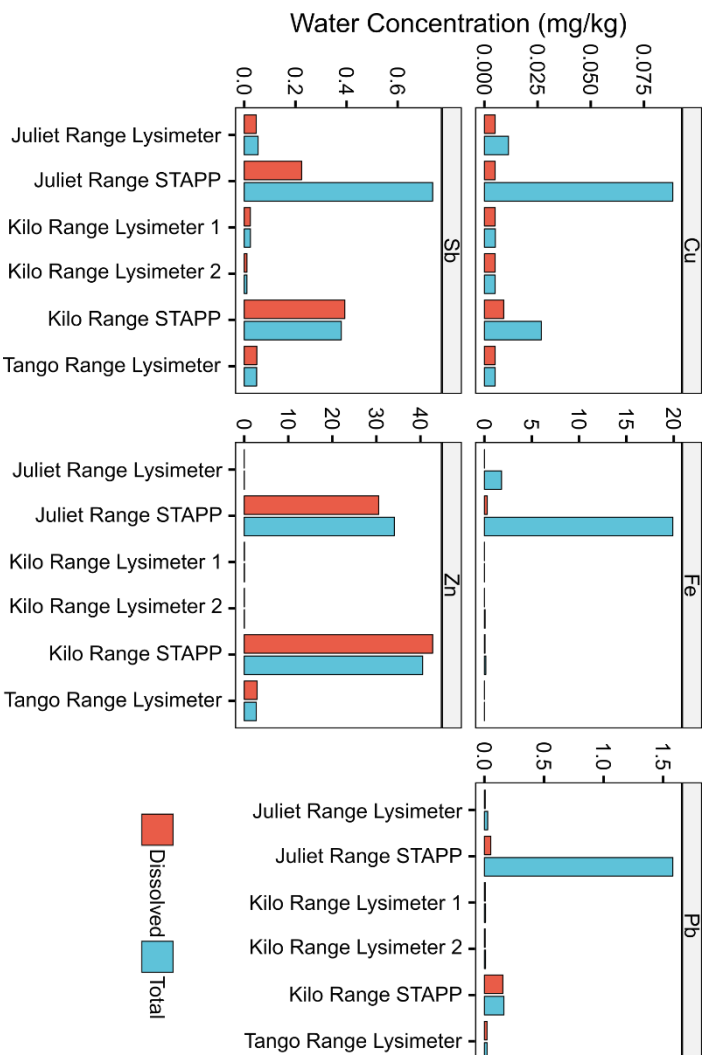
Table 6. Dissolved metal concentrations (mg/kg) in water samples collected from Camp Edwards ranges.

Analyte	Juliet Lysimeter	Juliet STAPP	Kilo Range Lysimeter 1	Kilo Range Lysimeter 2	Kilo STAPP	Tango Lysimeter	Blank
Arsenic	0.013	0.084	<0.005	0.006	0.038	<0.005	<0.005
Barium	<0.005	0.103	<0.005	<0.005	0.019	0.017	<0.005
Calcium	51.9	148	7.28	22.5	161	6.43	0.019
Copper	<0.005	<0.005	<0.005	<0.005	0.009	<0.005	<0.005
Iron	0.013	0.311	<0.005	<0.005	0.059	<0.005	<0.005
Magnesium	3.70	49.9	0.791	0.422	44.0	2.52	<0.005
Manganese	<0.005	0.720	<0.005	<0.005	0.088	0.014	<0.005
Sodium	4.79	19.8	2.45	2.73	16.5	7.65	<0.005
Nickel	<0.005	0.044	<0.005	<0.005	0.188	<0.005	<0.005
Lead	0.007	0.052	0.008	0.007	0.154	0.022	<0.005
Antimony	0.046	0.224	0.024	0.011	0.393	0.049	<0.005
Strontium	0.041	0.313	0.016	0.010	0.209	0.009	<0.005
Tungsten	<0.005	0.011	<0.005	<0.005	0.084	<0.005	<0.005
Zinc	<0.005	30.5	0.048	0.034	42.8	2.88	<0.005

Table 7. Total extractable metal concentrations (mg/kg) in water samples collected from Camp Edwards ranges.

