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Chemical durability assessment of enhanced low-activity waste glasses through EPA method 1313

May 2023

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U.S. DEPARTMENT OF ENERGY

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Executive Summary

This report describes the progress of the Glass Leaching Assessment for Durability (GLAD) program towards developing new strategies to understand the chemical durability of nuclear waste glasses that will be disposed in a near-surface disposal facility. The United States Environmental Protection Agency (EPA) Leaching Environmental Assessment Framework (LEAF), pH-dependent leach test (EPA Method 1313 and further abbreviated EPA 1313) was implemented to study the alteration behavior of low-activity nuclear waste (LAW) glasses at temperatures that are representative of the integrated disposal facility. Certain modifications were made to the EPA 1313 method, including constraining particle size, reducing the studied pH range, and reducing solution volume, to study LAW glass corrosion and provide more reproducible results. Implementation of the room temperature EPA 1313 method, which is closer to disposal conditions, could allow for more flexibility in the formulation of Hanford LAW glasses, as well as increased waste loading.

A series of sixteen enhanced LAW glasses were selected from a previous matrix, designed using statistical methods, to study glass alteration behavior with EPA 1313. This set of glasses is referred to as the LAW Phase 3 (LP3) matrix. A second glass matrix was also prepared based on a composition representative of vitrified material from the ~1500-year-old Broborg hillfort in Sweden, that is being used as an analogue to understand the long-term corrosion behavior of vitreous materials in a near-surface environment. Using the synthetic Broborg glass as a baseline, seven glasses were synthesized to establish a link between the composition of the LAW glasses and the composition of the Broborg analogue glass, and to broaden the compositional range tested using EPA 1313. This set of glasses is referred to as the join glass matrix.

Compositional modeling was used to fit the measured elemental release curves as a function of pH and concluded that high concentrations of elements such as silicon reduced the release of all elements across the pH range tested. This is consistent with elemental modeling of results from the Product Consistency Test (PCT) and Vapor Hydration Test (VHT); the tests currently used as requirements to assess durability of LAW glasses at the Hanford Waste Treatment & Immobilization Plant (WTP). The results demonstrate LAW glasses studied here provide a relatively similar elemental release through EPA 1313 despite the larger relative differences observed through PCT or VHT, suggesting that a future requirement could be defined that would allow broader compositional ranges for LAW glass formulations.

The ultimate goal of studying the Hanford LAW glass response to EPA 1313 is to assess the chemical durability of the wasteform. To facilitate the use of EPA 1313 for that purpose, additional tests should be performed on glasses containing key radioactive (Tc-99, I-129) and hazardous (Pb, Cr, Ba) components to better assess their leaching behavior under these test conditions.

Acknowledgments

The authors gratefully acknowledge the financial support provided by the U.S. Department of Energy Office of River Protection Waste Treatment and Immobilization Plant Project managed by Tom Fletcher, with technical oversight by Albert Kruger.

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Acronyms and Abbreviations

ANN	Artificial Neural Network
BSE	Backscatter electron imagining
DFLAW	Direct-feed low-activity waste
DIW	deionized water
DOE	U.S. Department of Energy
EPA	Environmental Protection Agency
EPMA	Electron Probe Microanalysis
EWG	Enhanced Waste Glass
GLAD	Glass Leaching Assessment for Durability
ICP-MS	inductively coupled plasma – mass spectroscopy
ICP-OES	inductively coupled plasma – optical emissions spectroscopy
IDF	Integrated Disposal Facility
LAW	low-activity waste
LEAF	Leaching Environmental Assessment Framework
LP3	LAW Phase 3
ML	Machine Learning
NQAP	Nuclear Quality Assurance Program
OM	optical microscopy
ORP	DOE Office of River Protection
PCT	Product Consistency Test following C1285-21
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
SEM	scanning electron microscopy
SPFT	Single pass flow through
S/V	surface area-to-solution volume
VHT	Vapor Hydration Test following ASTM C1663-18
wt%	weight percent
WTP	Waste Treatment and Immobilization Plant

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1.0 Introduction

The U.S. Department of Energy (DOE) Office of River Protection (ORP) requested Pacific Northwest National Laboratory (PNNL) to support the River Protection Project vitrification program in preparation for operations upon completion of startup activities including direct-feed low activity waste (DFLAW) (DOE 2012). PNNL has and continues to execute a number of critical activities in support of the vitrification flowsheet under a project titled "ORP Glass Support Work." One task in this project— Glass Leaching Assessment for Durability/Environmental Protection Agency (GLAD/EPA)—is the subject of this report. The GLAD/EPA task has the objective of developing strategies to implement EPA Method 1313 (called EPA 1313 throughout the report) to test low-activity nuclear waste (LAW) glass corrosion in realistic disposal conditions.

This report presents the EPA 1313 results of sixteen enhanced Hanford LAW Phase 3 (LP3) glasses and the join glasses, a series of glasses which link the LAW compositional region with a ~1500-year-old Hillfort glass, which is being used as an analogue to assess long-term corrosion. Section 1.1 summarizes the range of LAW glasses considered, Section 1.2 describes the selection of glasses from a previous LAW glass composition matrix, and Section 1.3 documents the quality assurance program used to prepare the quenched glasses.

1.1 Status of LAW Experimental Glass Composition Regions and Waste Loading Constraints

The Hanford Tank Waste Treatment and Immobilization Plant (WTP) project has previously developed glass property-composition databases and models to formulate LAW glass compositions and qualify LAW glasses for disposal (Kim et al. 2012, Vienna et al. 2021). These compositions fall in two general composition regimes:

1) WTP baseline glass compositions that are relatively low in waste loading. The formulation approach holds several components at nearly constant concentrations: Al_2O_3 (6.1 wt%), B_2O_3 (10.0 wt%), Fe_2O_3 (5.5 wt%), TiO_2 (1.4 wt%), ZnO (3.5 wt%), and ZrO_2 (3.0 wt%) while decreasing concentrations of CaO (2.0 to 7.0 wt%), Li_2O (0 to 4.3 wt%), and MgO (1.5 to 3.0 wt%) with increased Na_2O (5.4 to 21.0 wt%) (Kim et al. 2012). The WTP LAW facility will be commissioned using these glass compositions.

2) Enhanced waste glass (EWG) compositions are designed to expand the loading and waste feed compositional flexibility. A specific composition will be design for each waste by numerical optimization methods to simultaneously satisfy a number of property constraints with 90% confidence. These compositions cover a significantly broader composition space. On average, the loading in these glasses is more than 40 relative percent higher than those of the WTP baseline glasses (Lu et al. 2021).

The composition regions covered by the baseline and EWG domains are summarized in Table 1-1, as listed in (Piepel et al. 2007, Vienna et al. 2021).

Table 1-1. Comparison of glass composition regions and baseline glass target compositions in mass fractions (Piepel et al. 2007, Vienna et al. 2021)

Region	EWG Composition Region		WTP Baseline Model Region	
Oxide	Min	Max	Min	Max
Al_2O_3	0.0350	0.1475	0.035	0.090
B_2O_3	0.0600	0.1383	0.060	0.131
CaO	0.0000	0.1278	0.000	0.105
Cl	0.0000	0.0117	0.000	0.009
Cr_2O_3	0.0000	0.0063	0.000	0.006
F	0.0000	0.0130	0.000	0.004
Fe_2O_3	0.0000	0.1198	0.000	0.080
K_2O	0.0000	0.0809	0.000	0.054
Li_2O	0.0000	0.0582	0.000	0.058
MgO	0.0000	0.0502	0.000	0.050
Na_2O	0.0247	0.2657	0.025	0.230
P_2O_5	0.0000	0.0341	0.000	0.030
SO_3	0.0000	0.0163	0.000	0.010
SiO_2	0.3352	0.5226	0.384	0.521
SnO_2	0.0000	0.0503	0.000	0.000
TiO_2	0.0000	0.0400	0.000	0.030
V_2O_5	0.0000	0.0409	0.000	0.000
ZnO	0.0000	0.0582	0.010	0.054
ZrO_2	0.0000	0.0675	0.000	0.050
Others	0.0000	0.0216	0.0000	0.0028

1.2 Glass selection

In this work, sixteen LAW glasses were selected in the EWG composition region from an existing glass matrix (LP3) (Lonergan et al. 2020) and were subjected to EPA 1313 testing, the compositions are provided in Table 1-2. The glasses had already been synthesized and tested using the product consistency test (PCT) (ASTM 2021) and the vapor hydration test (VHT) (ASTM 2018). Glasses LP3-11, -13, -17, and -18 were not tested in the present study as there was limited sample readily available. The LP3 glasses were designed to be near the WTP contract (DOE 2000) pass-fail boundaries for VHT and PCT. The LP3 glasses either passed both VHT and PCT, failed both VHT and PCT, or failed VHT and passed PCT. Figure 1-1 provides a scatterplot matrix of the major elements for the LP3 glasses. Table 1-3 provides the measured PCT response and VHT alteration rates for the LP3 glasses (Lonergan et al. 2020).

The Broborg hillfort is an archaeological site located near Uppland, Sweden where Iron Age inhabitants produced a partially vitrified wall. The vitrified material has been subjected to near-surface conditions for over 1500 years, providing the opportunity to investigate long term alteration of glasses under environmental conditions, like those at the Hanford IDF, where the LAW glass will be disposed. A glass

(designated as the ‘dike’ sample) was synthesized in the laboratory by melting the amphibolitic rocks that the Iron Age people used as a source term to make the vitrified wall at Broborg. This glass was subjected to EPA 1313 testing to compare the alteration behavior with that for vitrified material altered over hundreds of years in the environment (Nava-Farias et al. 2021). The Broborg analogue glass has a composition that is outside the LP3 compositional matrix (Table 1-2), so a series of “join” glasses were synthesized along a compositional join from the Broborg archaeological glass to glass of similar composition to the Broborg glass but modified with the amount of sodium and boron that is present in LAW glass (20 wt% Na_2O and 10 wt% B_2O_3). We note that this is not a true compositional join as more than one element is changed at the same time, and there was no attempt to keep the molar ratios of all components equal. However, the strategy of increasing Na_2O and B_2O_3 compositions allows for a qualitative assessment of the impact of these constituents on the durability of the glasses. In addition, the join glasses provide an opportunity to understand how higher Na_2O and B_2O_3, found in LAW glasses, affect glass behavior in comparison to the Broborg hillfort glasses that contain little alkali and no boron. The naming scheme used for the join glasses was D*X*B*Y*N where *X* reflected the wt% addition of B_2O_3 and *Y* reflected the wt% addition Na_2O. Figure 1-2 provides the compositions of Broborg join glasses overlaid with a variety of LAW glass compositions. The join glasses were tested by EPA 1313 along with the LAW glasses to provide two distinct composition ranges for modeling of alteration behavior.

Table 1-2. Target Glass Compositions for LP3 (Lonergan et al. 2020) and join glasses in mass fraction.

Sample	Al_2O_3	B_2O_3	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SiO_2	SnO_2	V_2O_5	ZnO	ZrO_2	MoO_3	Re_2O_7	Others*	Total
LAWPh3-01-1	0.0658	0.1351	0.0446	0.0054	0.0068	0.0124	0.2413	0.3558	0.0308	0.0028	0.0207	0.0598	0	0	0.0189	1.0000
LAWPh3-02	0.0687	0.1316	0.0467	0.0005	0.0289	0.0108	0.2271	0.3492	0.0155	0.0285	0.0309	0.0477	0	0	0.0139	1.0000
LAWPh3-03	0.0650	0.0662	0.0409	0.0140	0.0501	0.0118	0.2159	0.3996	0.0158	0.0025	0.0303	0.0536	0	0	0.0344	1.0000
LAWPh3-04	0.0636	0.1276	0.0952	0.0037	0.0090	0.0042	0.2221	0.3580	0.0035	0.0276	0.0224	0.0339	0	0	0.0293	1.0000
LAWPh3-05_mod6	0.0624	0.0850	0.0247	0.0106	0.0293	0.0114	0.2274	0.3953	0.0202	0.0330	0.0215	0.0406	0	0	0.0387	1.0001
LAWPh3-06	0.0902	0.0673	0.1016	0.0024	0.0239	0.0114	0.2127	0.3578	0.0325	0.0230	0.0290	0.0333	0	0	0.0150	1.0000
LAWPh3-07	0.0980	0.0761	0.0884	0.0053	0.0078	0.0007	0.2389	0.3506	0.0073	0.0305	0.0310	0.0326	0	0	0.0328	1.0000
LAWPh3-08	0.0667	0.0648	0.0614	0.0011	0.0152	0.0019	0.2124	0.4399	0.0057	0.0005	0.0329	0.0629	0	0	0.0347	1.0000
LAWPh3-09	0.0782	0.0860	0.0923	0.0084	0.0550	0.0021	0.2129	0.3551	0.0024	0.0064	0.0229	0.0509	0	0	0.0275	1.0000
LAWPh3-10	0.0825	0.0636	0.0558	0.0079	0.0263	0.0034	0.2359	0.3550	0.0172	0.0384	0.0319	0.0621	0	0	0.0201	1.0000
LAWPh3-12	0.1112	0.1351	0.0276	0.0107	0.0023	0.0126	0.2143	0.3514	0.0093	0.0385	0.0279	0.0334	0	0	0.0257	1.0000
LAWPh3-14	0.0663	0.0826	0.0530	0.0003	0.0021	0.0009	0.2564	0.3956	0.0217	0.0214	0.0248	0.0377	0	0	0.0372	1.0000
LAWPh3-15	0.0730	0.0943	0.0867	0.0125	0.0414	0.0001	0.2171	0.3534	0.0169	0.0160	0.0294	0.0450	0	0	0.0142	1.0000
LAWPh3-16	0.0611	0.0692	0.0742	0.0033	0.0408	0.0079	0.2320	0.3656	0.0012	0.0362	0.0343	0.0571	0	0	0.0171	1.0000
LAWPh3-19	0.0670	0.0689	0.0849	0.0136	0.0308	0.0021	0.2143	0.4082	0.0108	0.0291	0.0211	0.0306	0	0	0.0186	1.0000
LAWPh3-20	0.0604	0.0893	0.0476	0.0112	0.0013	0.0007	0.2243	0.4120	0.0286	0.0364	0.0339	0.0353	0	0	0.0190	1.0000
Dike	0.1272	0.0000	0.0870	0.1278	0.0138	0.0665	0.0277	0.5348	0	0	0	0	0.0000	0.0000	0.0151	1.0000
D2B	0.1114	0.0165	0.0867	0.1253	0.0103	0.0860	0.0201	0.5282	0	0	0	0	0.0016	0.0000	0.0138	1.0000
D6B	0.1017	0.0557	0.0811	0.1163	0.0093	0.0795	0.0190	0.5231	0	0	0	0	0.0015	0.0000	0.0127	1.0000
D6B10N	0.0953	0.0527	0.0746	0.1067	0.0090	0.0730	0.0976	0.4770	0	0	0	0	0.0016	0.0006	0.0120	1.0000
D6B14N	0.0901	0.0492	0.0707	0.1007	0.0085	0.0696	0.1370	0.4608	0	0	0	0	0.0014	0.0004	0.0116	1.0000
D6B20N	0.0884	0.0484	0.0653	0.0921	0.0078	0.0673	0.1955	0.4219	0	0	0	0	0.0016	0.0005	0.0111	1.0000
D10B20N	0.0827	0.0887	0.0608	0.0854	0.0075	0.0620	0.1930	0.4088	0	0	0	0	0.0011	0.0002	0.0100	1.0000

*Others include Cl, Cr_2O_3, F, MoO_3, MnO_2, TiO_2 SO_3, and P_2O_5

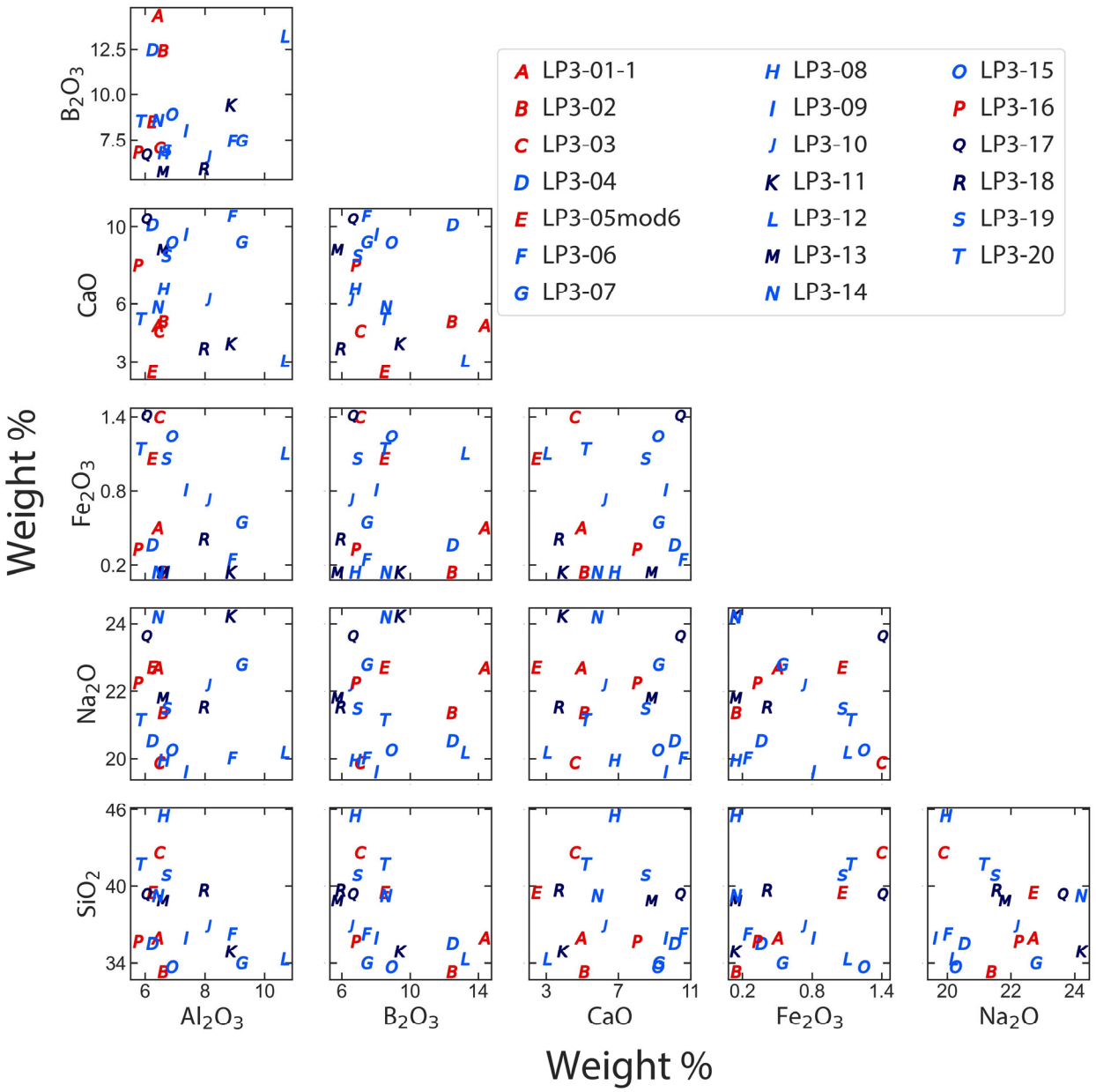

Figure 1-1. Scatterplot matrix of major elements in the LP3 compositional matrix. Glasses LP3-11, -13, -17, and -18 were not tested in the present study but compositions were taken from (Lonergan et al. 2020) are shown in dark blue. Blue symbols denote glasses are below the 2 g m^{-2} PCT response limit, red symbols denote glasses are above the PCT response limit.

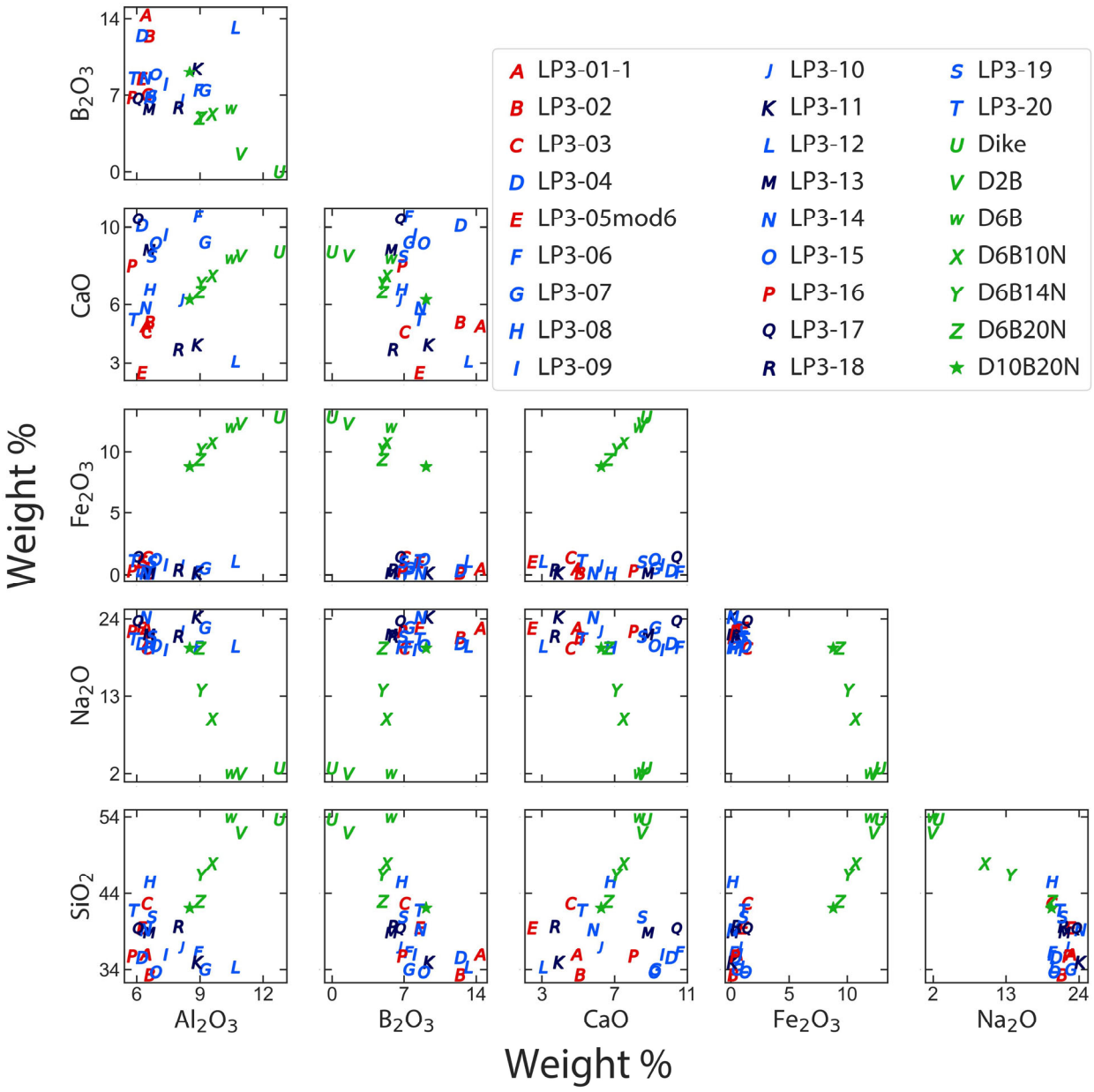

Figure 1-2. Scatterplot matrix of major elements in the LP3 compositional matrix with the "join" glasses. Glasses LP3-11, -13, -17, and -18 were not tested in the present study but compositions were taken from (Lonergan et al. 2020) are shown in dark blue. Blue symbols denote glasses are below the 2 g m^{-2} PCT response limit, red symbols denote glasses are above the PCT response limit, and green symbols denote join glasses

Table 1-3. Tabulated PCT and VHT responses for quenched LP3 glasses. The response limits for VHT and PCT are 50 g m^{-2} d^{-1} and 2 g m^{-2}, respectively (Lonergan et al. 2020). Similar information is not available for the join glasses.