Analyte	Juliet Lysimeter	Juliet STAPP	Kilo Lysimeter 1	Kilo Lysimeter 2	Kilo STAPP	Tango Lysimeter	Blank
Arsenic	0.015	0.090	0.005	0.006	0.037	<0.005	<0.005
Barium	0.012	0.109	<0.005	<0.005	0.023	0.021	<0.005
Calcium	48.3	139	6.89	20.9	150.7	6.12	0.008
Copper	0.011	0.089	0.005	<0.005	0.027	<0.005	<0.005
Iron	1.81	19.9	0.012	0.070	0.146	<0.005	<0.005
Magnesium	3.63	48.2	0.761	0.407	42.8	2.44	<0.005
Manganese	0.008	0.707	<0.005	<0.005	0.089	0.017	<0.005
Sodium	4.70	19.9	2.47	2.69	16.5	7.75	<0.005
Nickel	<0.005	0.053	<0.005	<0.005	0.183	<0.005	<0.005
Lead	0.027	1.58	0.009	0.009	0.164	0.023	<0.005
Antimony	0.054	0.737	0.024	0.010	0.380	0.048	<0.005
Strontium	0.043	0.313	0.019	0.014	0.208	0.012	<0.005
Tungsten	<0.005	0.119	<0.005	<0.005	0.091	<0.005	<0.005
Zinc	0.015	34.1	0.042	0.036	40.5	2.74	<0.005

Figure 2. Metal concentrations in water samples from Camp Edwards.



3.3 Batch reactions

The behavior and mobility of Pb and Sb as it relates to the three different amendments was investigated using batch reactions and shaker-table experiments to better understand the increased Sb concentrations following the lime addition to berm surface soils. A phosphate amendment was also evaluated since previous remediation efforts at Camp Edwards used this material to limit Pb mobility. An iron amendment was investigated because it has potential to immobilize both Pb and Sb concurrently. Concentrations of Pb and Sb in batch-reaction vessels are plotted by reaction time (Figure 3) and soil type (Figure 4) and reported in Table 8.

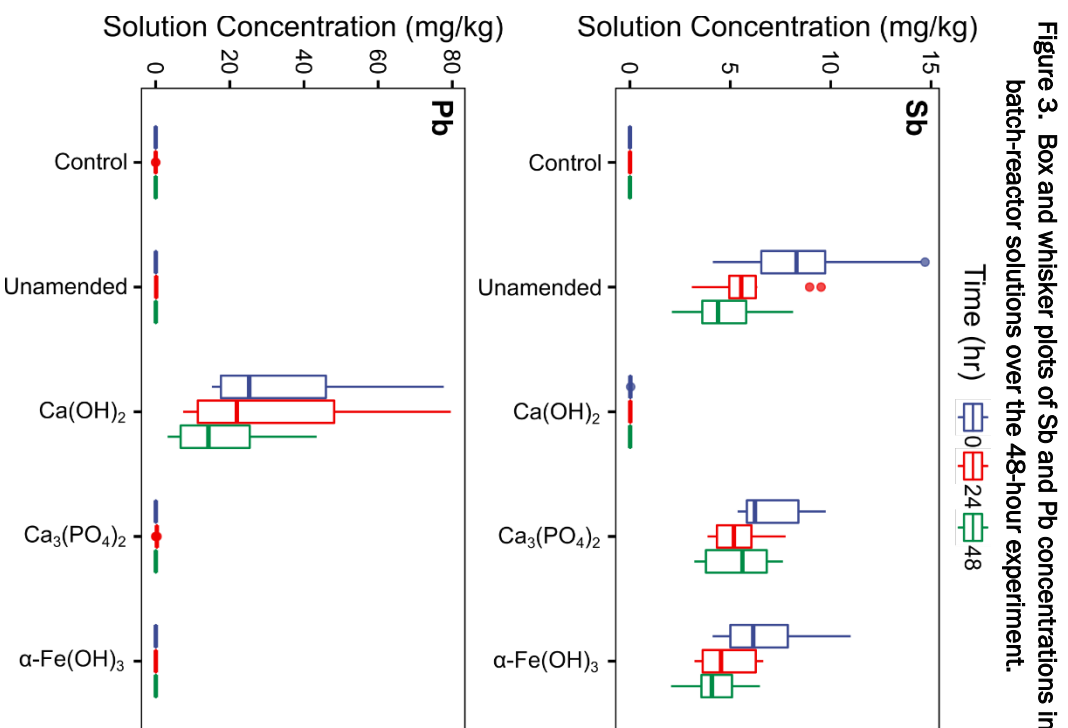


Figure 4. Box and whisker plots of Sb and Pb concentrations in batch-reactor solutions after 48 hours by soil used in the reactor.