Glass ID	Alteration rate (VHT) g m^{-2} d^{-1}	PCT-Na normalized release g m^{-2}
LAWPh3-01-1	>68.3	3.734
LAWPh3-02	>77.8	6.559
LAWPh3-03	>80.0	3.257
LAWPh3-04	30.1	1.628
LAWPh3-05-mod6	241	5.190
LAWPh3-06	6.48	0.993
LAWPh3-07	>85.5	1.168
LAWPh3-08	0.59	0.654
LAWPh3-09	55.7	1.653
LAWPh3-10	48.2	1.234
LAWPh3-12	78.6	0.960
LAWPh3-14	54.2	1.320
LAWPh3-15	33.5	1.475
LAWPh3-16	207	3.408
LAWPh3-19_mod1	38.9	1.096
LAWPh3-20	8.90	0.741

1.3 PNNL QA Program

This work was performed in accordance with the PNNL Nuclear Quality Assurance Program (NQAP). The NQAP complies with the United States Department of Energy Order 414.1D, Quality Assurance, and 10 CFR 830 Subpart A, Quality Assurance Requirements. The NQAP uses NQA-1-2012, Quality Assurance Requirements for Nuclear Facility Application as its consensus standard and NQA-1-2012, Subpart 4.2.1, as the basis for its graded approach to quality.

The NQAP works in conjunction with PNNL's laboratory-level Quality Management Program, which is based upon the requirements as defined in the United States Department of Energy (DOE) Order 414.1D, Quality Assurance, and 10 CFR 830, Nuclear Safety Management, Subpart A, Quality Assurance Requirements.

The information contained in this report should not be used as design input or operating parameters without additional qualification.

2.0 Test Methods

This section describes how the data were obtained for the sixteen LP3 glasses, the Broborg analogue glass, and seven join glasses that bridge the compositional gap between LAW glass and the Broborg analogue glass. The descriptions include the EPA 1313 methods, solution analysis, and sample imaging and chemical mapping methods. Glasses were melted and characterized previously (Lonergan et al. 2020). Tests were performed in duplicate for LP3-04, LP3-08, LP3-10, LP3-15, and LP3-16.

2.1 EPA Method 1313

The EPA 1313 is a standardized test developed by the EPA that has been modified as part of the ORP program in order to develop a method to assess leaching of LAW glasses. The EPA 1313 method and the modifications will be described in the subsequent sections.

2.1.1 Modifications to EPA 1313

Through the ORP program, the procedure for the EPA 1313 was altered for use with LAW glasses The following modifications were performed to the method:

1. Reduced mass of sieved and washed glass powder for each trial from 10 g to 1 g.
2. Reduced volume of solution per each trial to 10 mL.
3. Reduced number of target pH values to 12, 10, 9, 8, 7, 5.5.

Modification 1 was performed to reduce the sample mass and modification 2 was performed to maintain a glass mass to solution volume ratio of 1:10 (as specified in the procedure). These two modifications greatly reduced the time for sample preparation, particularly since a significant amount of material is lost during the size reduction. Modification 3 reduced the number of samples needed, the number of titrations required, and the amount of laboratory consumables (testing vessels, syringes, chemicals, etc.) required to perform the EPA 1313 test. If performed with hazardous or radiological materials, these modifications would also limit the radiological exposure time. Although the number of pH values tested was reduced, the range chosen is relevant to the conditions within nuclear waste disposal facilities, such as the IDF (Zhang et al. 2013).

2.1.2 EPA 1313 Procedure

Quenched glasses were used for EPA 1313 following the standard PCT protocol (ASTM 2021). EPA 1313 constrains the particle sizes to 75-150 µm, which is the same particle size range as required by the PCT protocol. The EPA 1313 test used here involves placing 1 g quantities of prepared glass powder in vials containing 10 mL of solution prepared with various amounts of acid. The initial step is a “titration” step where the amount of acid equivalents needed to achieve desired final pH values are determined. The number of acid equivalents required is dependent on the glass composition. Once the number of acid equivalents has been determined, a final set of tests are conducted to achieve ±0.5 pH units of 12, 10, 9 ,8, 7, or 5.5. In addition, a “natural pH” test is conducted where the material is only contacted with deionized water. Details of EPA 1313 are provided by Thorpe et al. (Thorpe et al. 2018, Thorpe et al. 2021).

The steps of the standard EPA 1313 test method are as follows:

Glass preparation

1. Glass was crushed in a tungsten carbide mill.

2. Crushed glass powder was sieved using -100 +200 mesh (75-150 µm) sieves.
3. Sieved glass powders were washed with deionized water three times by swirling the water and glass in a beaker then using an ultrasonic cleaner until the water was no longer cloudy.
4. After washing with deionized water, powders were washed with absolute ethanol three times by swirling the ethanol and glass in a beaker by hand then using an ultrasonic cleaner until the solution was no longer cloudy.
5. The glass powders were dried in an oven at 90 °C for 4-12 hours.
6. Glass powder was weighed into 1.0000 ± 0.0005 g samples.

Solution preparation

7. 10 mL solutions were prepared by mixing deionized water and either 1 M HNO_3 or 1 M KOH to -0.0381, 0, 0.0005, 0.001, 0.003, 0.0075, 0.15, and 0.30 M acid equivalents where a negative acid equivalent value represents a basic solution (prepared by diluting 1 M KOH in deionized water), a value of zero indicates that deionized water was used, and a positive value represents an acidic solution (prepared by diluting 1 M HNO_3 in deionized water).

Initial titration

8. An initial titration was performed by combining the 1.0 g glass powder samples with the 10 mL -0.0381, 0, 0.0005, 0.001, 0.003, 0.0075, 0.15, and 0.30 M solutions in polypropylene bottles. This mixture of 1 g of glass (sized to between 75 and 150 µm) and 10 mL of solution results in a surface area to solution volume (*S/V*) ratio of roughly 2000 m^{-1} (assuming a spherical particle geometry).
9. The glass-solution systems were tumbled for 48 hours end-over-end at 28 ± 2 revolutions per minute at room temperature. Sample masses were recorded prior to tumbling.
10. After 48 hours, systems were removed from the tumbler, weighed, and the powder was allowed to settle for 15 minutes.
11. The pH of the eluate was measured using an electronic pH probe, calibrated with 4 pH buffers (range 4-11.45) during the same day.
12. A titration curve of eluate pH (after tumbling for 48 hours) versus equivalents of acid was prepared to help interpolate the necessary acid or base additions to achieve the target pH of 12, 10, 9 ,8, 7, or 5.5 after tumbling each sample for 48 hours.
13. Titrations were repeated until the pH of the eluates were within ±0.5 pH units of 12, 10, 9, 8, 7, or 5.5.
14. Once the target pH values were achieved, the necessary acid/base additions were used according to the following steps.

Final EPA 1313 test

15. 1 g of glass (sized to between 75 and 150 µm) was added to10-mL solutions made with the acid volumes from Step 14.
16. The glass-solution systems were placed on an end-over-end tumbler set at 28 ± 2 revolutions per minute.
17. After tumbling for 48 hours, the powder was allowed to settle for 15 minutes then 1-2 mL of solution was removed, placed in a separate container, and the pH of the 1-2 mL aliquots were measured.
18. The remaining 8-9 mL of eluates were syringe filtered using a 0.45 µm pore size syringe filter.
19. Filtered eluates were acidified to make ~0.1 M HNO_3 solution and refrigerated until solution analysis.

20. The glass powders were washed with deionized water, then with ethanol, and dried at 90 °C for 12 hours.
21. Solution analysis using inductively inductively coupled plasma-optical emission spectroscopy (ICP-OES) and inductively couple plasma-mass spectroscopy (ICP-MS)

Elements Na, Ca, Al, Si, and B were measured by ICP-OES while Mo and Re were measured by ICP-MS) at Vanderbilt University. The ICP-OES analysis was carried out on an Agilent 5110 VDV (Agilent Technologies, Santa Clara, CA) and ICP-MS was carried out on a Perkin Elmer model NexION 2000B (Perkin Elmer Corporation, Waltham, MA). ICP-OES followed the EPA Method 6010D (SW-846) protocol and ICP-MS followed the EPA Method 6020B (SW-846) protocol.

The solution pH for a glass and deionized sample is referred to as the "natural pH". The normalized elemental release, NL_i (g m^{-2}), of element i was calculated using the following equation:

$$NL_i = \frac{C_i}{f_i \times (S/V)} \tag{2.3}$$

where C_i is the content of element i in solution (g m^{-3}), f_i is the mass fraction of element i in the sample prior to chemical alteration, and S/V is 2000 m^{-1}, as described above. An estimated equivalent thickness (E_{th}, m) can then be estimated with the following equation:

$$E_{th} = \frac{NL_B}{\rho} \tag{2.4}$$

Where ρ is the density of the glass in kg/m^3 (assumed to be 2.7 kg/m^3).

2.2 Electron Probe Microanalysis (EPMA)

The dried, post-reaction glass powders were mounted in epoxy and polished with 400, 600, 800, and 1200 grit SiC grinding paper then with 1 µm diamond paste. Polished samples were sputter coated with 2 nm Ir and imaged with scanning electron microscopy (SEM) and electron probe microanalyses (EPMA). The polished powder cross sections were analyzed using a JEOL 8530F EPMA operated in backscatter electron (BSE) mode with the measurement parameters of 15 kV accelerating voltage and a 10 nA probe current.

3.0 Results and Discussion

This section describes the test results for the EPA 1313 for the LP3 glasses, the Broborg analogue and the join glasses, and a comparison is made to VHT and PCT results. The aim of the comparison is to see if there are similar compositional dependencies for the EPA 1313 as there is for VHT and PCT. The relationship between acid addition and pH is described in Section 3.1. The normalized elemental releases (NL_i) from LAW glasses, the Broborg analogue and the join glasses after the EPA 1313 test are summarized in Section 3.2. Comparing the results between PCT and VHT methods is given in Section 3.3. The impact of LAW glass composition on NL_i during the EPA 1313 test is modeled in Section 3.4. The effect of acid equivalent on NL_i is given in Section 3.5.

3.1 Relationship between EPA 1313 pH and acid addition

In this section, a compositional mixed linear model, designed using artificial neural network (ANN) models, is presented. The compositional model was used to calculate the titration curves for the sixteen LP3 glasses. Appendix A further elaborates on the ANN models used to understand the relationship of the titration curves and the glass compositions. The general shape of the titration curves is a sigmoid, which was approximated using polynomial functions. The relationship of acid/base addition and the final pH after tumbling for 48 hours is summarized in Appendix A. Values for elemental releases are tabulated in Appendix B.

From Figure A-3 and Figure A-5, it was apparent that the model coefficients vary exponentially with composition; therefore, a mixed linear composition-property model was fit to data of the form:

$$pH = \left(\frac{E}{A}\right)^{1/5} + B \qquad (3.1.1)$$

$$A = \sum A_i 10\hat{}(x_i) \qquad (3.1.2)$$

$$B = \sum B_i 10\hat{}(x_i) \qquad (3.1.3)$$

where E is the acid equivalents (in units of Eq./g-dry material), and x_i is composition in mole fraction, A_i and B_i are fitting coefficients for oxide component, i.e., a positive acid equivalent value indicates that an acidic solution was added to a sample whereas a negative value indicates that a basic solution was added to a sample. The fitting results from using the mixed linear model are shown in Figure 3-1, trace plots of the coefficients are shown in Figure 3-2, and the coefficients are listed in Table 3-1. A 20% validation split was performed prior to fitting. The mixed linear model provides an improved R^2 correlation coefficient and an improved root mean squared error (RMSE) over the ANN models with fewer fitting coefficients.

In Figure 3-2, the trace plots for Al_2O_3 and SiO_2 show that the amount of acid addition required to achieve a final target pH is slightly higher for lower fractions of Al_2O_3 and SiO_2 in glass. The opposite is observed for Na_2O and B_2O_3 where the required acid addition required to achieve a final target pH is slightly higher for higher fractions of Na_2O and B_2O_3 in glass. However, large variations were observed for these trace plots, represented by the shaded areas in Figure 3-2.

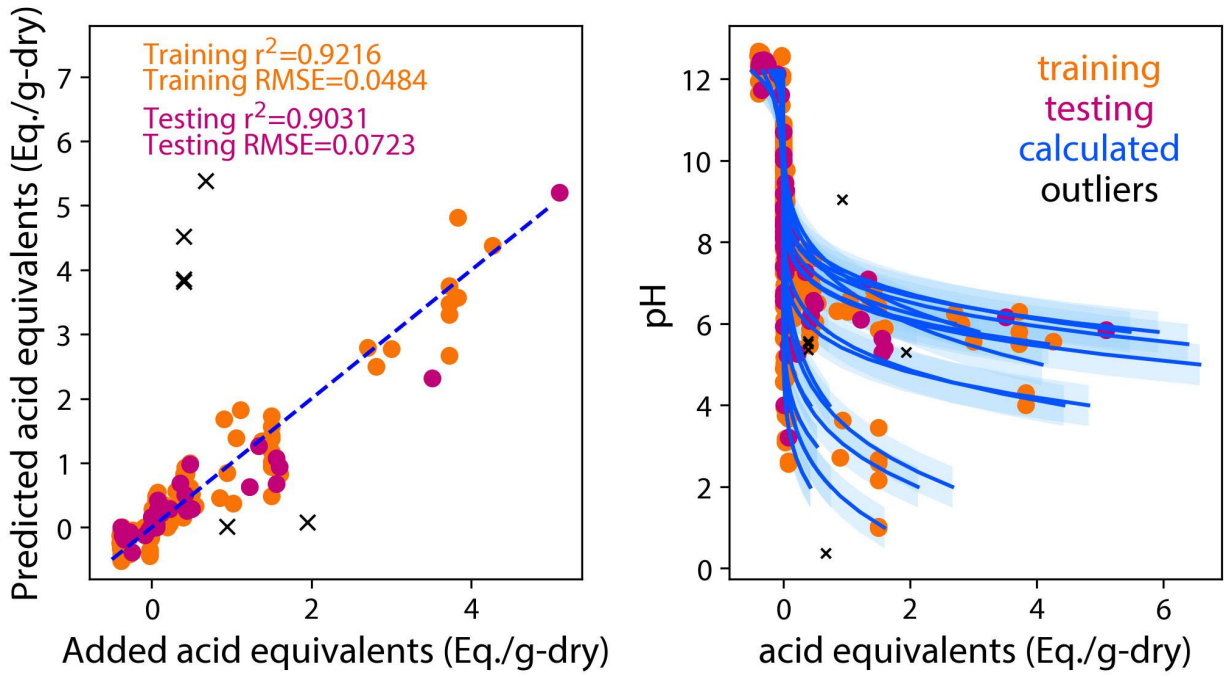

Figure 3-1. (left) comparison of measured acid equivalents to predicted acid equivalents from the mixed linear compositional model and (right) titration curves for LP3 and join glasses and join glasses, the shaded region shows the fit curve with a ±0.5 pH unit offset.

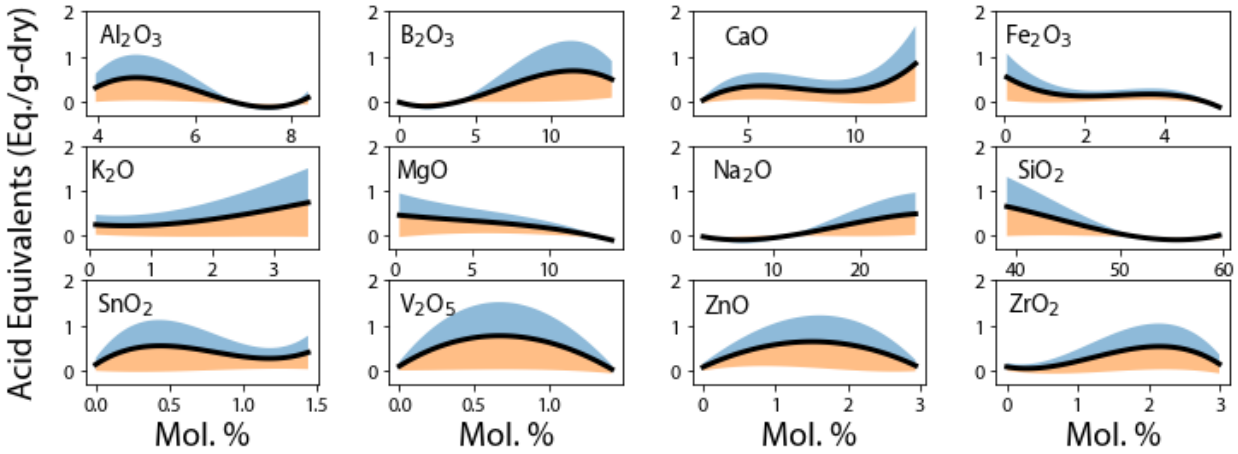

Figure 3-2. Trace plots demonstrating component effects on predicted acid equivalents. The 99% confidence interval (which combines the statistical variation of the standard deviation and the number of samples per data point, n=22) is shown as shaded areas above and below the lines.

Table 3-1. Mixed linear composition model coefficients

Component	A_i	B_i
Al_2O_3	0	20.24
B_2O_3	-1.94E-04	12.48
CaO	-7.28E-04	11.02
Fe_2O_3	1.08E-03	-51.36
K_2O	-1.64E-03	21.84
MgO	0	27.72
Na_2O	0	22.26
SiO_2	0	6.52
SnO_2	3.04E-03	0
V_2O_5	-1.47E-03	-51.89
ZnO	0	-26.37
ZrO_2	0	-15.10

3.2 Normalized elemental releases from EPA 1313 testing of LP3 glasses

The normalized elemental releases for the sixteen LP3 glasses after EPA 1313 are shown in Figure 3-3, which compares the NL_i values for i = Al, B, Ca, Na, and Si. Tables of NL_i values are compiled in Appendix B. The values of NL_B, NL_{Ca}, and NL_{Na} are similar for most glasses across most of the pH range. However, NL_{Ca} tends to be lower above pH 10. NL_{Al} and NL_{Si} are generally lower, particularly at lower pH. Figure 3-5 shows the pairwise trends in distribution of elemental releases from the LP3 glasses, the Broborg analogue and the join glasses after EPA 1313 testing. Based on Figure 3-5, the behavior of most glasses is similar such that the normalized elemental releases are higher for the pH 5.5 and pH 12 tests than exhibited by the natural pH tests. These trends are similar to those found for longer-term, higher-temperature static tests (see Reiser et al. (2021) for example). For LP3 glasses, the normalized elemental releases of Na and B were highest at pH < 6 and had a minimum at 10 ≤ pH < 12. The NL_{Al}, however, exhibits a minimum near pH 8. Furthermore, Figure 3-5 shows that the normalized releases of certain elements such as Na, Ca, and B are highly correlated.

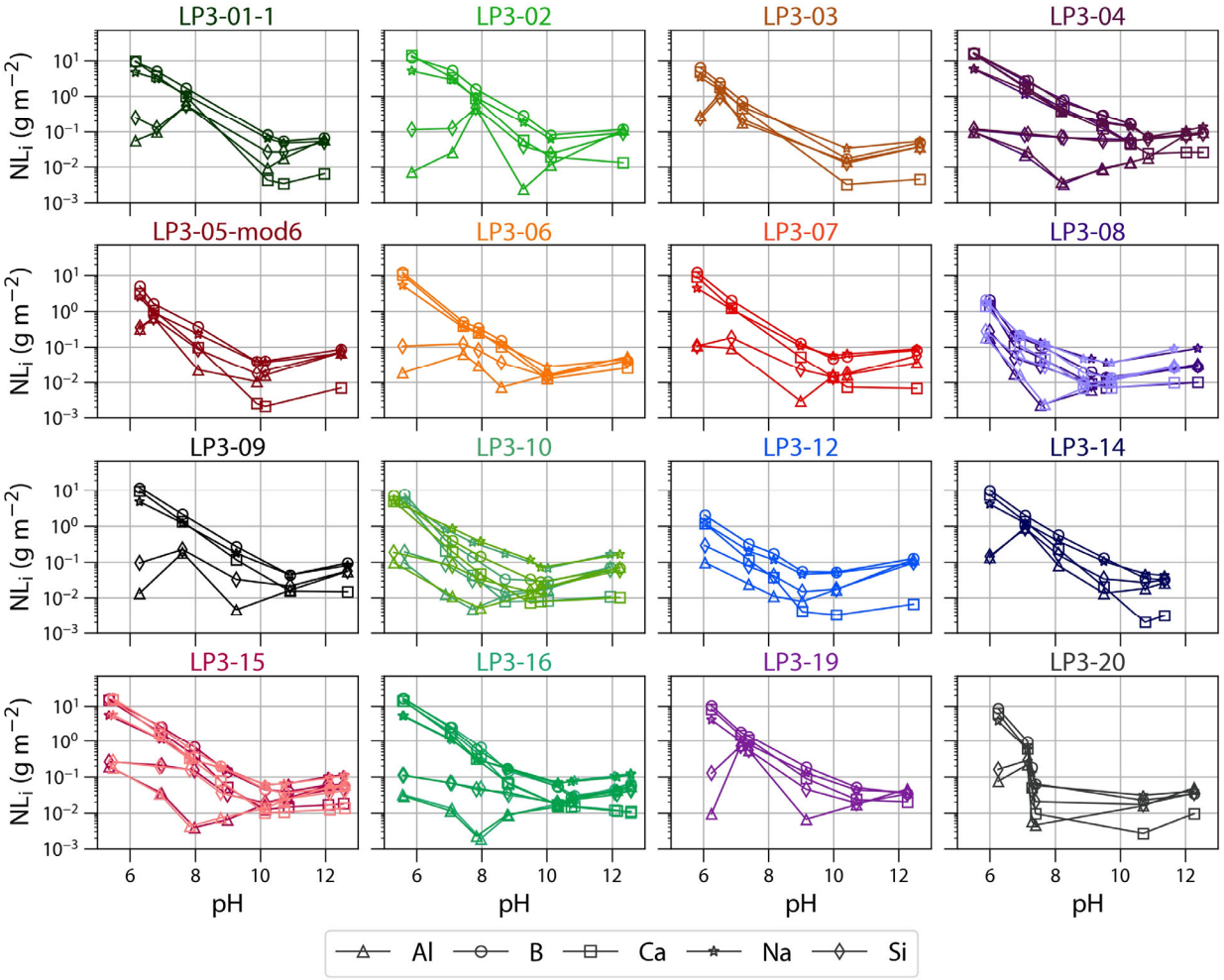

Figure 3-3. EPA 1313 normalized elemental releases for 16 LP3 glasses

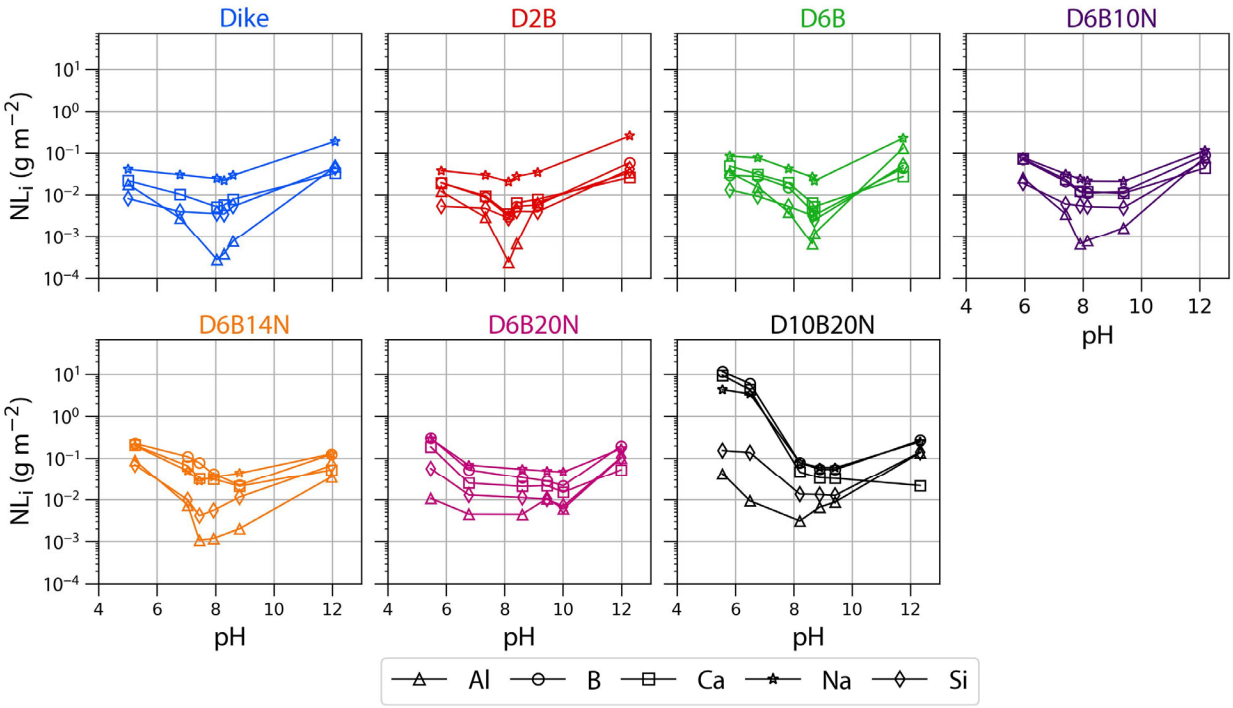

Figure 3-4. EPA 1313 normalized elemental releases for join glasses

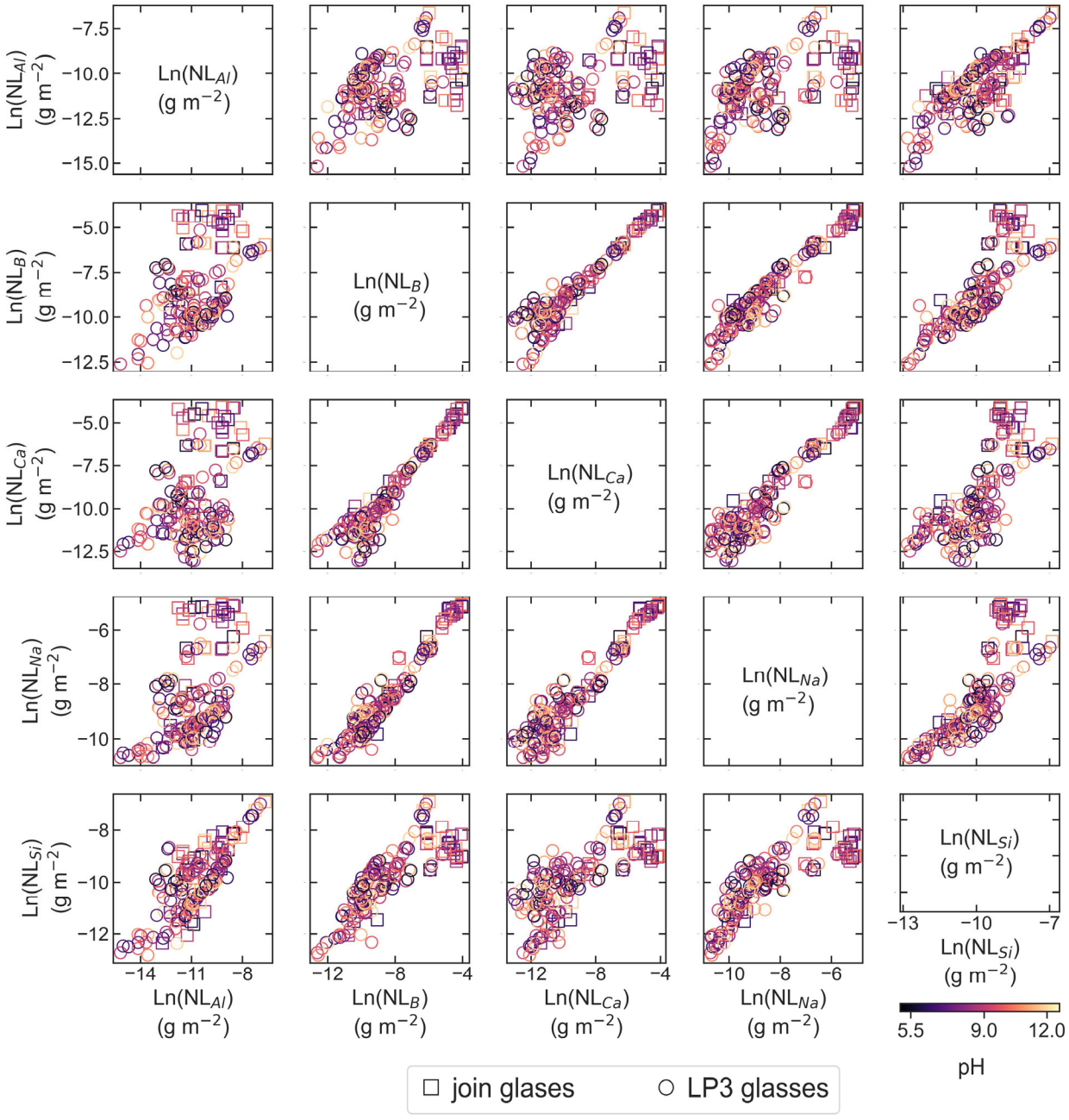

Figure 3-5. Pairwise distribution of the natural log of normalized elemental releases for LP3 and join glasses. In this figure, LP3 glasses are shown using circles and join glasses are shown using squares. The color of each data point represents the pH of the final solution, as indicated with the color bar on the bottom right

3.3 Comparison of Durability Test Results for LP3 Glasses

This section compares EPA 1313 results with the previously published LP3 VHT and PCT results (Lonergan et al. 2020) in order to see if relative releases correspond with VHT rates and PCT releases and to determine if the three tests result in the same alteration mechanism. No similar comparison is available for the join glasses as PCT and VHT have not been conducted with these glasses. In order to compare

results from the three tests, dissolution rates in µm yr^{-1} were determined based on conversion of normalized release values to equivalent layer thicknesses for EPA 1313 and PCT results. The dissolution rate for the VHT was determined through a direct measurement of the altered coupon. The values used to determine the rates are tabulated in Table 1-3, Table B-1, and Table B-2.

For the lowest pH samples of the room temperature EPA 1313 (pH ≈ 5.5), the calculated alteration rates exceed those observed in the 90 °C PCT (see Figure 3-6). As the target pH increases, the calculated alteration rates in EPA 1313 fall below that which was calculated for the PCT. The measured room temperature pH of the solutions in the PCT tended to be about 10. As a result of the lower temperature, the alteration rates at final pH ≈ 10 in the EPA 1313 was much lower for the same pH as the PCT. The lower temperature of the EPA 1313 is closer to the average temperature of the IDF on the Hanford site (~15°C) than the PCT or VHT.

After converting dissolution rates to natural log in µm yr^{-1}, the test responses at the different temperatures were compared on an Arrhenius plot (Figure 3-6). Other authors have used Arrhenius plots to compare alteration measurements performed under different timescales (Neeway et al. 2018, Nava-Farias et al. 2021). For all cases, the alteration depths observed by VHT were significantly higher than those observed during EPA 1313 at pH values above pH=7 (see Figure 3-6). This is expected as the VHT is a 200 °C-test. Glasses LAWPh3-01-1, LAWPh3-02, LAWPh3-03, LAWPh3-05mod6 and LAWPh3-16, which failed the PCT and VHT requirements (2.0 g/m^2 and 50 g/m^2/d, respectively) also exhibited a large degree of normalized Na release during EPA 1313 testing at final pH = 5.5. Glass LAWPh3-08, which exhibited the lowest VHT alteration rate and PCT normalized Na release, also exhibited the among the lowest EPA 1313 NL_{Na} values.

The apparent activation energies (E_a), calculated based on the slope of the Arrhenius plots, ranged from 29 kJ mol^{-1} to 63 kJ mol^{-1} (Figure 3-6). An apparent E_a value (47 ± 5 kJ mol^{-1}) within this range was also found for a synthetic Broborg glass in a similar comparison of VHT, PCT, and EPA 1313 results (Nava-Farias et al. 2021) However, these values are lower than the range of apparent E_a values (81 kJ mol^{-1} to 90 kJ mol^{-1}) found for three high level waste glass compositions tested using single pass flow through (SPFT) (Neeway et al. 2018). Neeway et al. (2018) concluded that this range of E_a values is associated with the breaking of the Si-O-Si bond, as it is too high to be associated with a diffusion-controlled process, with apparent E_a values reported to be 30 kJ mol^{-1} to 50 kJ mol^{-1}. Neeway at al. (2018) also compared their results with those for several other glass compositions, tested using different methodologies, and this resulted in a larger range of apparent E_a values (53 kJ mol^{-1} to 90 kJ mol^{-1}), however, variations in glass dissolution rate measurements due to the different test methodologies could not be ruled out. In this report, a larger set of glass compositions was tested using the VHT, PCT, and EPA 1313 test methodologies, and the lower range of apparent E_a values suggests that the rate-controlling mechanism of glass dissolution may not be the breaking of the Si-O-Si bond, with diffusion-controlled processes potentially playing a role. In addition, the range of apparent E_a values found for glasses of different compositions suggests that the rate-controlling mechanism of glass dissolution may be composition dependent. Identifying the rate-controlling mechanism is not necessary for use of the EPA1313 standard test, but a mechanistic understanding of how the glass is altered under the test conditions increases the technical defensibility for using the test to predict the behavior of the glass under disposal conditions. Further examination of the alteration layers on the solid phases by techniques such as time-of-flight secondary ion mass spectrometry (TOF-SIMS), may aid in understanding any observed differences in the test methods.

In addition to the Arrhenius plots, the calculated equivalent thickness (E_{th}) from EPA 1313 was compared with E_{th} from EPMA measurements on cross sections of the altered glass material. In Figure B-1, the E_{th} values calculated from the NL_B at pH ≈ 5.5 are compared with E_{th} values determine by measuring the thickness of a chemically and morphologically different alteration layer on the glass surface in EPMA

images (from Appendix C) for specific glasses (LAWPh3-01-1, LAWPh3-03, LAWPh3-06, LAWPh3-07, LAWPh3-08, LAWPh3-09, LAWPh3-10, and LAWPh3-20). This pH was selected as it resulted in the highest NL_i values and, by extension, the largest layer thicknesses for measurement by EPMA. The calculated E_{th} values generally matched the trends in thickness from the examined cross sections. The EPMA images for the glasses altered at pH 5.5 show a depletion in Na in the alterations layer, suggesting incongruent dissolution and ion exchange processes may be taking place at low pH, but the alteration mechanisms will likely change as a function of increasing pH.

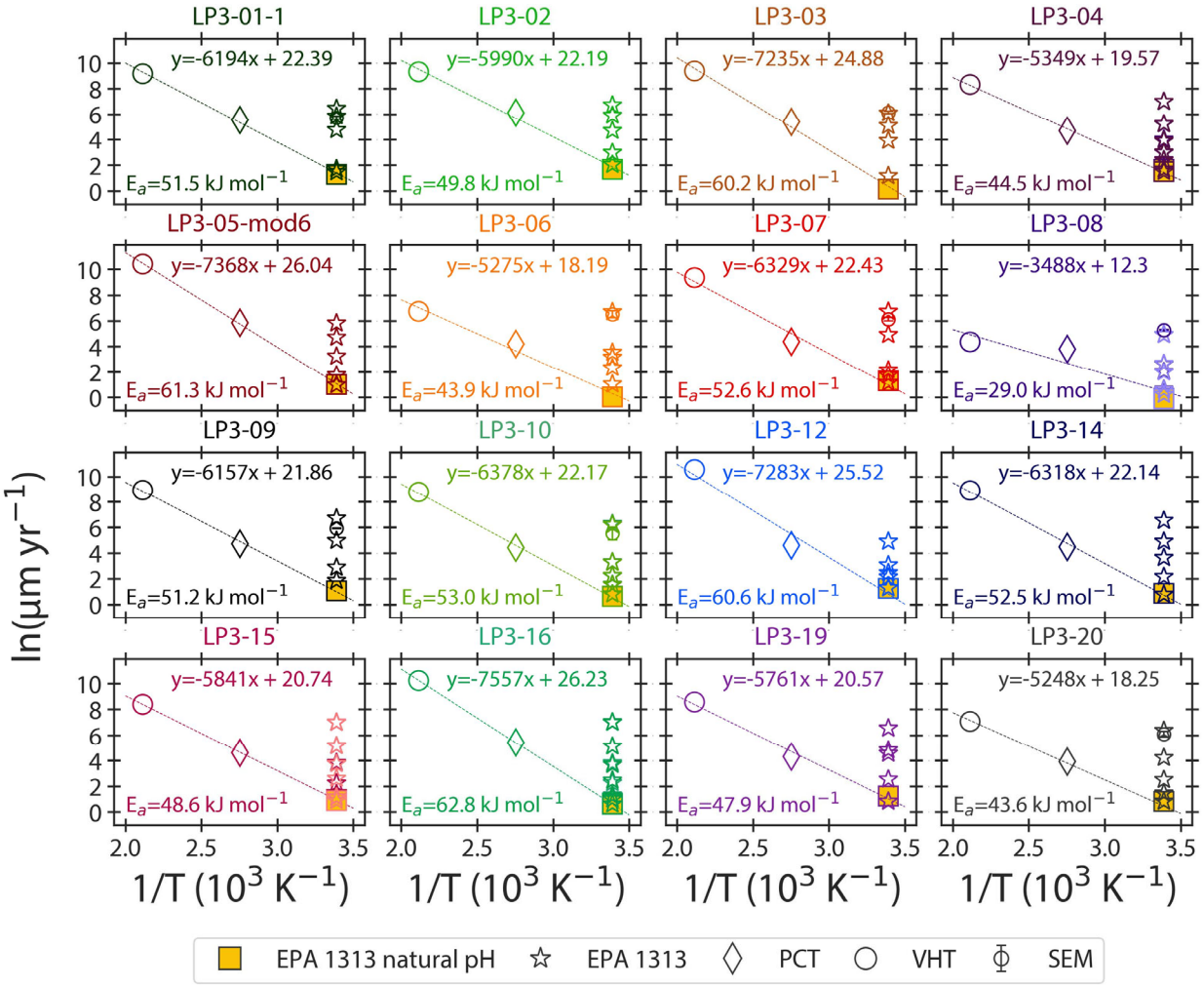

Figure 3-6. Comparison of alteration rates from PCT, VHT, and EPA 1313 of LP3 glasses (PCT and VHT were not performed on join glasses). In this figure the 'natural pH' refers to the alteration with no acid or base added (acid equivalents equal zero). A line was fit through the VHT data, PCT data, and natural pH point in the EPA 1313.

3.4 Compositional modeling – elemental release

The normalized elemental releases for Na and B were fit based on the general alteration rate equation proposed by Strachan (2017):

$$r = k_i \left[\exp\left(-\frac{E_{aH^+}}{RT}\right) a_H^{\eta_a} + \exp\left(-\frac{E_{aH_2O}}{RT}\right) + \exp\left(-\frac{E_{aOH^-}}{RT}\right) a_{OH}^{\eta_b} \right] \quad (3.4.1)$$

where, r is the glass dissolution rate (g m^{-2} d^{-1}), k_i is the intrinsic rate constant (g m^{-2} d^{-1}), a_H is the activity of the hydronium ion (unitless, where a_H=10^[pH]), a_{OH} is the activity of the hydroxide ion (unitless, where a_{OH}=10^[pOH]), η_a is the reaction order for the hydronium ion (unitless), η_b is the reaction order for hydroxide (unitless), E_{aH+} is the activation energy for the dissolution in an acidic solution (J mol^{-1}), E_{aH2O} is the activation energy for the dissolution in a neutral solution (J mol^{-1}), E_{aOH-} is the activation energy for the dissolution in a basic solution (J mol^{-1}), R in the denominator is the gas law constant (J mol^{-1} K^{-1}), and T is temperature (K).

Since EPA 1313 is performed at a constant temperature and a constant time, equation. 3.4.1 can be rewritten as:

$$\ln(NL_i) = a + b\mathrm{pH} + c\mathrm{pOH} \quad (3.4.2)$$

where a, b, and c are fit constants that are nominally equal to $\ln(k_i)\cdot E_{aH2O}/RT$, $\ln(k_i)\cdot E_{aH+}/RT\cdot\eta_a$, and $\ln(k_i)\cdot E_{aOH-}/RT\cdot\eta_b$. We acknowledge that the activities of the various water species are not fixed during the experiment so we cannot accurately capture the evolution of the η terms.

Based on the general alteration rate in equation 3.4.1, a simplified stepwise linear model was fit to the data where two linear profiles were fit to NL_i versus the end-of-experiment pH (one corresponding to the low pH range and the other to the high pH range), see Appendix D. The stepwise linear model in Appendix D accounted for compositional differences between elemental releases for LAW glasses, but did not account for the shift in minimization observed between join glasses and LAW glasses. The simplest form of the compositional effects can be represented as $a = \sum A_j x_j$ where x_j is the mole fraction of component j and b and c are composition independent. This would fit the data under the assumption that η_a and η_b are compositionally independent. Alternatively, if as suggested by Vienna et al (2018) t, k_i is composition independent, then composition effects can be expressed as:

$$\ln(NL_i) = a_0 + \mathbf{a}^{\mathbf{T}}\mathbf{x} + \begin{cases} b \cdot pH & if \quad pH < p \\ \mathbf{c}^{\mathbf{T}}\mathbf{x} \cdot pH & if \quad pH \geq p \end{cases} \quad (3.4.3)$$

where **a** and **c** are coefficient vectors, **x** is the composition vector (where the components are in mole fraction), a_0 is a constant, and p is the pH cutoff. Table 3-2 summarizes the fitting coefficients for NL_{Na} and NL_B for the trial including only LAW glasses and Table 3-3 summarizes the fitting coefficients for the trial including both LAW and join glasses. The overall fits for both trials are shown in Figure 3-7. In Figure 3-7, outliers are shown as red points, these values were identified during the least-squares fitting where outliers were datapoints whose square difference was greater than 2.95 from the mean value. A systematic argument cannot be made for why these values are outliers, however, these outliers were consistent among the fitting trials and were either LP3-20 at pH=6.25, LP3-20 at pH=7.29, LP3-20 at pH=7.40, D10B20N at pH=6.50, D6B20N at pH=5.47, D6B20N at pH=6.78, or D2B at pH=9.13, see Table 3-3.

Table 3-2. Linear model coefficients fit to ln(NL_i) data of LP3 glasses using Equation 3.4.3.

Component (mole fr.)	Na		B	
	a_i	c_j	a_j	c_j
Al_2O_3	-9.31	14.90	-36.52	27.95
B_2O_3	18.93	-16.10	13.72	-18.61
CaO	12.94	-7.99	0	-14.23
Fe_2O_3	0	0	0	0
K_2O	25.43	-23.05	36.48	-31.09
MgO	0	0	-8.28	0
Na_2O	27.85	-18.12	19.79	-24.34
SiO_2	7.44	-6.63	-5.90	-8.36
SnO_2	-10.58	0	0	0
V_2O_5	0	0	2.46	0
ZnO	0	0	-38.25	0
ZrO_2	0	0	-58.17	0
p	11.5		11.5	
a_0	-6.00		8.60	
b	-0.924		-1.162	
R^2	0.937		0.955	
RMSE	0.407		0.441	
Outliers	2		2	

Table 3-3. Linear model coefficients fit to ln(NL_i) data of LP3 and join glasses using Equation 3.4.3.