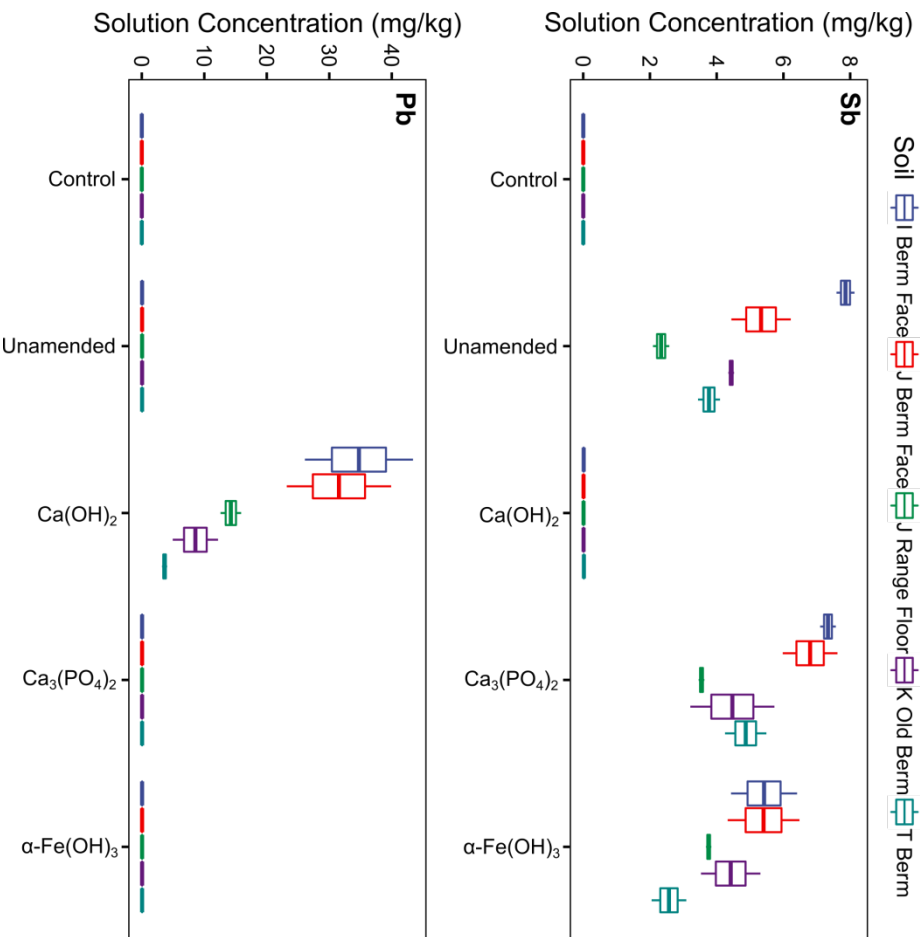


Table 8. Concentrations (mg/kg) of lead and antimony in batch-reaction solutions.

Range Soil	Type	Amendment	Lead (mg/kg)			Antimony (mg/kg)			pH		
			0 hr	24 hr	48 hr	0 hr	24 hr	48 hr	0 hr	24 hr	48 hr
Old Kilo Berm	Control	None	<0.005	<0.005	<0.005	<0.005	0.009	<0.005	7.4	7.7	7.8
Old Kilo Berm	Control	None	<0.005	0.005	<0.005	<0.005	0.011	<0.005	7.5	7.5	7.6
Juliet Berm Face	Control	None	<0.005	<0.005	<0.005	<0.005	0.009	<0.005	7.5	7.5	7.5
Juliet Berm Face	Control	None	<0.005	<0.005	<0.005	<0.005	0.008	<0.005	7.3	7.3	7.4
Tango Berm	Control	None	<0.005	<0.005	<0.005	<0.005	0.006	<0.005	7.3	7.3	7.4
Tango Berm	Control	None	<0.005	0.005	<0.005	<0.005	<0.005	<0.005	7.3	7.3	7.3
India Berm Face	Control	None	<0.005	<0.005	<0.005	<0.005	0.007	<0.005	7.6	7.5	7.5
India Berm Face	Control	None	<0.005	<0.005	<0.005	<0.005	0.011	<0.005	7.5	7.5	7.8
Juliet Range Floor	Control	None	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	6.9	7	7.3
Juliet Range Floor	Control	None	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	6.9	6.9	7.1
Old Kilo Berm	Spike	None	<0.1	0.126	<0.1	6.12	5.64	4.52	7.2	7.3	7.3
Old Kilo Berm	Spike	None	<0.1	<0.1	<0.1	7.83	6.35	4.35	7.2	7.3	7.5
Juliet Berm Face	Spike	None	<0.1	0.14	<0.1	8.36	6.03	6.22	7.5	7.5	7.7
Juliet Berm Face	Spike	None	<0.1	0.158	<0.1	8.23	4.92	4.44	7.4	7.6	7.8

Table 8 (cont.). Concentrations (mg/kg) of lead and antimony in batch-reaction solutions.