Component (mole fr.)	Na		B	
	a_i	c_j	a_j	c_j
Al_2O_3	-11.58	21.77	-34.49	32.22
B_2O_3	12.79	-15.92	16.15	-22.91
CaO	7.80	-10.34	0	-14.40
Fe_2O_3	0	0	0	0
K_2O	21.24	-25.63	37.27	-37.01
MgO	0	0	-12.96	0
Na_2O	9.57	-14.34	12.26	-19.37
SiO_2	0	-6.59	-4.23	-9.99
SnO_2	0	-52.75	0	0
V_2O_5	0	0	9.86	0
ZnO	0	0	-30.53	0
ZrO_2	0	0	-56.33	0
p	11.5		11.5	
a_0	1.918		8.94	
b	-0.844		-1.14	
R^2	0.884		0.940	
RMSE	0.543		0.520	
Outliers	2		4	

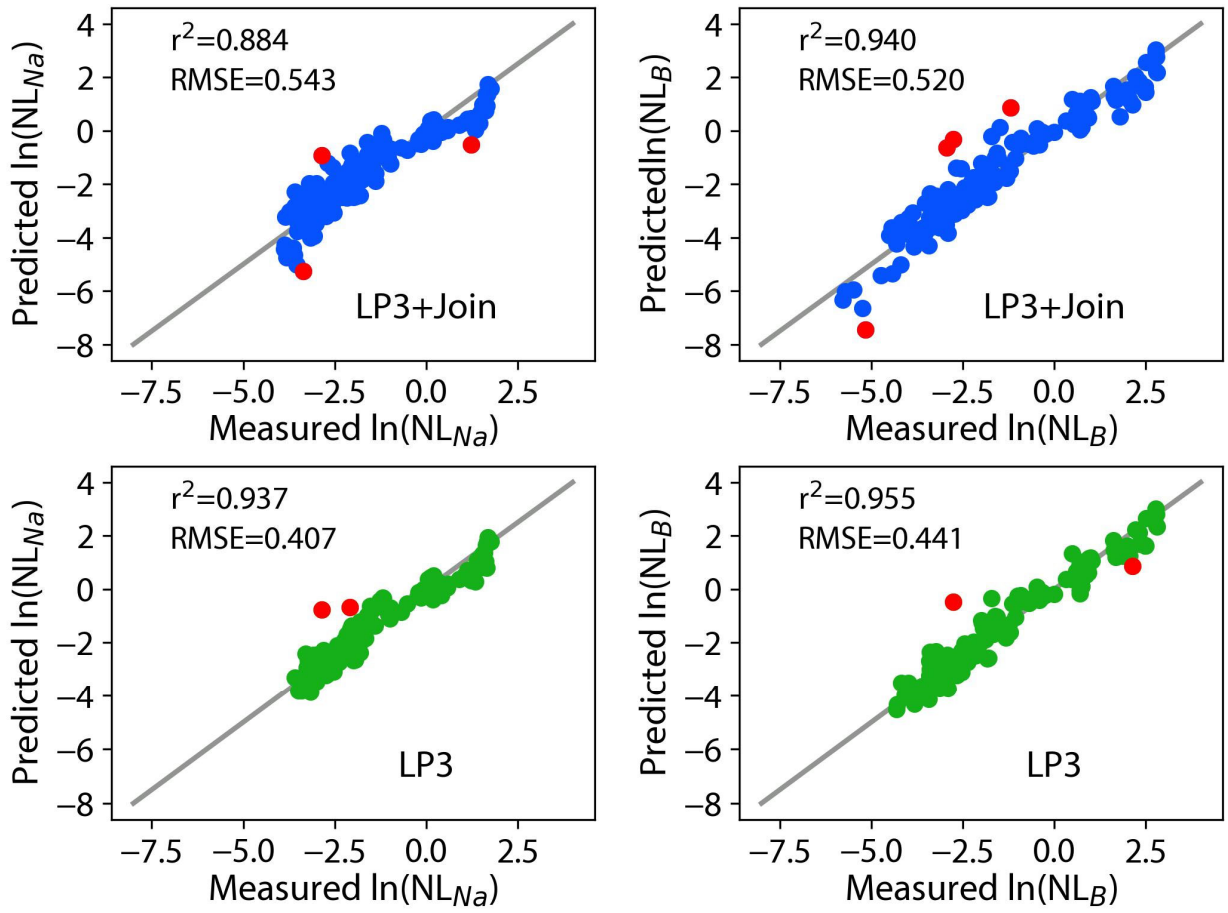

Figure 3-7. Comparison of measured versus calculated NL_{Na} values (left columns) and NL_B values (right columns) from EPA 1313 of LP3 glasses. The solid line represents the unity line. Outliers are shown in red

3.5 Effect of acidification on EPA 1313 responses

EPA 1313 requires the addition of various amounts of acid, depending on glass composition, to reach a target pH at the end of the experiment. To remove the influence of glass composition, the amount of acid added can be kept constant and the effects of the acid addition on the elemental release of glasses can be examined. As described in Section 3.1, the trends observed for the compositional effect on the amount of acid addition to achieve a target pH was subject to significant variation. This section describes the effect of acid addition on the resulting elemental release of glass during the EPA 1313, independent of glass composition.

To understand the glass response as a function of acid addition, the elemental releases of Na and B were plotted versus acid addition. Figure 3-8 shows a plot of the values for $\ln(NL_i)$ versus acid addition (in Eq./g-dry). All of the measured datapoints fall near a logarithmic curve which, when viewed in a plot of $\ln(NL_i)$ versus the decadic logarithm of acid addition, reveals only four outlier points (two for D10B20N and two for LAWPh3-20 near pH = 7). This suggests that there is a dominant effect of the acid concentration since ln(E) explains > 96% of the variation in $\ln(NL_i)$ independent of glass composition and therefore, the glass composition effects cannot amount to more than 4% of the variation in test response.

A similar trend was observed by Vienna et al. (2018) when considering composition effects on the alteration rate of select LAW glasses.

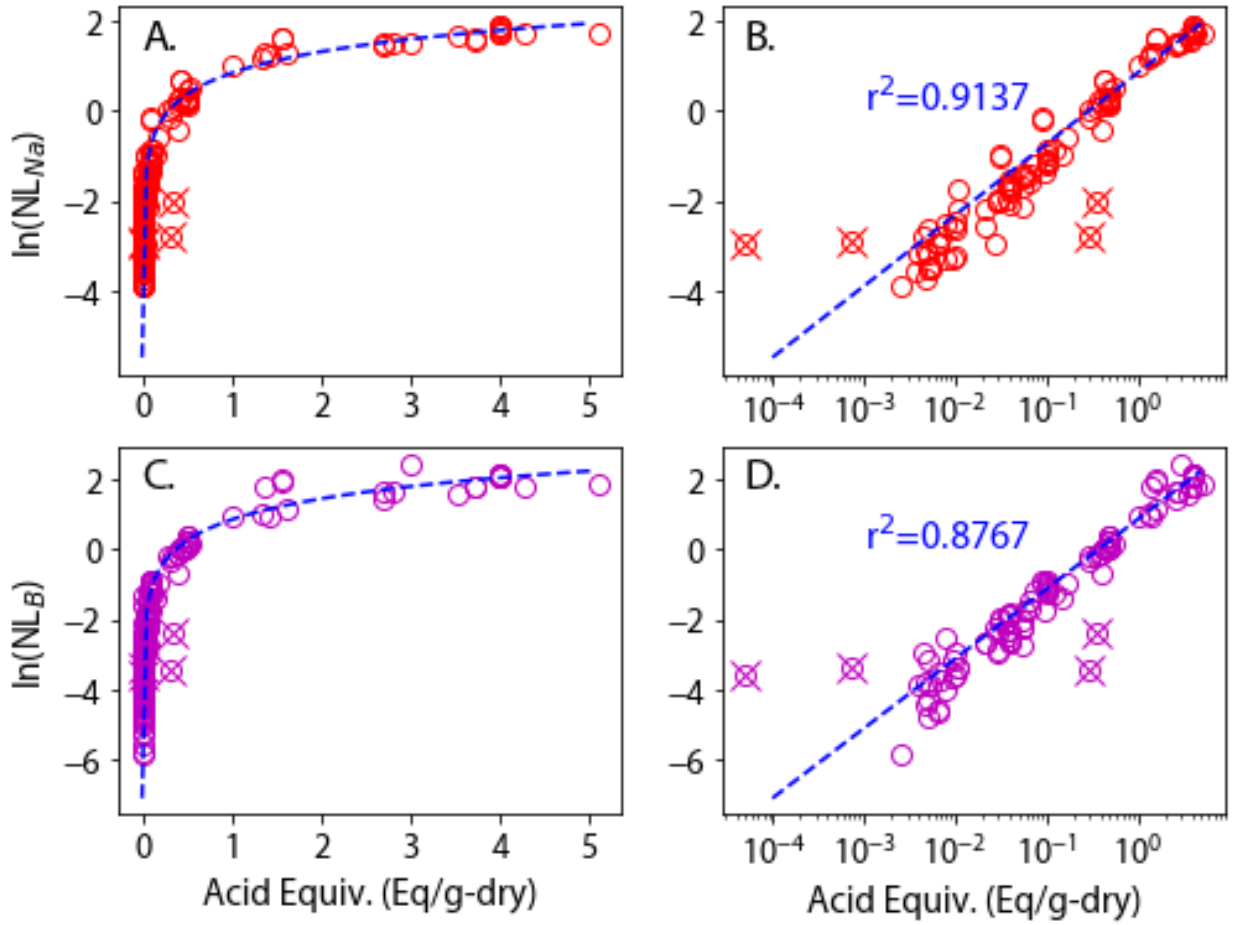

Figure 3-8. (A) $\ln(NL_{Na})$ values vs acid equivalents. (B) semilog plot of $\ln(NL_{Na})$ values versus the logarithm of acid addition. (C) $\ln(NL_B)$ values vs acid equivalents. (D) semilog plot of $\ln(NL_B)$ values versus the logarithm of acid addition. Outliers are shown with X's.

4.0 Summary

The objective of this report was to summarize data for EPA 1313 conducted on sixteen enhanced LAW glasses, named LP3 glasses, which had previously been subjected to rigorous characterization, and chemical durability testing by PCT and VHT. EPA 1313 was also conducted on a glass that is representative of vitrified material from the ~1500-year-old Broborg hillfort in Sweden, and seven glasses that bridge the compositional gap between LAW glass and the Broborg analogue glass.

At low end-of-experiment pH values (< 5.5) the extent of alteration from room temperature EPA 1313 testing in some cases exceeded that which was observed in 90 °C PCTs. However, for comparable end-of-experiment pH (near pH 10) the normalized degree of alteration observed during EPA 1313 testing was nearly an order of magnitude lower than PCT, and approximately two orders of magnitude lower than the VHT. This is expected as the EPA 1313 test is performed at a lower temperature (room temperature) than the PCT (90 °C) and VHT (200 °C). These results highlight the importance of evaluating pH when considering a test to assess the relative durability of a glass. Initial attempts at modeling composition and acid addition effects on normalized losses of Na, B, and Ca from EPA 1313 was promising. This modeling demonstrated that high concentrations of elements such as silicon reduced the release of all elements across the range of acid additions. Increased Si concentrations has also been shown to decrease the extent of glass dissolution in PCT and VHT. The EPA 1313, PCT and VHT test responses at the different temperatures were compared on an Arrhenius plot and used to determine the apparent activation energy for glass dissolution. The range of apparent activation energy values found suggests that the rate-controlling mechanism may not be the breaking of the Si-O-Si bond, and that diffusion-controlled processes could be playing a role in glass dissolution. In addition, the relatively wide range of apparent activation energy values found for these glasses of different composition suggests that the mechanism of glass dissolution is likely to be composition dependent.

The logarithm of acid addition was found to explain ~96% of the variation in logarithm normalized losses, suggesting that glass composition effects may play a secondary role in the EPA 1313 test response. Overall, that the LP3 glasses provide a closer range of elemental release values through EPA 1313 than the larger relative differences observed through PCT or VHT, suggesting that a future constraint could be defined that would allow broader compositional ranges for LAW glass formulations

4.1 Future work

Based on the findings outlined in this report, future work should aim to mitigate the large influence of acid addition on the elemental releases, and provide a better understanding of the compositional effect on elemental release in the EPA 1313 test. This could be achieved by performing EPA 1313 on a matrix of glasses and using fixed acid additions rather than fixed target pH. This would have the added benefit of eliminating the titration step.

Measurement of alteration layer thickness on the glass powders using electron microscopy was challenging due to the buildup of electrical charge on the epoxy that surrounds relatively small powder particles. Conducting EPA 1313 with glass monoliths may allow for more reliable characterization of alteration layers using electron microscopy. Future work should also evaluate the use of state-of-the-art solid phase characterization techniques to measure the alteration layer chemistry with higher resolution. Development of these techniques would allow the alteration behavior in the EPA 1313 test to be compared with alteration of natural and/or man-made ancient analogs under conditions similar to those in IDF disposal.

Future EPA 1313 testing should investigate the effect of solution composition on the normalized elemental release, e.g., by the addition of dissolved species to reproduce the porewater chemistry at the IDF. This guarantees that the effect of porewater constituents, including the solution concentration of Si, on the response to EPA1313 to be understood.

The ultimate goal of studying the Hanford LAW glass response to EPA 1313 is to assess the chemical durability of the wasteform. To facilitate the use of EPA 1313 for that purpose, additional tests should be performed on glasses containing key radioactive (Tc-99, I-129) and hazardous (Pb, Cr, Ba) components in glass to better assess their leaching behavior under these test conditions.

Glass testing should be expanded to incorporate the suite of EPA tests defined in LEAF and responses should be compared to kinetics of corrosion in IDF disposal conditions. This may include variants of those methods to better simulate the conditions in IDF (such as saturated silica conditions or under simulated groundwater conditions). This would facilitate the development of a screening criteria for glasses that are acceptable for disposal in IDF.

5.0 References

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Appendix A – EPA 1313 Titration Results and Machine Learning

This appendix provides tables of the titration results and figures for the titration curves for the sixteen enhanced LP3 glasses and seven join glasses. Figures are provided for the two artificial neural network (ANN) models and one linear compositional model. The ANN model #1 was comprised of four dense layers of 16, 32, 32, and 16 perceptrons with hyperbolic tangent activation functions, 622 datapoints, learning rates of 0.001, and L1 regularization penalties. The Tensorflow algorithm within Anaconda Python was utilized to develop these models. The inputs for ANN model #1 were pH and weight percent of Al_2O_3, B_2O_3, CaO, Fe_2O_3, MgO, Na_2O, and SiO_2. ANN model #2 fit the coefficients for a polynomial of the form shown below:

$$pH = \left(\frac{E}{A}\right)^{1/3} + B \qquad (A.1)$$

where E is the acid addition (in mol/L), pH = -log[H^+], and A and B are functions of the composition vector, x, such that A=f($\tilde{x}$), B=f($\tilde{x}$), and C=f($\tilde{x}$). The terms comprising the composition vector were mole fractions of Al_2O_3, B_2O_3, CaO, Fe_2O_3, K_2O, MgO, Na_2O, SiO_2, SnO_2, V_2O_5, ZnO, and ZrO_2. Figure A-1 provides the raw data used for the fitting of these models and Table A-1 lists these values. Figure A-2-Figure A-5 compare the goodness-of-fit for these models and show the estimated variation of the model coefficients as a function of composition.

Table A-1. Eluate pH as a function of acid equivalents after tumbling for 48 hours. Equivalents of Acid are in units of eq per gram dry glass.

LP3-01-1		LP3-02		LP3-03		LP3-04		LP3-05mod6		LP3-06		LP3-07	
Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH
0.0000	10.715	0.0000	10.117	0.0000	10.402	0.4000	5.51	0.0000	10.153	0.0000	10.59	0.0000	10.412
-0.0950	11.969	-0.3700	12.338	-0.3570	12.643	0.0500	7.17	-0.3170	12.478	-0.4000	12.57	-0.3880	12.548
0.3880	7.725	0.0380	9.275	-0.0900	12.132	0.0100	8.19	0.3000	6.725	0.0940	7.899	0.0068	9.982
1.4230	6.822	0.0390	9.266	-0.0330	11.612	0.0040	9.47	0.5000	6.477	0.1370	7.492	0.0541	8.988
3.5120	6.171	0.3490	7.813	0.0610	8.513	0.0000	10.32	0.0040	7.891	0.1470	7.432	0.4733	6.869
0.0050	10.22	1.3380	7.094	1.2200	6.101	-0.0010	10.85	0.0640	8.097	1.1060	6.326	3.7193	5.804
		5.1016	5.85	0.5415	6.513	-0.0150	12.02	1.0160	6.303	-0.2850	12.467	0.0000	9.73
				0.8499	6.315	-0.0300	12.54			0.0400	8.598	-0.3880	12.33
				1.6005	5.901					4.2647	5.572	0.0068	9.195
				0.1650	7.199							0.0541	7.623
												0.4733	6.551
												3.7193	5.578
												0.0000	9.854
												-0.3880	12.285
												0.0068	8.954
												0.0541	7.625
												0.4733	6.562
												3.7193	5.511
												-0.3790	12.297
												0.0080	8.759
												0.0070	9.76
												0.0200	8.695
												0.0320	8.753
												0.0000	10.22
												-0.3980	12.585
												0.0070	8.096
												0.0149	7.8
												0.0224	7.817
												0.0340	7.639

Table A-1 (cont.). Eluate pH as a function of acid equivalents after tumbling for 48 hours.

LP3-08		LP3-09		LP3-10		LP3-12		LP3-14		LP3-15		LP3-16	
Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH
0.0000	9.729	0.0000	10.91	0.0000	9.815	Trail #1		0	10.761	0.4000	5.35	0.4000	5.57
-0.3920	11.652	-0.3880	12.68	-0.3810	12.236	0.0000	10.132	-0.0250	11.362	0.0500	6.97	0.0500	7.09
0.0064	8.872	0.0068	10.511	0.0105	9.493	-0.3920	12.683	0.1210	8.099	0.0100	7.98	0.0100	7.97
0.0290	7.679	0.0541	9.767	0.0318	7.956	0.0064	8.899	0.4320	7.082	0.0040	8.99	0.0040	8.82
0.0541	6.922	0.4733	7.617	0.0918	7.102	0.0290	8.258	0.4320	7.082	0	10.15	0	10.32
0.4181	5.865	3.7193	6.296	1.5608	5.3	0.0541	7.589	1.0550	6.605	-0.0010	10.87	-0.0010	10.84
0.0000	9.576			0.0000	10.029	0.4181	6.073	2.8090	5.995	-0.0150	12.09	-0.0150	12.08
-0.3920	12.384			-0.3810	11.947	Trail #2		0.0220	9.496	-0.0300	12.55	-0.0300	12.57
0.0064	9.115			0.0105	8.714	0.0000	10.088						
0.0290	7.546			0.0318	7.711	-0.2750	12.448						
0.0541	6.743			0.0918	6.891	0.0064	9.04						
0.4181	5.493			1.5608	5.634	0.0300	8.163						
0.0000	9.789			0.0000	10.29	0.0580	7.4						
-0.3880	11.945			-0.3880	12.226	0.5000	6.05						
0.0068	8.41			0.0068	9.99								
0.0541	6.924			0.0541	8.606								
0.4399	5.787			0.4399	6.231								
3.8282	4.303			3.8282	4.001								
0.0064	8.798			0.0517	8.682								
0.0302	7.24			0.2217	6.966								
0.0541	6.977			0.3916	6.49								
0.4181	5.705			1.5000	5.851								
0.0064	9.287			0.1367	6.541								
0.0290	8.184			0.0517	7.76								
0.0541	7.005			0.0897	7.163								
0.4181	5.475			0.1277	6.636								
0.0000	9.729			1.5957	5.399								
-0.3920	11.652			0.0100	8.48								
0.0064	8.872			0.0300	9.467								
0.0290	7.679												
0.0541	6.922												
0.4181	5.865												

Table A-1 (cont.). Eluate pH as a function of acid equivalents after tumbling for 48 hours.

LAWPh3-19mod1		LAWPh3-20		D10B20N		D6B20N		D6B14N		D6B10N	
Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH
0.037	9.156	0	10.271	0	8.543	0	10.22	0	9.457	0	6.681
0	10.705	-0.287	12.392	-0.381	12.459	-0.381	12.487	-0.381	12.455	-0.381	12.429
-0.243	12.268	0.012	9.266	0.005	8.102	0.005	9.179	0.005	8.839	0.005	8.255
0.3008	7.398	0.033	8.466	0.01	8.871	0.01	7.577	0.01	7.793	0.01	6.119
0.352	7.271	0.193	6.6	0.03	8.898	0.03	7.54	0.03	6.59	0.03	4.639
0.4032	7.152	1.948	5.296	0.075	8.241	0.075	7.109	0.075	4.741	0.075	3.68
2.7028	6.251			1.5	6.424	1.5	3.446	1.5	2.681	1.5	2.58
				0	8.889	0	10.007	0.9	2.726	0	8.958
				-0.381	12.328	-0.366	11.987	0	7.447	-0.368	12.389
				0.0045	9.418	0.005	9.454	-0.367	11.978	0.0046	8.441
				0.0773	8.207	0.01	9.598	0.0049	8.825	0.005	7.358
				1.3766	6.501	0.075	8.601	0.0097	7.932	0.0114	6.955
				3	5.56	0.9398	9.044	0.0282	7.055	0.02	5.839
						0.01	6.777	0.0647	5.239	0	8.151
						0.075	5.472			-0.356	12.185
						0.9398	3.631			0	9.389
										0.0048	7.901
										0.0053	7.408
										0.0212	5.945

D6B		D2B		Dike	
Equivalents of Acid (eq/g)		Equivalents of Acid (eq/g)	Eluate pH	Equivalents of Acid (eq/g)	Eluate pH
0	5.645	0	8.944	0	8.046
-0.381	12.328	-0.381	12.475	-0.366	12.096
0.0050	6.414	0.0050	6.693	0.0036	8.609
0.0100	5.004	0.0100	3.944	0.0000	8.291
0.0300	3.749	0.0300	3.184	0.0053	6.785
0.0750	3.212	0.0750	2.628	0.0100	5.014
1.5000	2.160	1.5000	1.027	0	8.970
0	8.446	0	9.059	0.0036	4.907
-0.371	12.396	-0.367	12.457	0.0000	8.684
-0.2783	12.433	0.0000	9.319	0.0053	4.576
-0.2474	12.354	0.0025	8.38	0.0100	4.000
0.0046	8.632	0.0050	7.237	0	8.242
0.0091	7.376	0.0072	5.932	-0.381	12.495
0	8.707	0	8.141	0.0050	6.559
-0.345	11.741	-0.353	12.288	0.0100	5.18
0.0000	8.634	0.0000	9.128	0.0300	3.1
0.0049	7.817	0.0026	8.412	0.0750	2.570
0.0096	6.772	0.0050	7.338	1.5000	1.000
0.0100	5.810	0.0077	5.831		
		0	8.835		
		0	9.324		

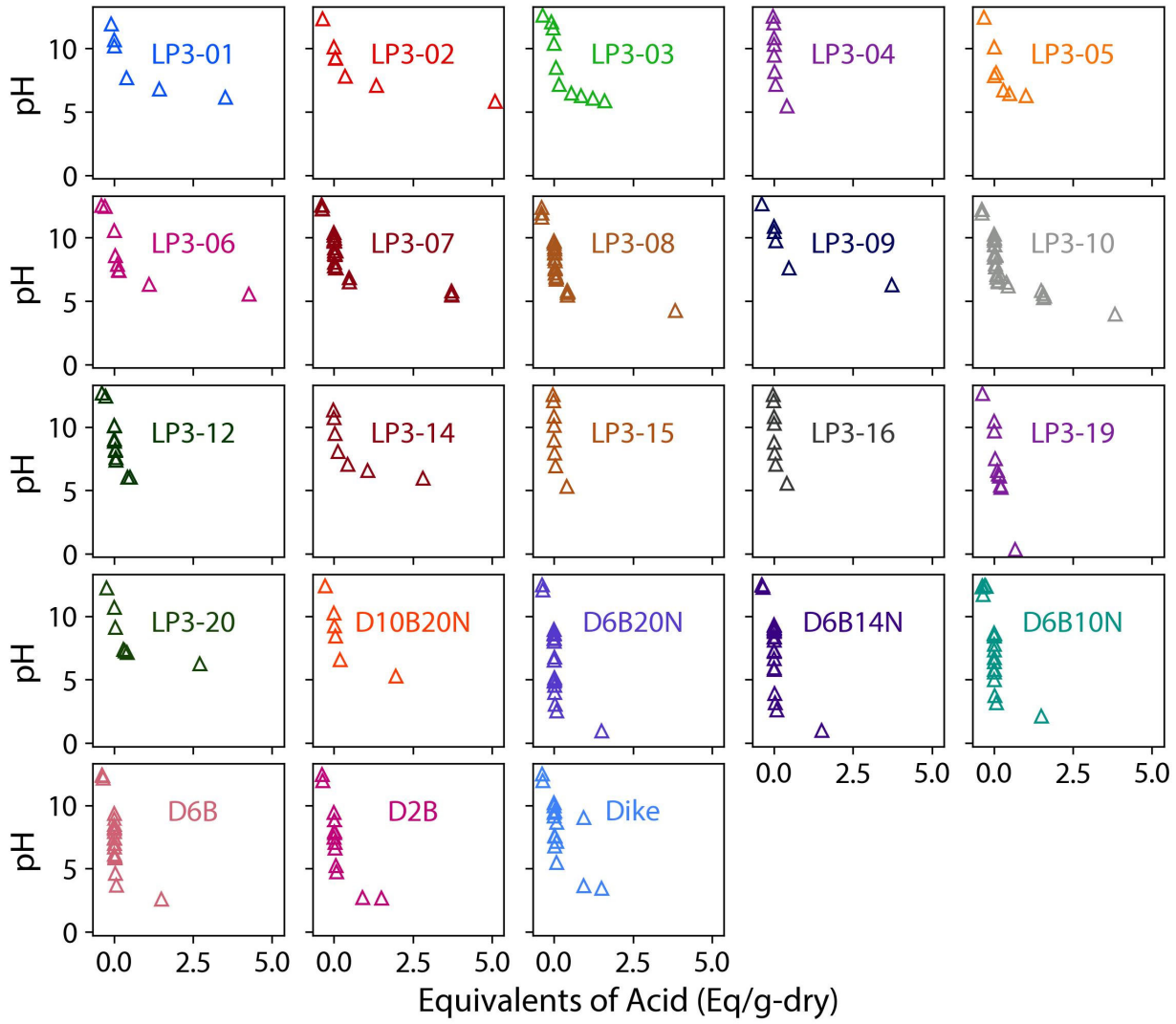

Figure A-1. Final eluate pH versus acid equivalents for the glasses tested in this study. Experimental details are provided in Section 2.6.

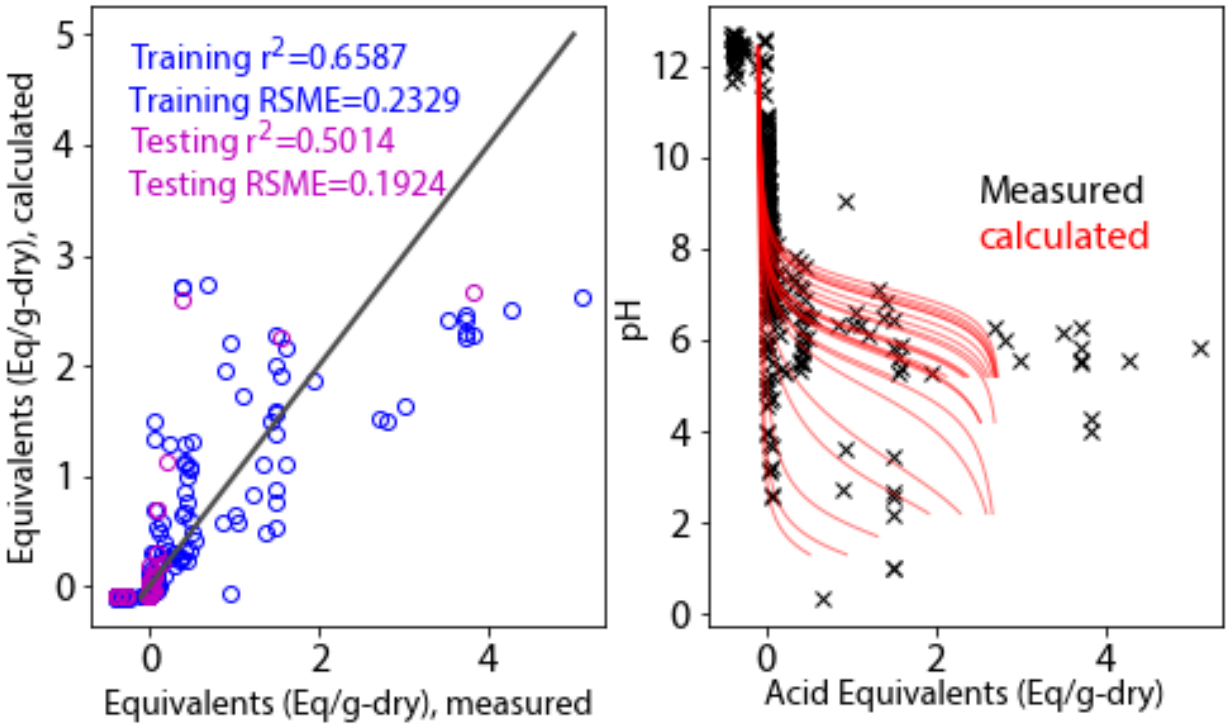

Figure A-2. (left) Comparison of measured acid equivalents to compositional ANN model #1 and (right) titration curves for LP3 glasses and join glasses. The solid line on the left is the unity line. On the left, the training dataset is shown in blue and the testing dataset is shown in magenta.

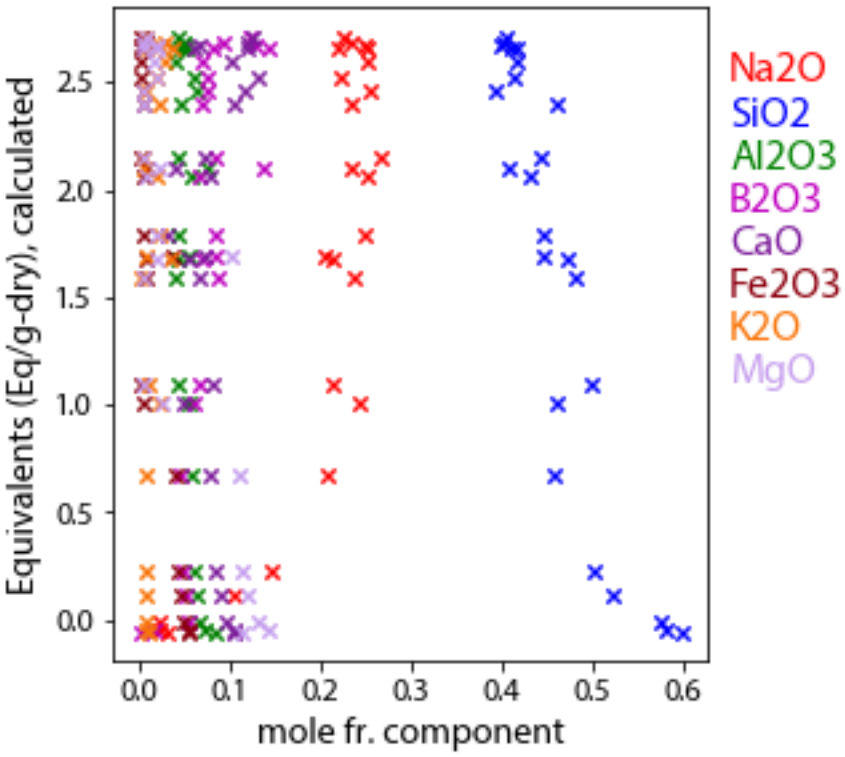

Figure A-3. Calculated acid equivalents as a function of mole fraction component for 8 oxides fit using ANN model #1

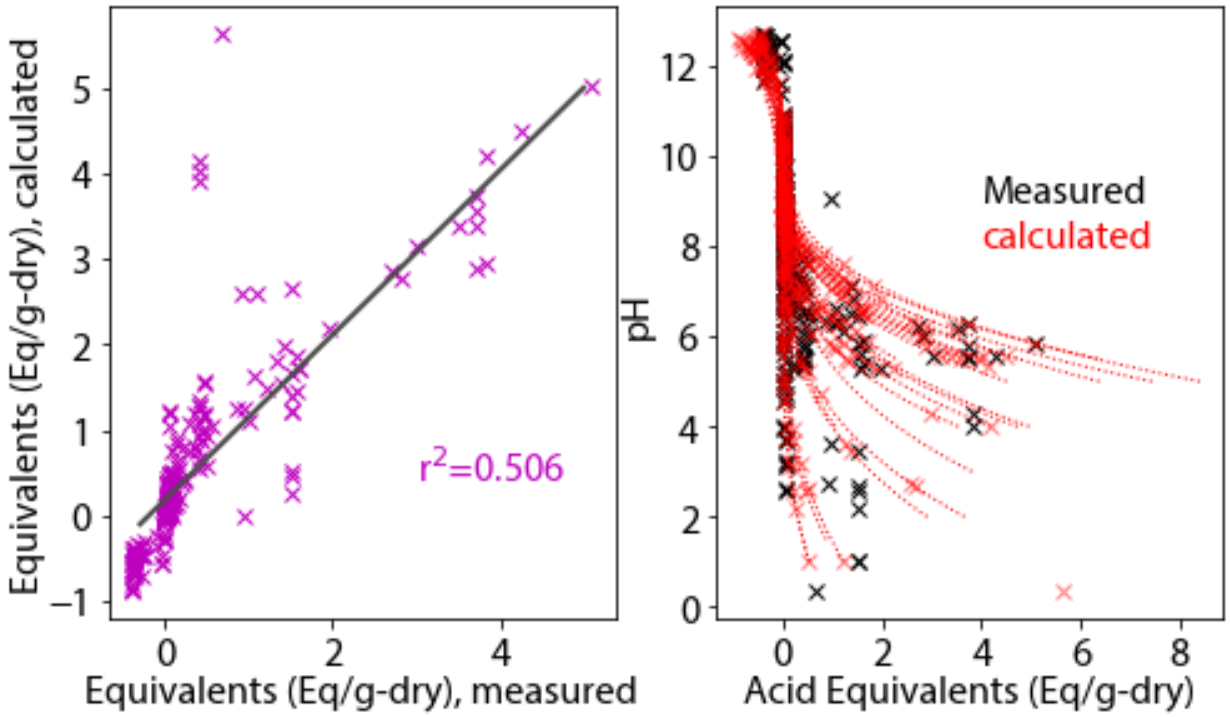

Figure A-4. (left) Comparison of measured acid equivalents to compositional ANN model #2 using a cubic fitting function and (right) titration curves for LP3 glasses and join glasses. The solid line on the left is the unity line.

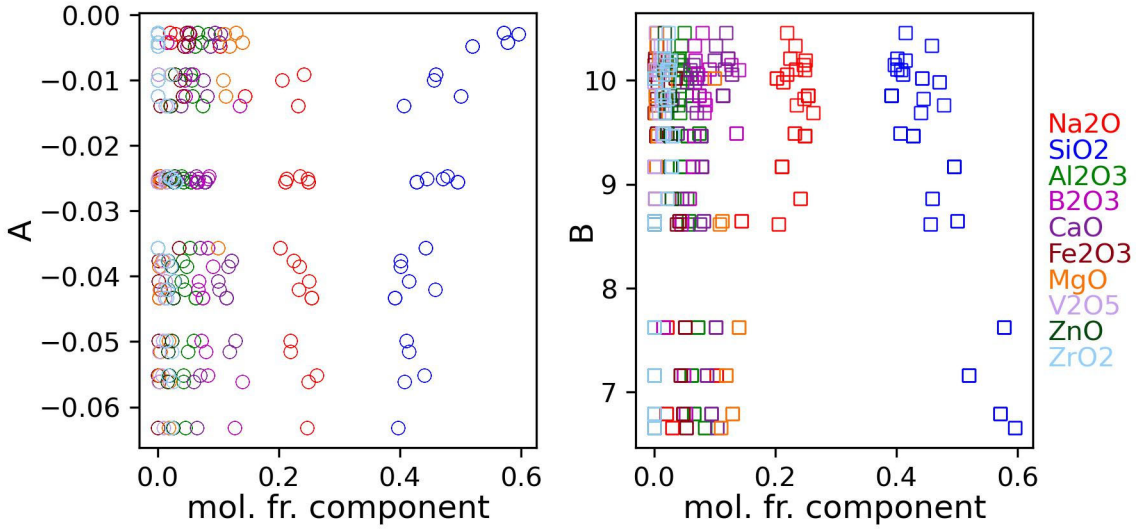

Figure A-5. Variation of model coefficients as a function of composition for ANN model #2

Appendix B – Compiled EPA 1313 Results

This appendix presents the normalized elemental releases for the sixteen LP3 glasses tested with the EPA 1313 test. Glasses LP3-04, LP3-08, LP3-10, LP3-15, and LP3-16 were run in duplicate. Figure B-1 and Figure B-3 compare the calculated equivalent layer thickness, E_{th}, with the alteration layer thickness measured by cross-sectional SEM. The calculated equivalent layer thickness, E_{th}, was calculated using Equation 2.4. Figure B-4 – Figure B-13 compare the normalized elemental releases of Al, B, Ca, Na, or Si, either in separate plots or on the same plot. Figure B-14 – Figure B-34 group the elemental releases of Al, B, Ca, Na, and Si per individual glass. Table B-1 lists the normalized elemental releases for the LAWPh3 glasses. Table B-2 lists the normalized elemental releases for the join glasses.

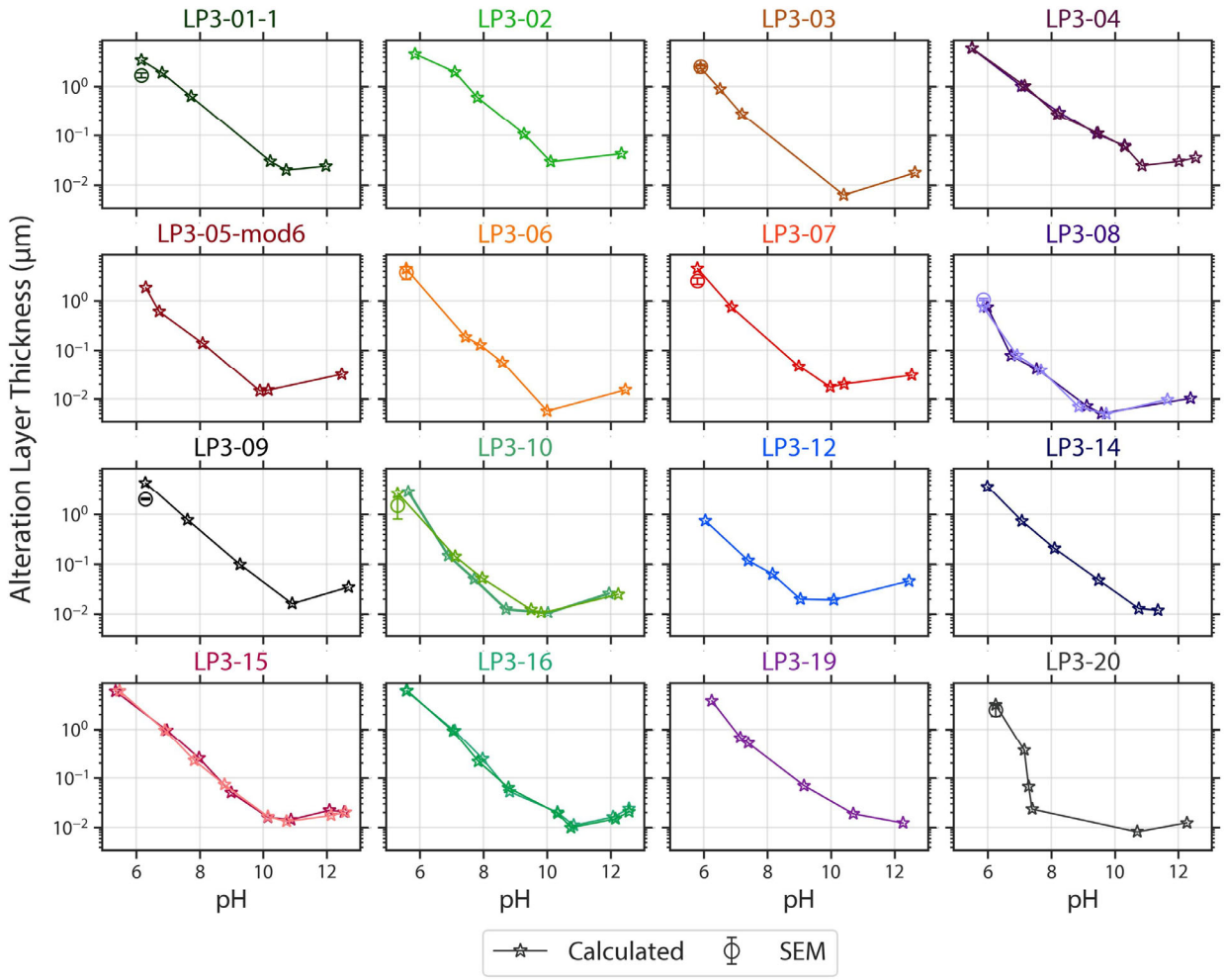

Figure B-1. Calculated alteration layer thickness, Eth, calculated using normalized release of B for 16 LP3 glasses

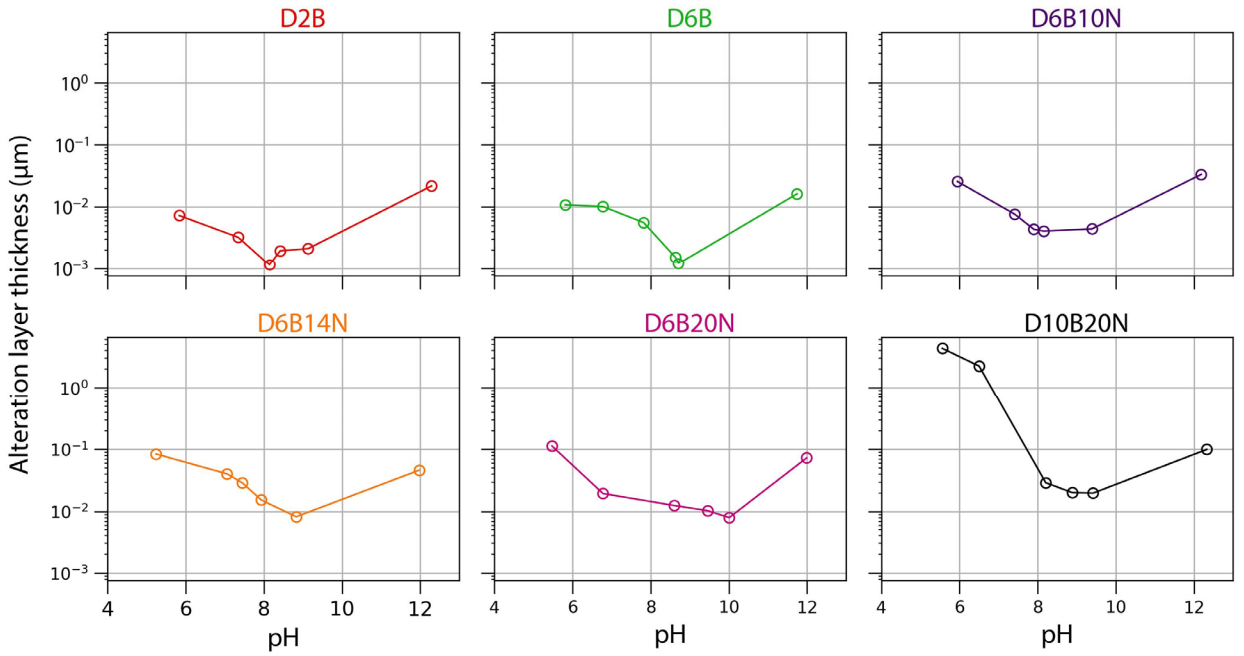

Figure B-2. Calculated alteration layer thickness (E_{th}) calculated using normalized release of B for 6 join glasses (note Dike glass is excluded as it did not contain boron)

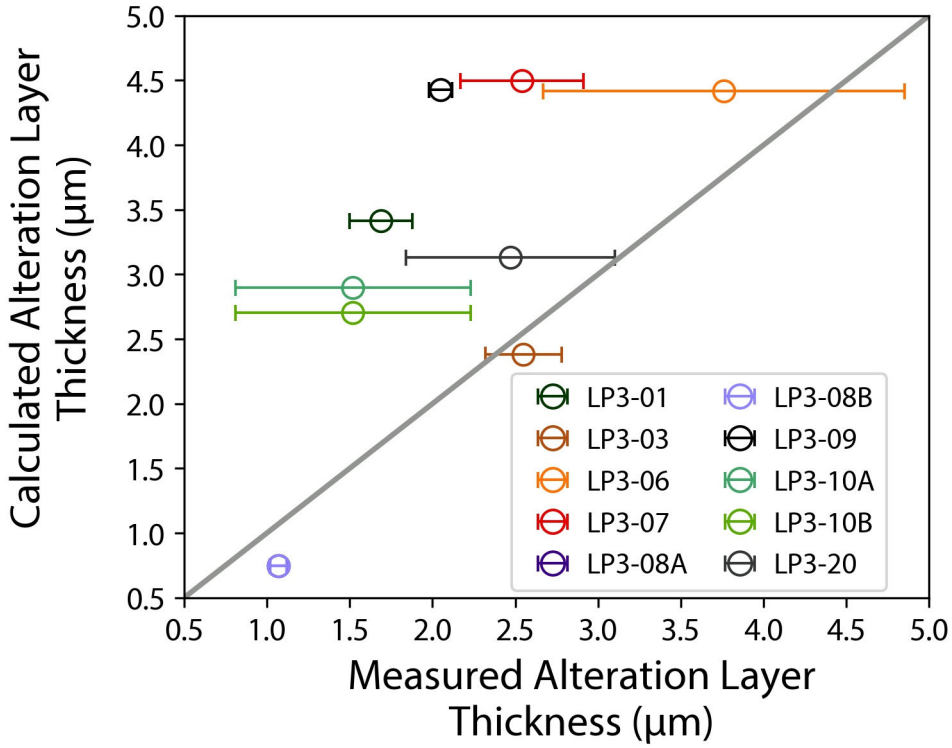

Figure B-3. Comparison of EPMA measured alteration layer thickness and calculated equivalent layer thicknesses for LAW glasses tested by EPA 1313 at pH = 5.5 ± 1 based on the normalized release of B.