Range Soil	Type	Amendment	Lead (mg/kg)			Antimony (mg/kg)			pH		
			0 hr	24 hr	48 hr	0 hr	24 hr	48 hr	0 hr	24 hr	48 hr
Tango Berm	Spike	None	<0.1	0.171	<0.1	10.1	5.03	3.44	7.4	7.5	7.7
Tango Berm	Spike	None	<0.1	0.221	<0.1	8.62	5.46	4.10	7.4	7.6	7.7
India Berm Face	Spike	None	<0.1	0.157	<0.1	14.7	9.52	8.13	7.6	7.8	8
India Berm Face	Spike	None	<0.1	<0.1	<0.1	14.5	8.95	7.59	7.5	7.8	8
Juliet Range Floor	Spike	None	<0.1	0.308	<0.1	4.13	3.08	2.09	7.2	7.6	7.8
Juliet Range Floor	Spike	None	<0.1	0.277	<0.1	5.92	3.46	2.58	7.2	7.4	7.7
Old Kilo Berm	Spike	Ca(OH) ₂	28.8	79.6	12.2	0.025	0.021	0.008	11.8	11.5	11.3
Old Kilo Berm	Spike	Ca(OH) ₂	16.3	9.42	4.95	0.023	0.026	0.015	11.7	11.4	11.3
Juliet Berm Face	Spike	Ca(OH) ₂	28.2	30.2	23.2	0.042	0.027	0.014	11.8	11.6	11.5
Juliet Berm Face	Spike	Ca(OH) ₂	70.2	54.1	39.9	0.027	0.018	0.006	11.9	11.6	11.4
Tango Berm	Spike	Ca(OH) ₂	22.3	7.79	4.05	0.024	0.024	0.018	11.8	11.4	11.3
Tango Berm	Spike	Ca(OH) ₂	17.5	7.43	3.24	0.025	0.022	0.022	11.7	11.7	11.2
India Berm Face	Spike	Ca(OH) ₂	77.7	68.4	43.4	0.028	0.032	0.019	11.9	11.7	11.5
India Berm Face	Spike	Ca(OH) ₂	51.6	26.2	26.1	0.026	0.021	0.010	11.9	11.6	11.5
Juliet Range Floor	Spike	Ca(OH) ₂	15.2	17.2	12.6	0.019	0.017	0.009	11.8	11.6	11.4
Juliet Range Floor	Spike	Ca(OH) ₂	17.9	17.6	15.9	0.021	0.021	0.011	11.8	11.5	11.5
Old Kilo Berm	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.316	<0.1	6.31	3.87	3.21	8.4	8.1	7.9
Old Kilo Berm	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.292	<0.1	9.4	6.11	5.73	8.1	7.8	7.7
Juliet Berm Face	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.314	<0.1	8.67	7.76	7.62	7.9	7.8	7.8
Juliet Berm Face	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.398	<0.1	5.69	5.06	5.98	7.9	8	7.8
Tango Berm	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.332	<0.1	9.75	6.35	5.49	7.9	7.8	7.7
Tango Berm	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.318	<0.1	7.58	4.70	4.25	7.7	7.8	7.7
India Berm Face	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.339	<0.1	5.72	5.30	7.10	8.1	8.5	8.2
India Berm Face	Spike	Ca ₃ (PO ₄) ₂	<0.1	0.387	<0.1	6.15	5.88	7.57	8	8.4	8.2
Juliet Range Floor	Spike	Ca ₃ (PO ₄) ₂	<0.1	<0.1	<0.1	6.15	4.22	3.46	7.5	7.6	7.6
Juliet Range Floor	Spike	Ca ₃ (PO ₄) ₂	<0.1	<0.1	<0.1	5.37	4.16	3.63	7.4	7.5	7.6
Old Kilo Berm	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	7.6	4.84	3.53	7.4	7.6	7.5
Old Kilo Berm	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	11	6.65	5.31	7.5	7.6	7.6
Juliet Berm Face	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	8.31	6.38	6.48	7.7	7.9	7.9
Juliet Berm Face	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	6.61	4.54	4.33	7.7	7.7	8
Tango Berm	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	4.12	3.22	2.05	7.6	7.7	7.8
Tango Berm	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	5.05	3.33	3.09	7.6	8	7.8
India Berm Face	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	4.67	3.63	4.43	7.8	8	8.4
India Berm Face	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	7.96	6.27	6.41	7.8	7.9	8.1
Juliet Range Floor	Spike	αFe(OH) ₃	<0.1	-	<0.1	5.68	-	3.84	7.4	7.6	7.7
Juliet Range Floor	Spike	αFe(OH) ₃	<0.1	<0.1	<0.1	4.99	4.18	3.68	7.3	7.5	7.8