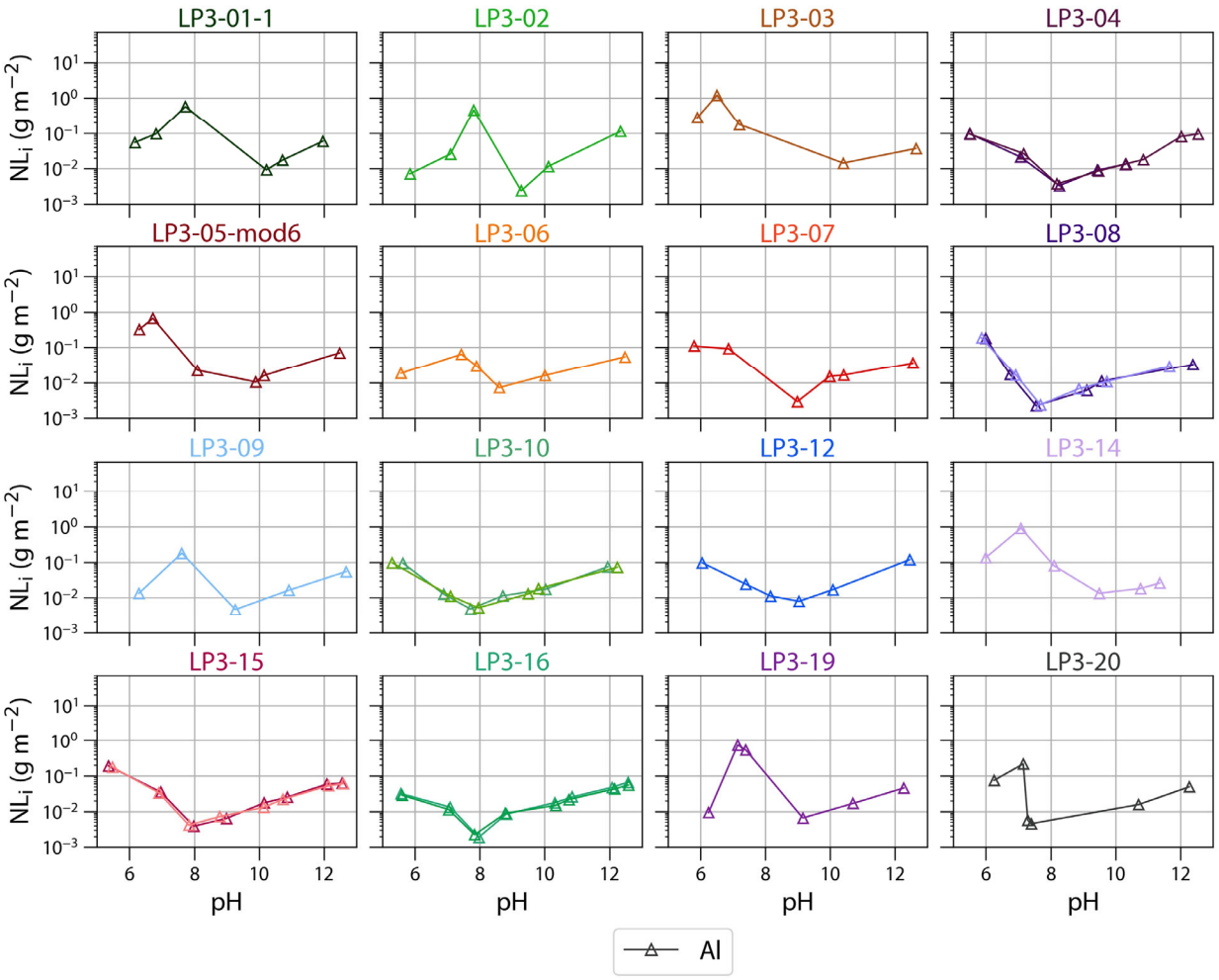

Figure B-4. EPA 1313 NL_{Al} for the 16 LP3 glasses

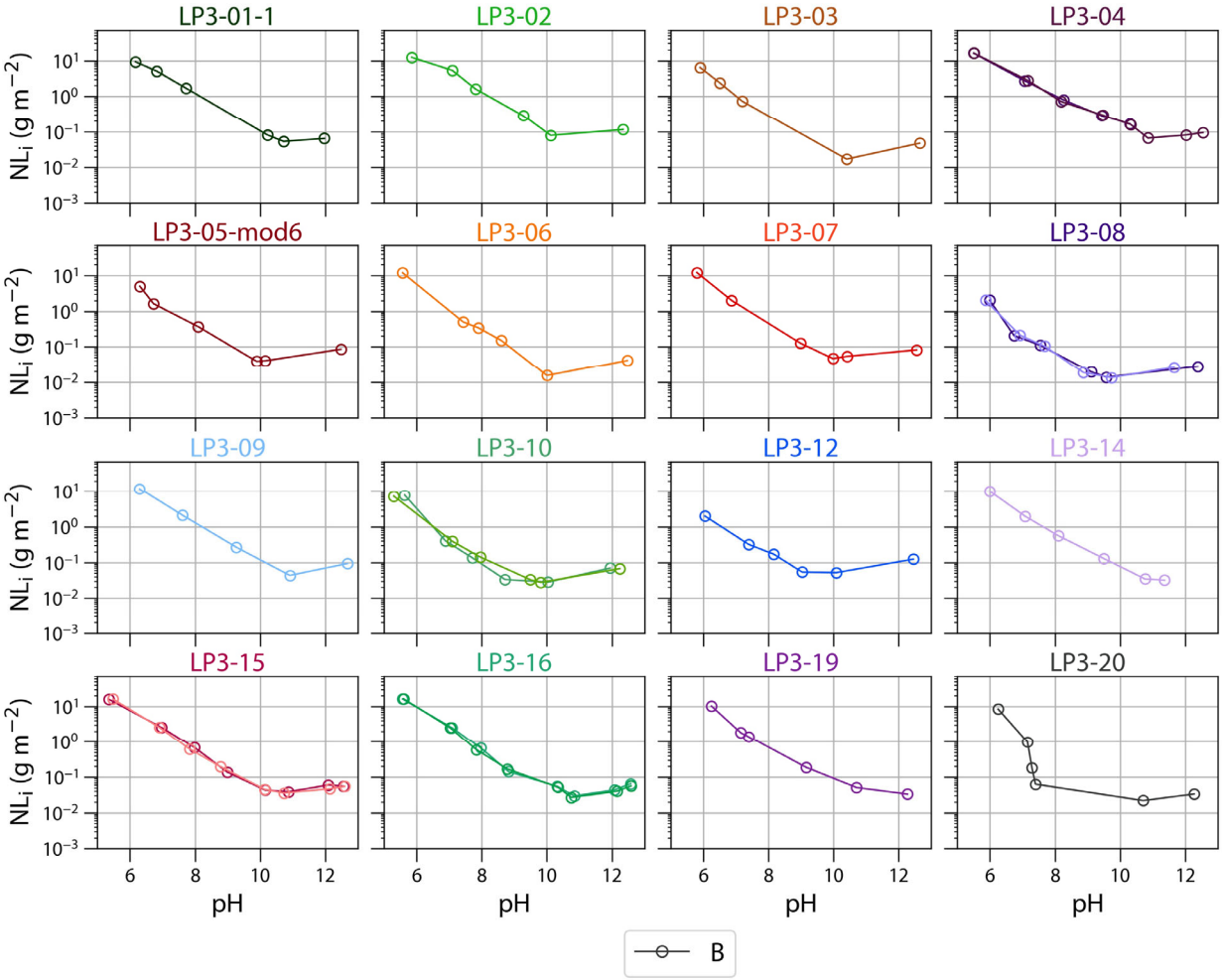

Figure B-5. EPA 1313 NL_B for the 16 LP3 glasses

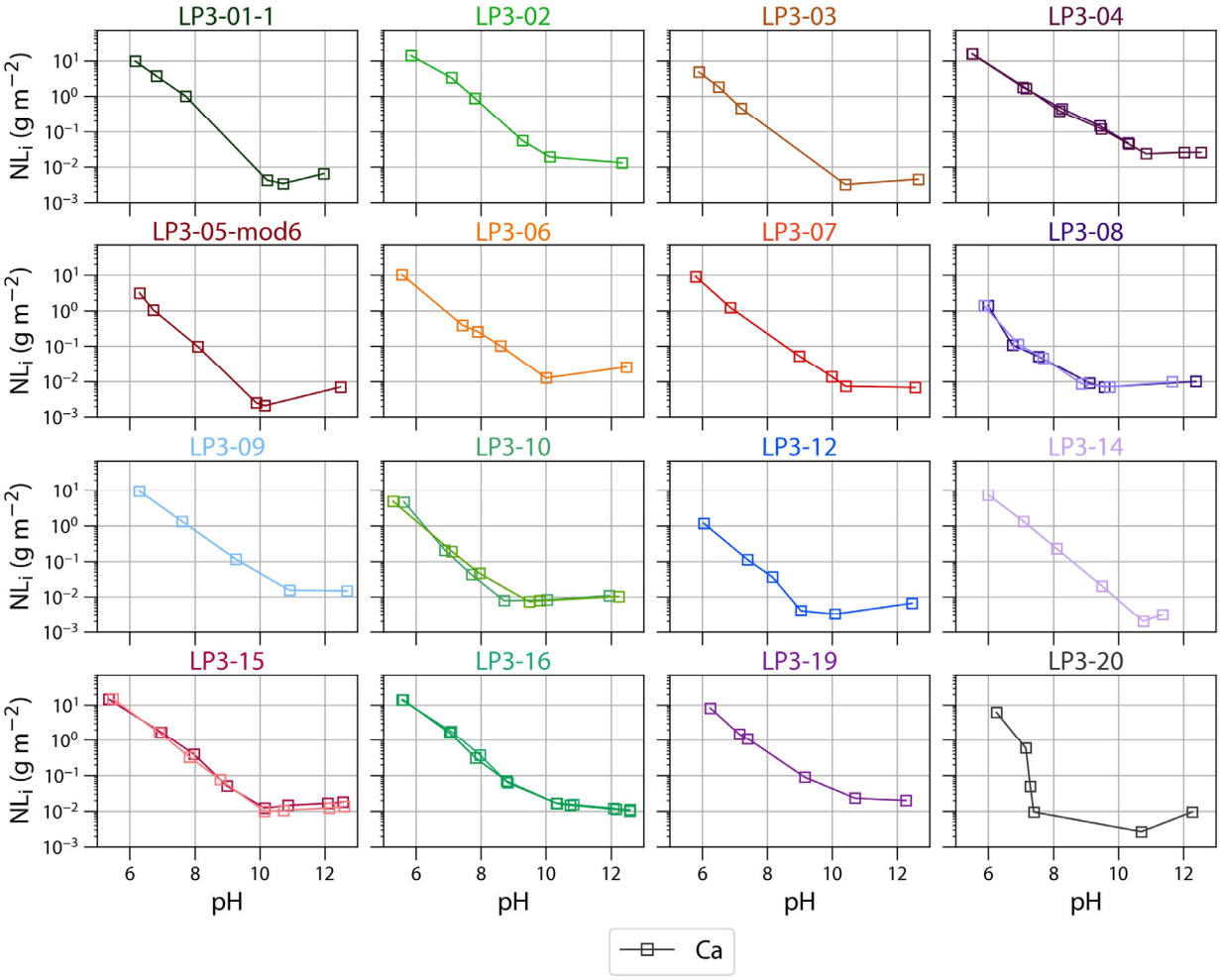

Figure B-6. EPA 1313 NL_{Ca} for the 16 LP3 glasses

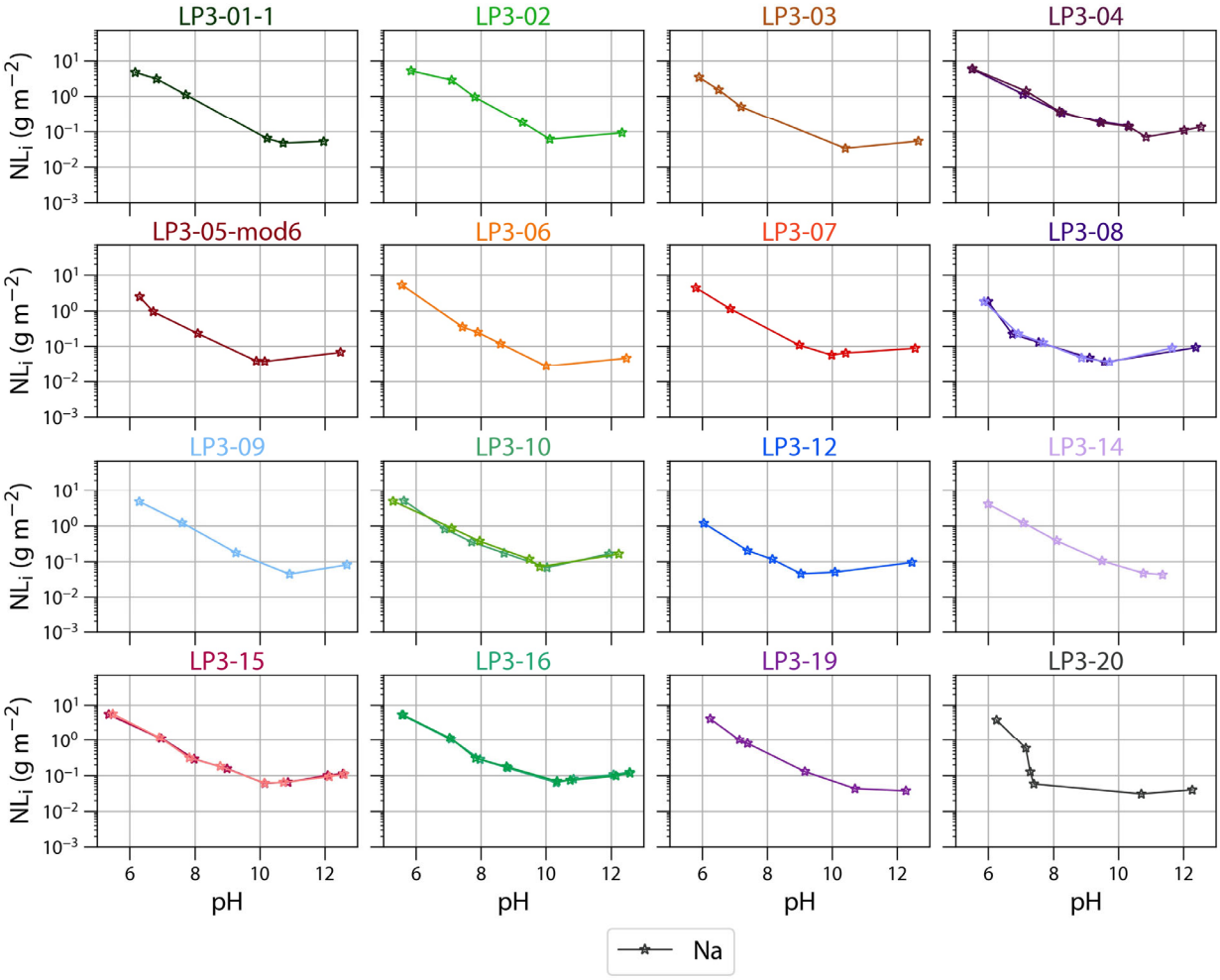

Figure B-7. EPA 1313 NL_{Na} for the 16 LP3 glasses

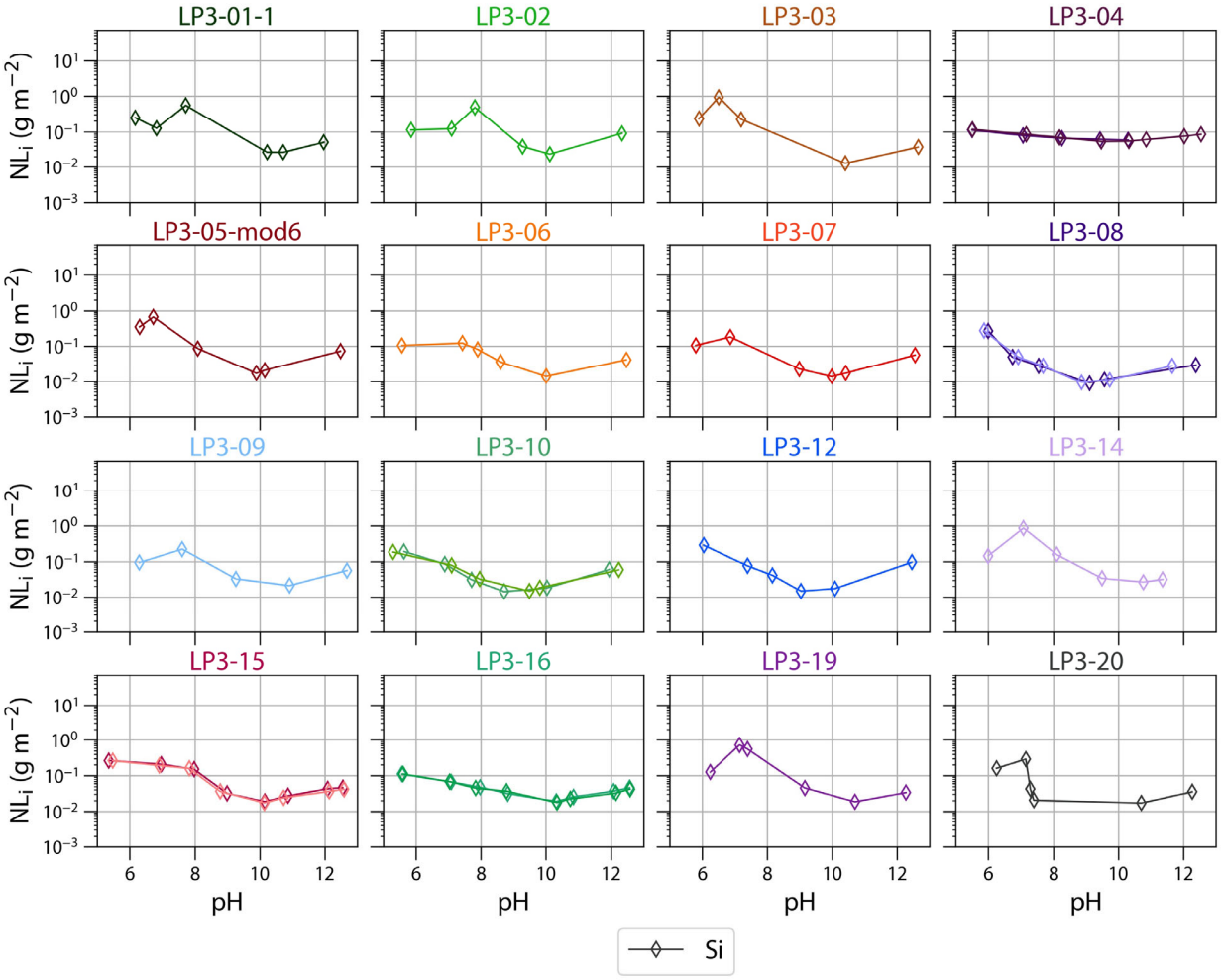

Figure B-8. EPA 1313 NL_{Si} for the 16 LP3 glasses

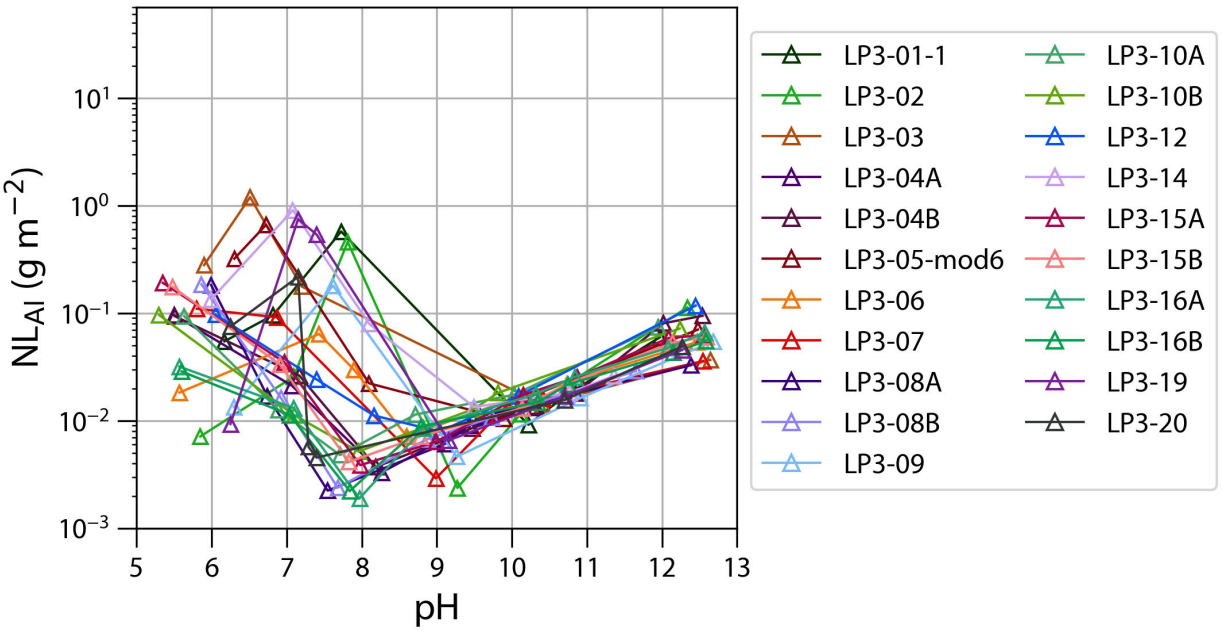

Figure B-9. EPA 1313 normalized elemental release of Al for LP3 glasses

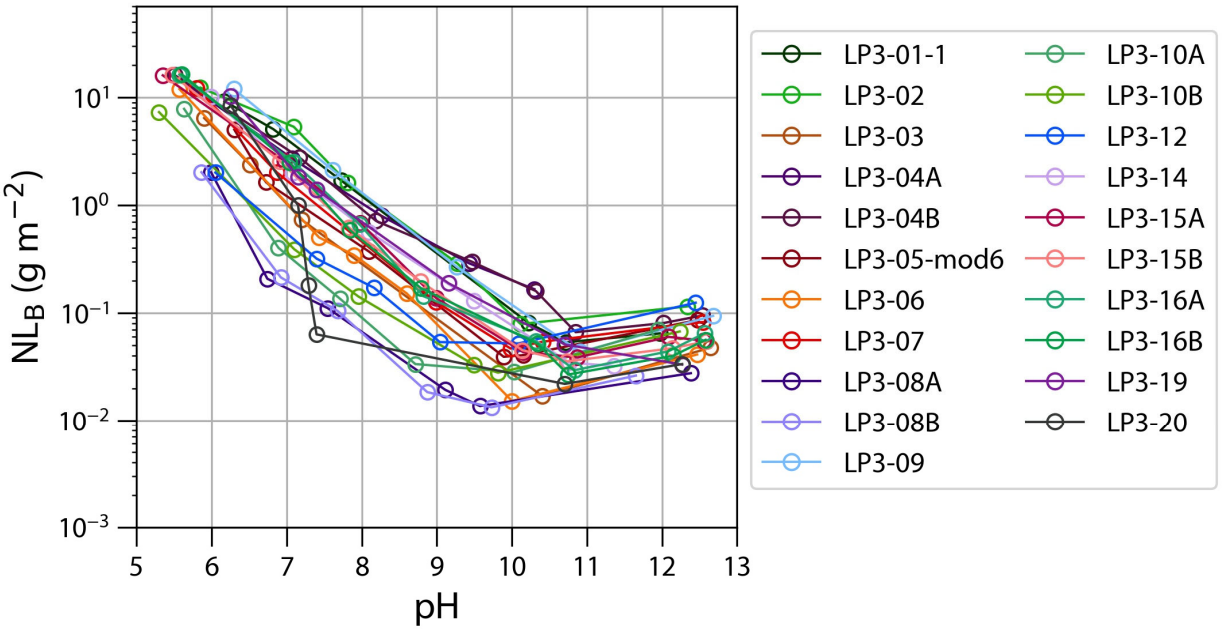

Figure B-10. EPA 1313 normalized elemental release of B for LP3 glasses

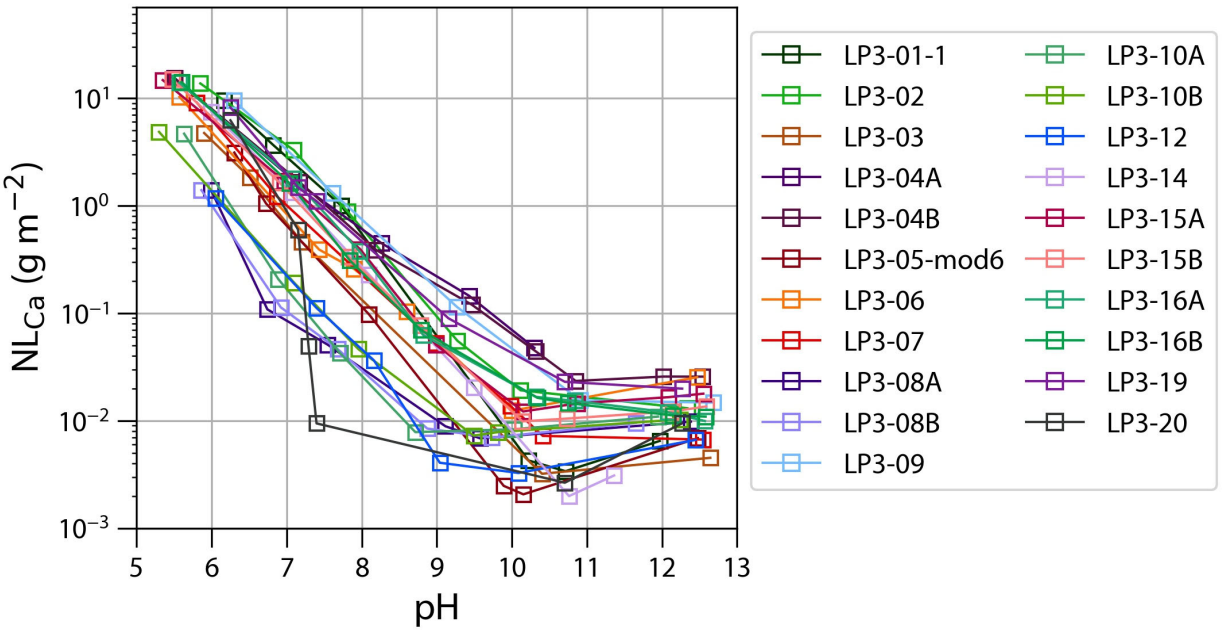

Figure B-11. EPA 1313 normalized elemental release of Ca for LP3 glasses

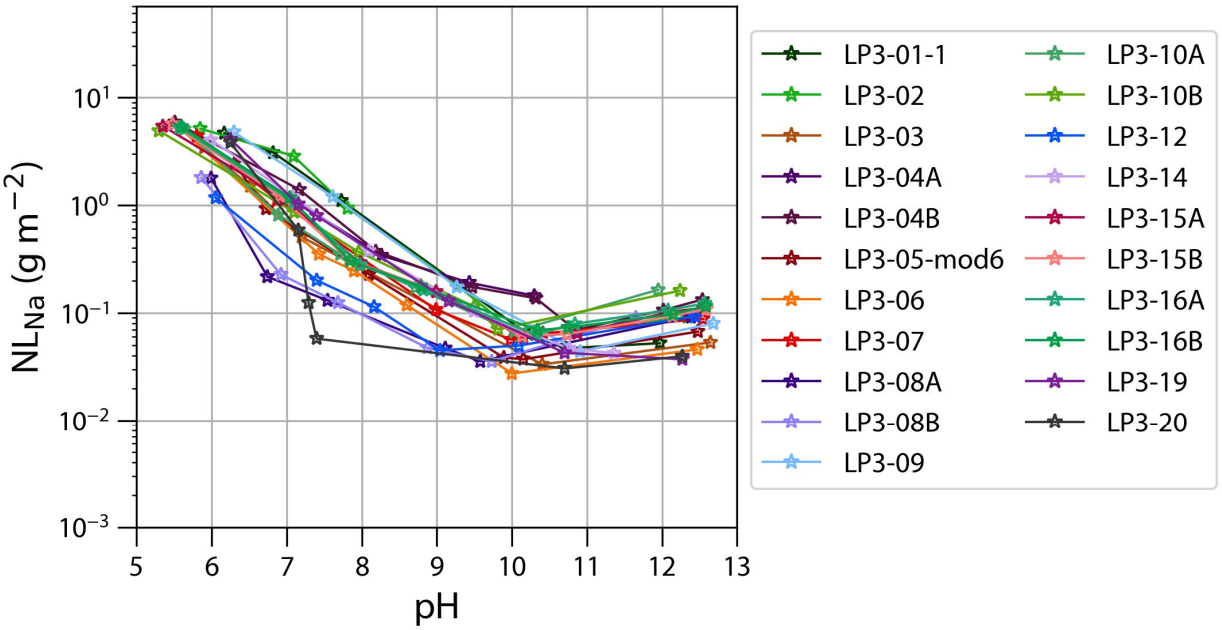

Figure B-12. EPA 1313 normalized elemental release of Na for LP3 glasses

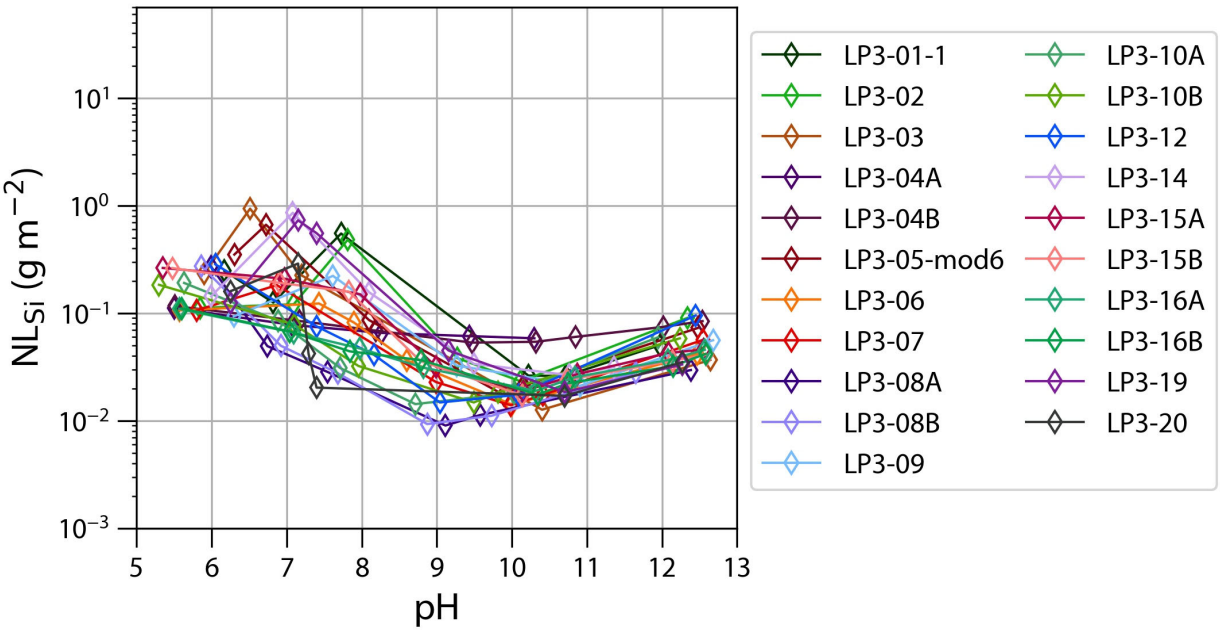

Figure B-13. EPA 1313 normalized elemental release of Si for LP3 glasses

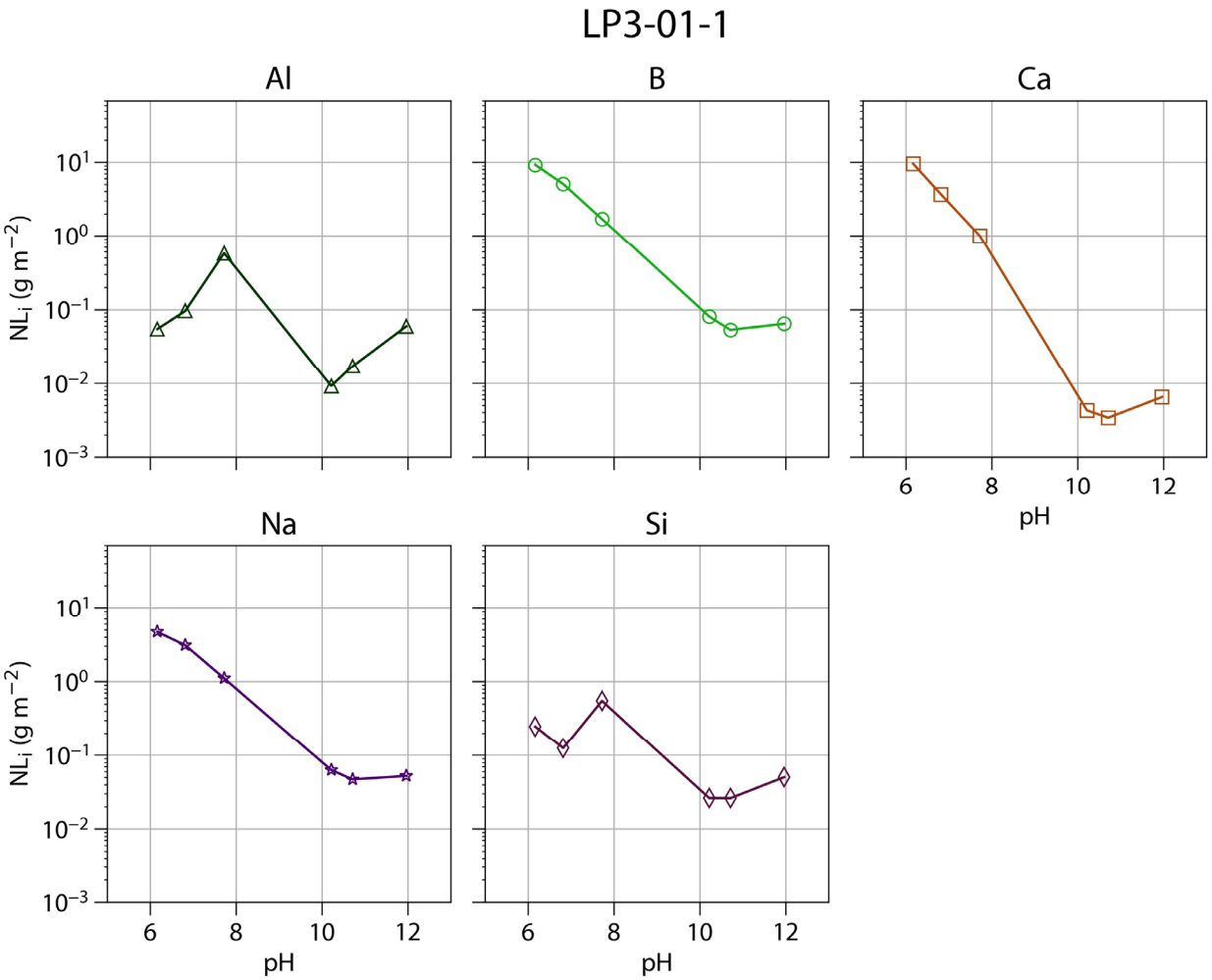

Figure B-14. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-01-1

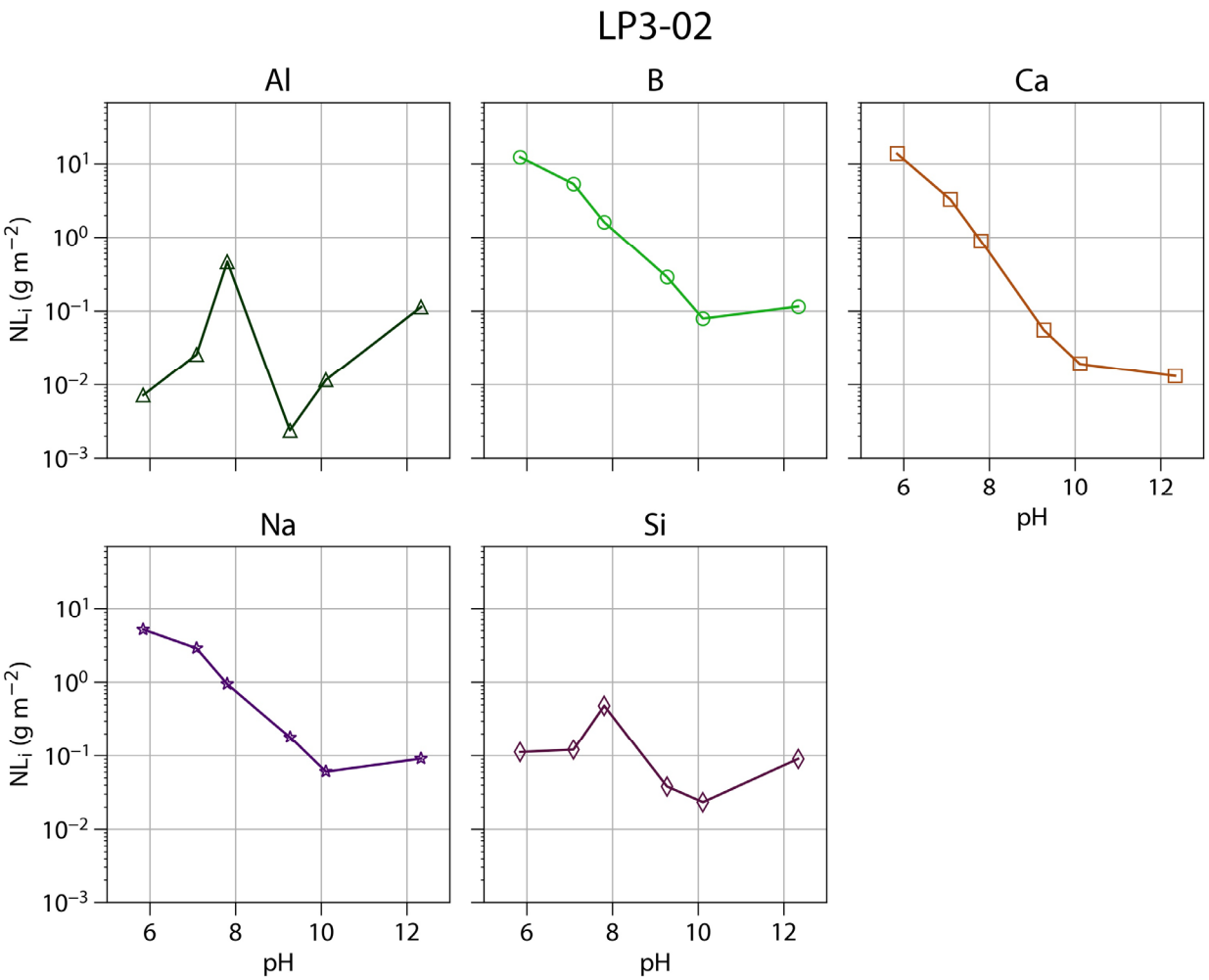

Figure B-15. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-02

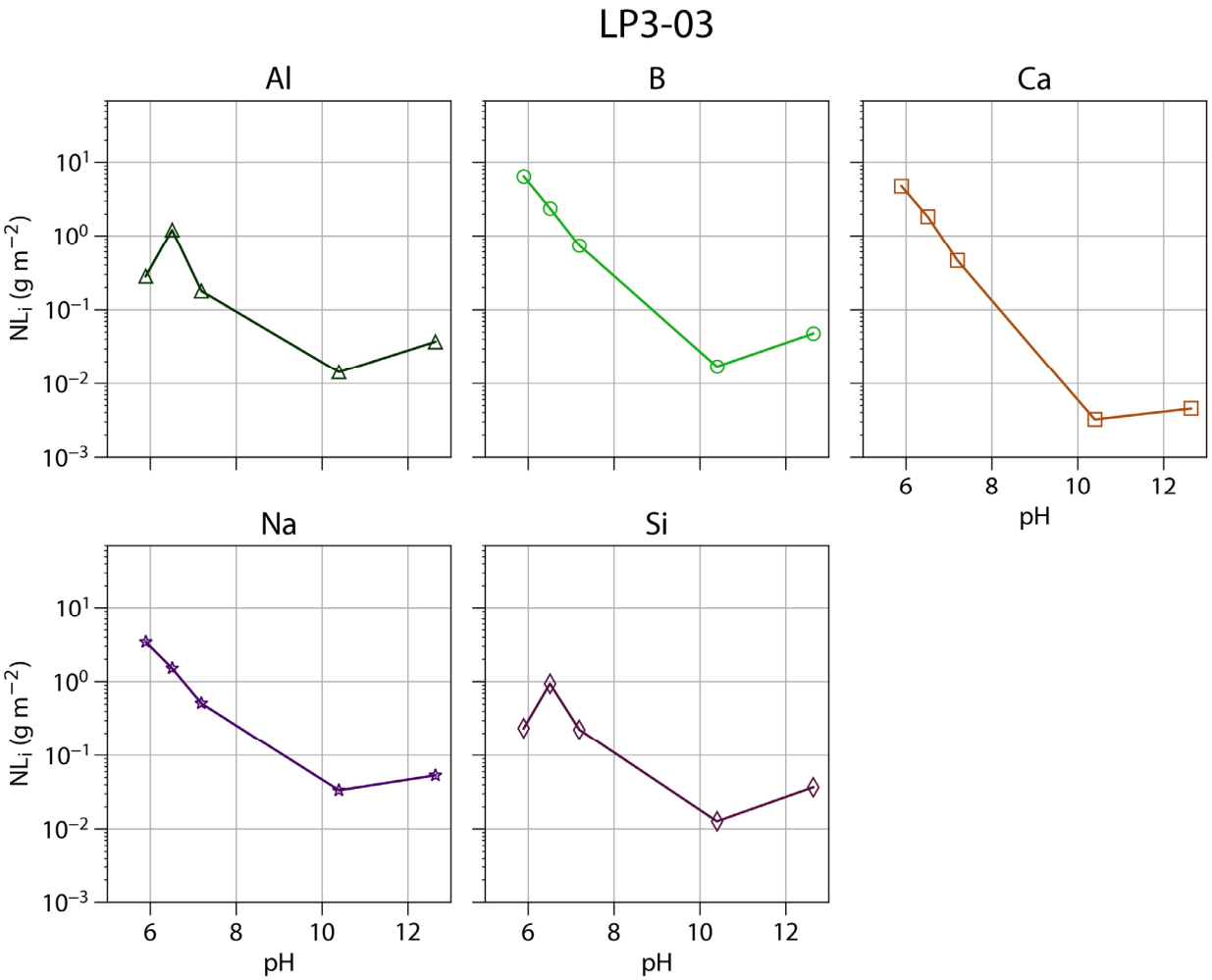

Figure B-16. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-03