Note: Dashes refer to sample results not determined

3.3.1 Untreated soils

The concentrations of Pb and Sb in the control soil reactor solutions were generally less than 5 ppb throughout the 48-hour experiment. These low concentrations reflect the relatively slow dissolution of in situ Pb and Sb in JBCC soils. In soils spiked with metallic Pb (0.5 g) and Sb (0.1 g) and unamended, concentrations were slightly increased for Pb but greatly increased for Sb compared with the control group. These observations highlight the overall slow oxidation of metallic Pb versus Sb oxidation, which is kinetically fast on the order of roughly 2 hours (Ilgen et al. 2014). Consequently, the mobility of Sb will be greater than Pb following the introduction of leaded projectiles into unamended JBCC soils. Concentrations of Sb in the metal(loid) spiked unamended soils decreased over time, suggesting sorption to in situ soil minerals or organic matter. Therefore, an increase in Sb soil pore-water concentrations can be expected immediately following firing with leaded ammunition; however, with increasing time, Sb levels will decline.

3.3.2 Lime addition

For the lime (calcium hydroxide) amendment, concentrations of Pb increased initially compared to the spiked soils with no amendments, likely attributable to the overall increase in pH from the dissolution of the lime (hydroxide added into the system). The aqueous solution for the lime addition for this batch reaction exhibited pH values between 11.2 and 11.9, resulting from a relatively large addition of lime to the system (0.5 g lime per 25 g soil and 200 mL deionized water). Previously, Pb (depending on the speciation) has been shown to be less soluble at pHs ranging from about 7.5 to 10.5 for the minerals cerussite (PbCO_3) and hydrocerussite ($\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$) (Sheetz 2004). Previous work has shown that Pb can form slightly soluble Pb-hydroxide complexes at pH 12 (Clevenger and Dave 1998). However, the upper pH limit for mobilizing the Pb species found in the soils at Camp Edwards is approximately 11. After 48 hours reaction time of the lime to the soil, the soil constituents likely began buffering the system. Thus, pH and Pb concentrations decreased concomitantly (Table 8). Further work is needed to constrain the lower pH limit for Pb mobility.

For Sb, the lime addition had an opposite effect on its behavior as compared to Pb, which highlights the often diverging behavior between Pb and Sb. Concentrations of Sb (Table 8) were lower in the lime-amended soils

than in the spiked unamended soils. This observation is contrary to the expected cause-and-effect relationship between liming activities at JBCC SAR berms and increasing Sb levels. The overall mobilization versus retention of Sb in soils is controlled by the aqueous and solid-phase speciation (Sb[III] versus Sb[V]). Without determining the speciation, it is difficult to estimate mobility. The lime addition in this study did not initiate mobilization of Sb, instead likely precipitating Sb as a solid-phase species. We suspect that the ratio of lime to soil volume was much higher in our batch experiments compared to the field additions, which may explain our unexpected results. Our proposed column experiments will help to explore the relationship between Sb mobilization as a result of lime additions. Further work is also needed to estimate the lower pH limit for Sb precipitation and if the dominant phase is Sb(III) or Sb(V), which will dictate solubility as a function of changes in pH.

3.3.3 Phosphate addition

The phosphate (calcium phosphate tribasic) addition had little effect on Pb mobilization compared to the control and the spiked unamended batches, which was expected. Lead phosphates are one of the most stable forms of Pb in the environment, and they have been shown to rapidly form when phosphate amendments are added to Pb-contaminated soils (Cao et al. 2008). Antimony concentrations were relatively unchanged with the addition compared to the spiked unamended batch reactions (Table 8). In some batches, concentrations of Sb were higher than the spiked soils; and in other batches, Sb concentrations were lower. Thus, there is a potential that the phosphate amendment did mobilize Sb as seen in previous studies (Griggs et al. 2011) but was only observed to a small extent in this study. Column experiments should provide a clearer insight into the processes involved and the observations from JBCC.

3.3.4 Iron addition

Iron addition was selected for this study as a potential remediation option because previous studies have shown it to have a natural affinity for immobilizing both Pb and Sb at a variety of pH values (Scheinost et al. 2006; Ackermann et al. 2009; Okkenhaug et al. 2016). Iron (III) hydroxide was chosen for our tests due to its high surface area and because it does not alter solution pH upon addition, unlike other iron-based amendments, particularly iron (II). Additionally, the subsurface sediments at JBCC contain

elevated levels of iron hydroxide, which may explain the lack of Pb mobilization at the SARs despite leaded ammunition usage for over 75 years. We found that the iron amendment in this study immobilized Pb to a greater extent than Sb (Table 8). Concentrations of Pb in the spiked soils ranged from below 0.1 mg/kg up to 0.308 mg/kg (308 ppb), which in comparison to the iron-amended concentrations, were significantly higher. The Pb concentrations were all below 0.1 mg/kg for the iron-amended reactions, showing it is an effective remediation option for Pb.

For Sb, there was little difference between the iron-amended batches and the spiked unamended soil batches. While Sb and Pb both have a natural affinity for iron, the results from this batch study suggest that, with potentially limited iron sorption sites (0.25 g α Fe(OH)₃ added to the system per 25 g soil), Pb is potentially more competitive for sorption sites than Sb. With a higher amount of iron added to the system, it is possible that competitive sorption will not take place and both Pb and Sb will become immobilized. Further work is needed to determine what amount of iron (III) hydroxide is needed to supply enough sorption sites for both Pb and Sb.

4 Conclusions

Previously reported Sb concentrations exceeded the OMMP Action Levels of 6 ppb beginning in 2012 and 2013 for the Juliet and Kilo Ranges and 2016 for the Tango Ranges. Understanding what caused elevated Sb concentrations particular to the Camp Edwards ranges is important for assessing future risks and the potential for off-site migration.

The three amendments chosen for this batch study had varying effects on solution concentrations of Sb and Pb and solution pH for the different reactions. The iron amendment successfully immobilized Pb relative to the spiked reactors. The phosphate and iron amendments had little effect on Sb solution concentrations relative to the unamended reactors, whereas the lime amendment reduced Sb concentrations nearly to the method detection limit but increased solution Pb concentrations. While the alkaline conditions (pH ~11.5) brought about by the lime amendment seem successful in suppressing dissolution of Sb, it appears to promote the dissolution of Pb. Further study is needed to constrain the pH effects for Pb versus Sb mobilization as a function of lime additions.

A decreasing temporal trend in Pb and Sb concentrations is apparent for all amendments and the unamended reactors, indicative of a potential kinetic limitation for interaction of dissolved Pb with adsorption sites on both the amendments and natural soil surfaces. Longer reaction times are needed to confirm the reaction kinetics and to determine equilibrium conditions.

Concentrations of the Pb and Sb solutions depended on the soil used in the batch-reaction tests. Juliet Range Berm Face and India Range Berm Face soils had the greatest Sb concentrations in the unamended and the Phosphate- and iron-amended soil tests. The highest Pb concentrations were observed in the lime-amended soil test. These soils had the lowest organic matter and iron concentrations, highlighting the importance of individual site soils on metal mobility.

5 Recommendations

This report details ERDC-CRREL's Tasks 1 and 2 of the Statement of Work dated 23 May 2018 covering batch-reaction experiments and supplemental JBCC SAR data analysis, respectively. The behavior of Pb and Sb was highly dependent on pH and availability of sorption sites. However, longer reaction times, greater than the 24 hours, are needed to confirm the reaction kinetics and to determine equilibrium conditions. Multiday and multiweek soil column studies would help to better understand the reaction kinetics. For Sb, the overall mobilization versus retention in soils is controlled by the aqueous and solid-phase speciation [Sb(III) versus Sb(V)]; and without determining the speciation, it is difficult to estimate mobility. Additionally, further work is needed to estimate the lower pH limit for Sb precipitation and if the dominant phase is Sb(III) or Sb(V), which will dictate solubility as a function of changes in pH. Implementing these recommendations would assist in our understanding of the cause of the increased Sb mobility observed at Camp Edwards SARs.

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