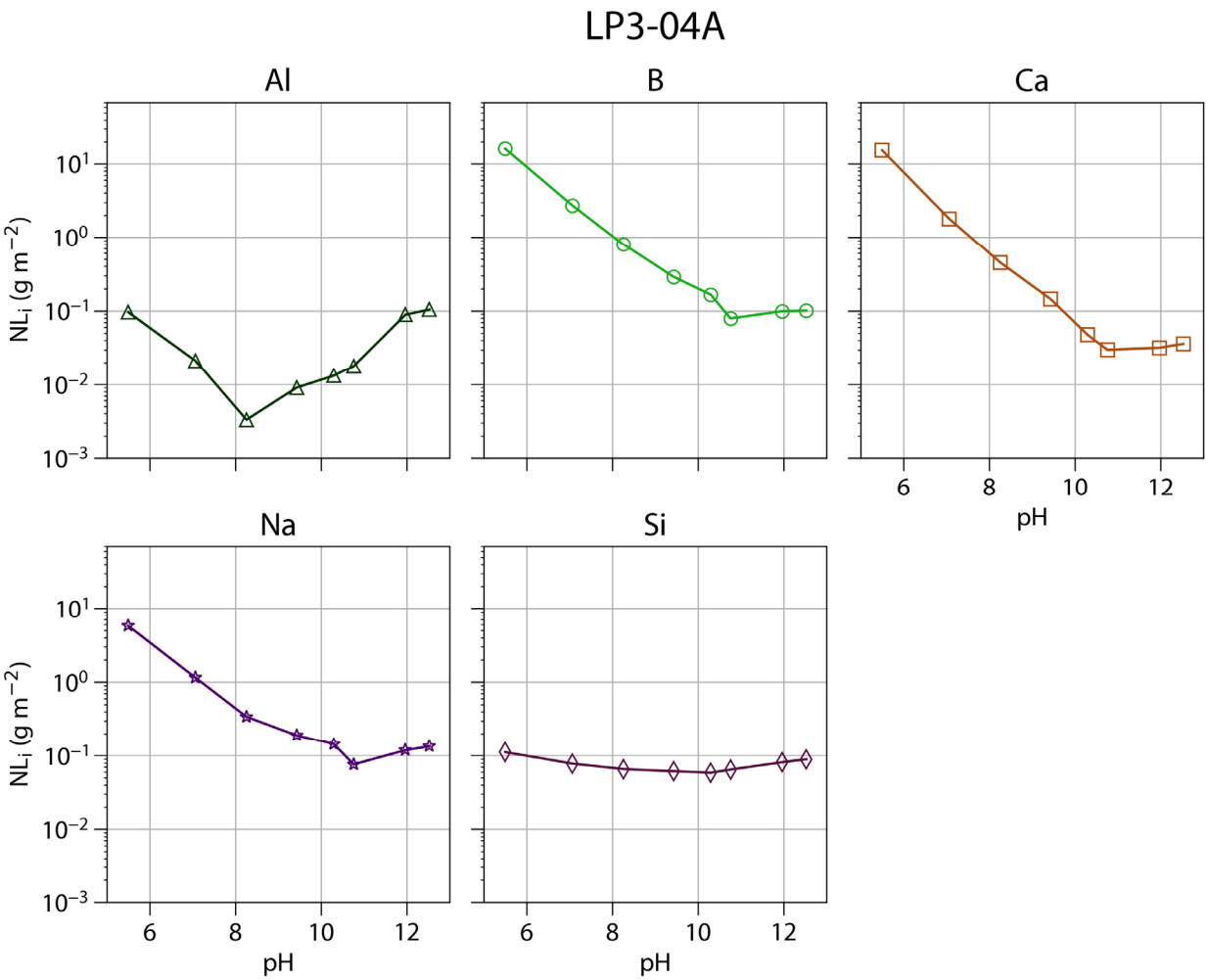

Figure B-17. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-04 (trial #1)

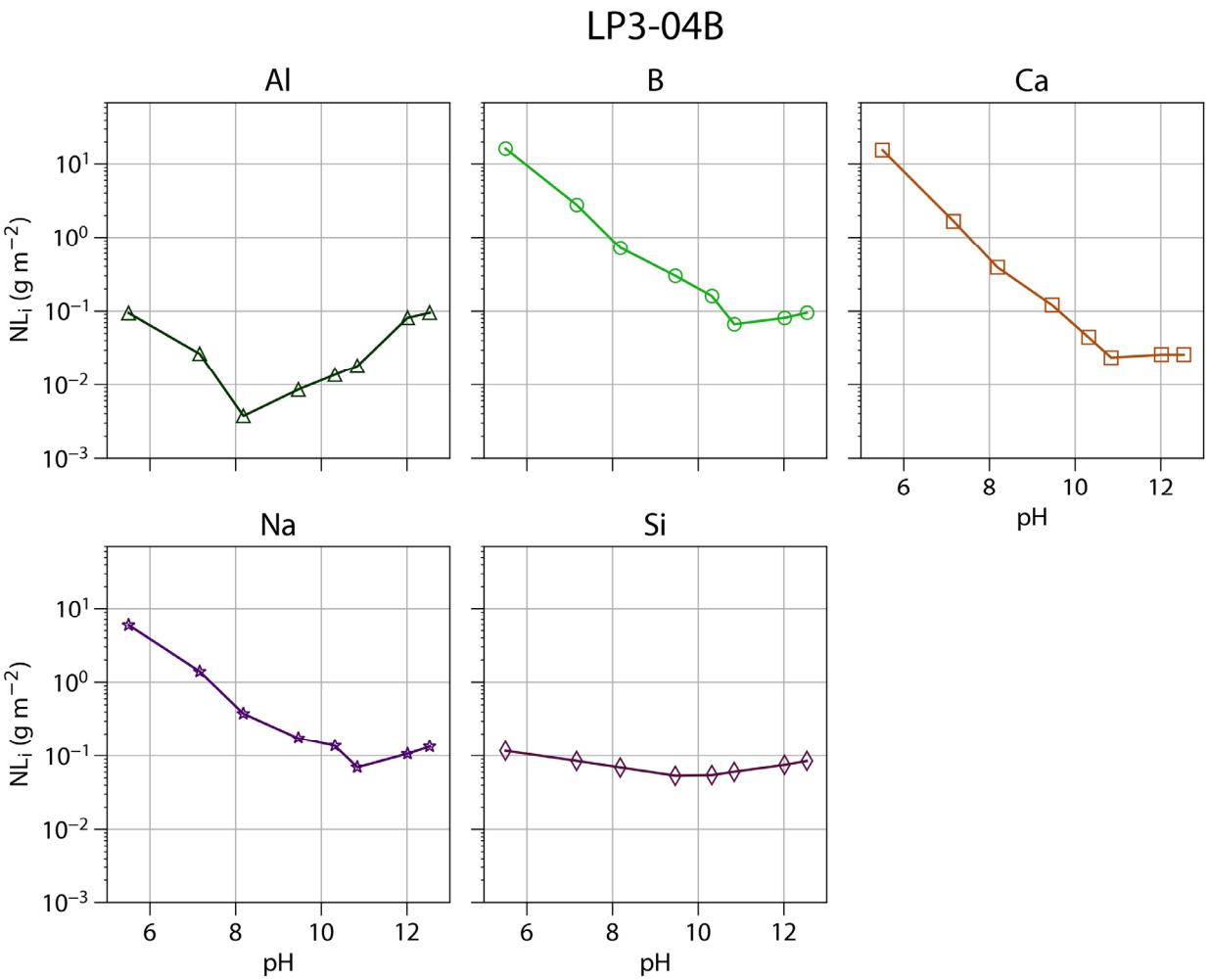

Figure B-18. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-04 (trial #2)

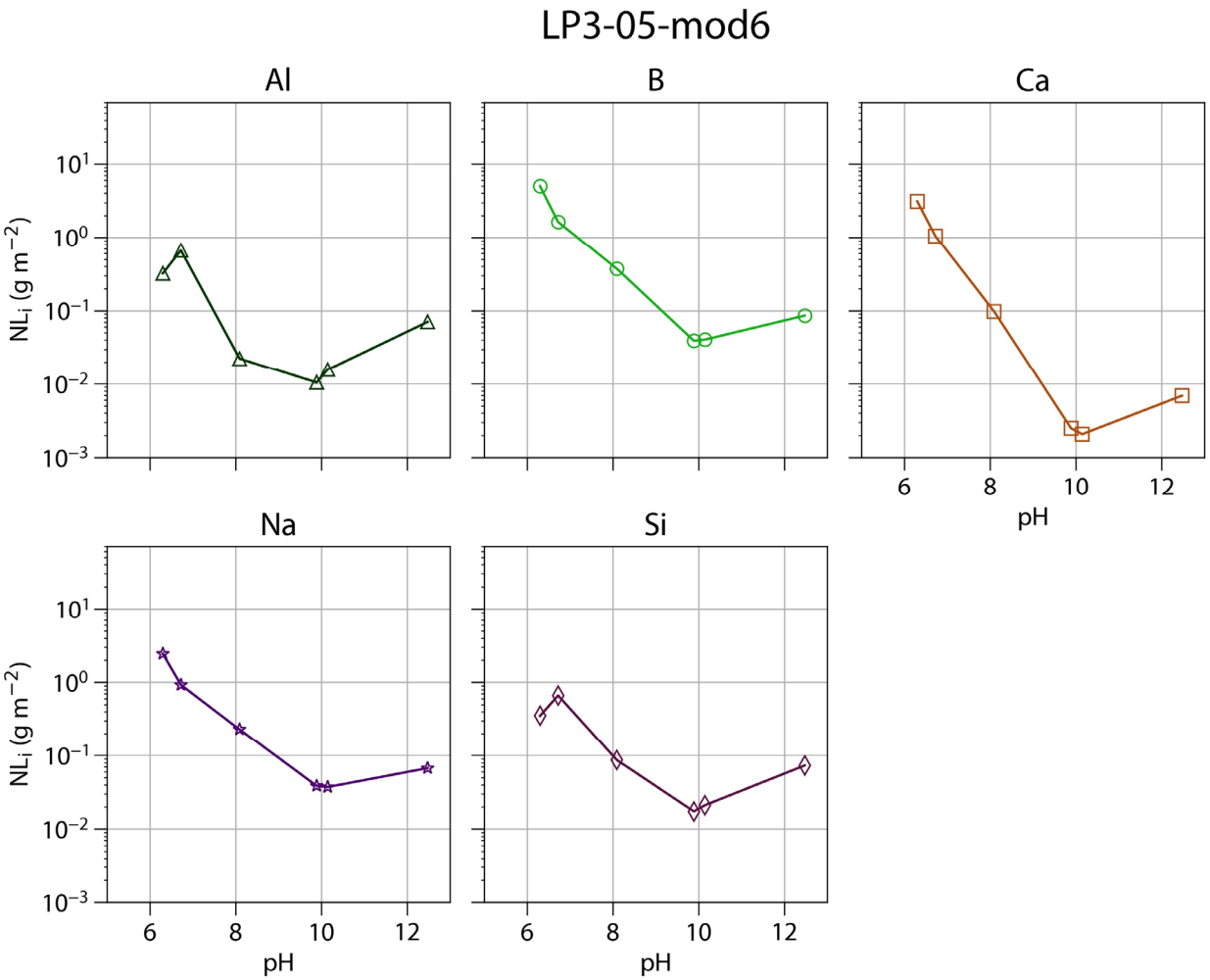

Figure B-19. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-05mod6

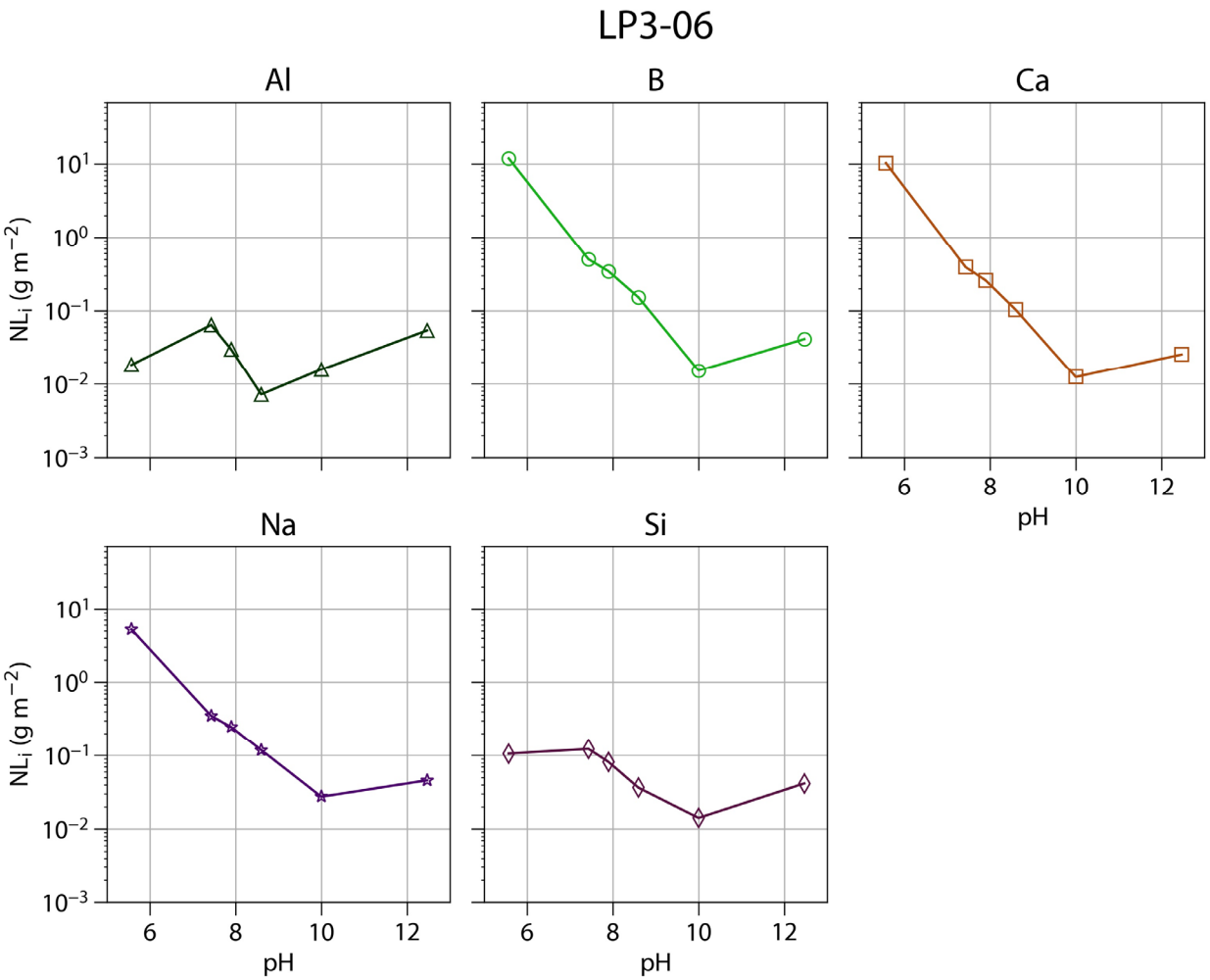

Figure B-20. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-06

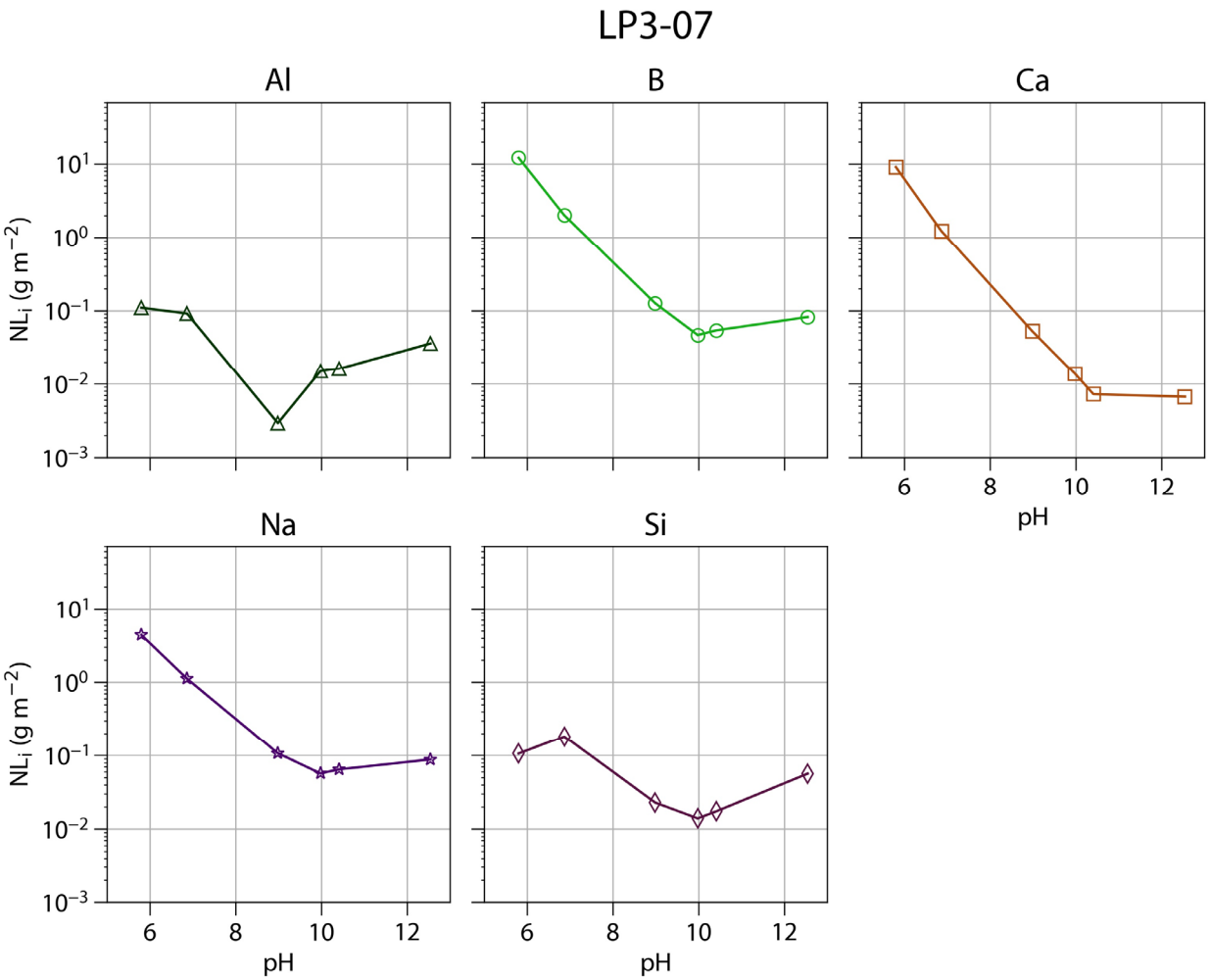

Figure B-21. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-07

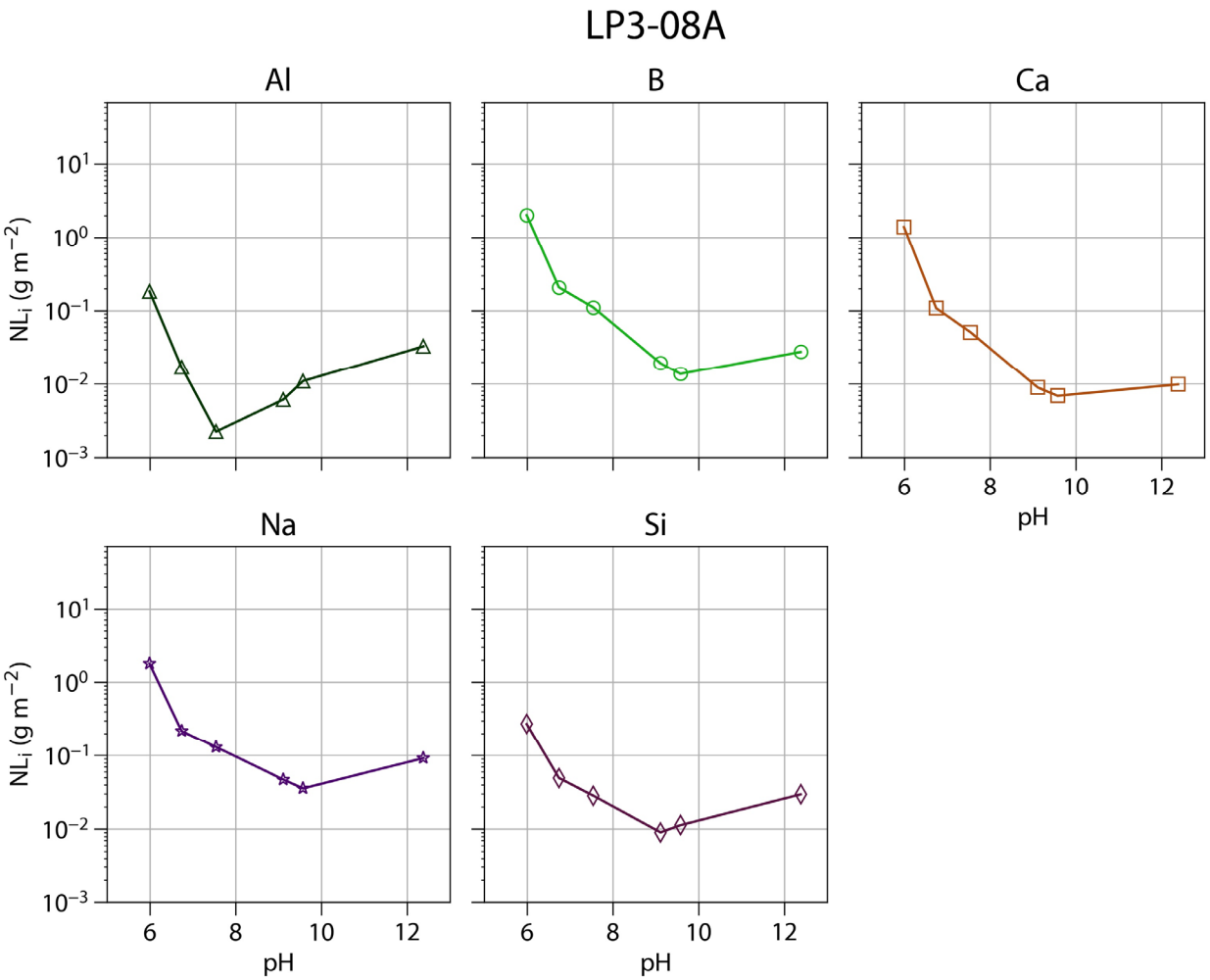

Figure B-22. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-08 (trial#1)

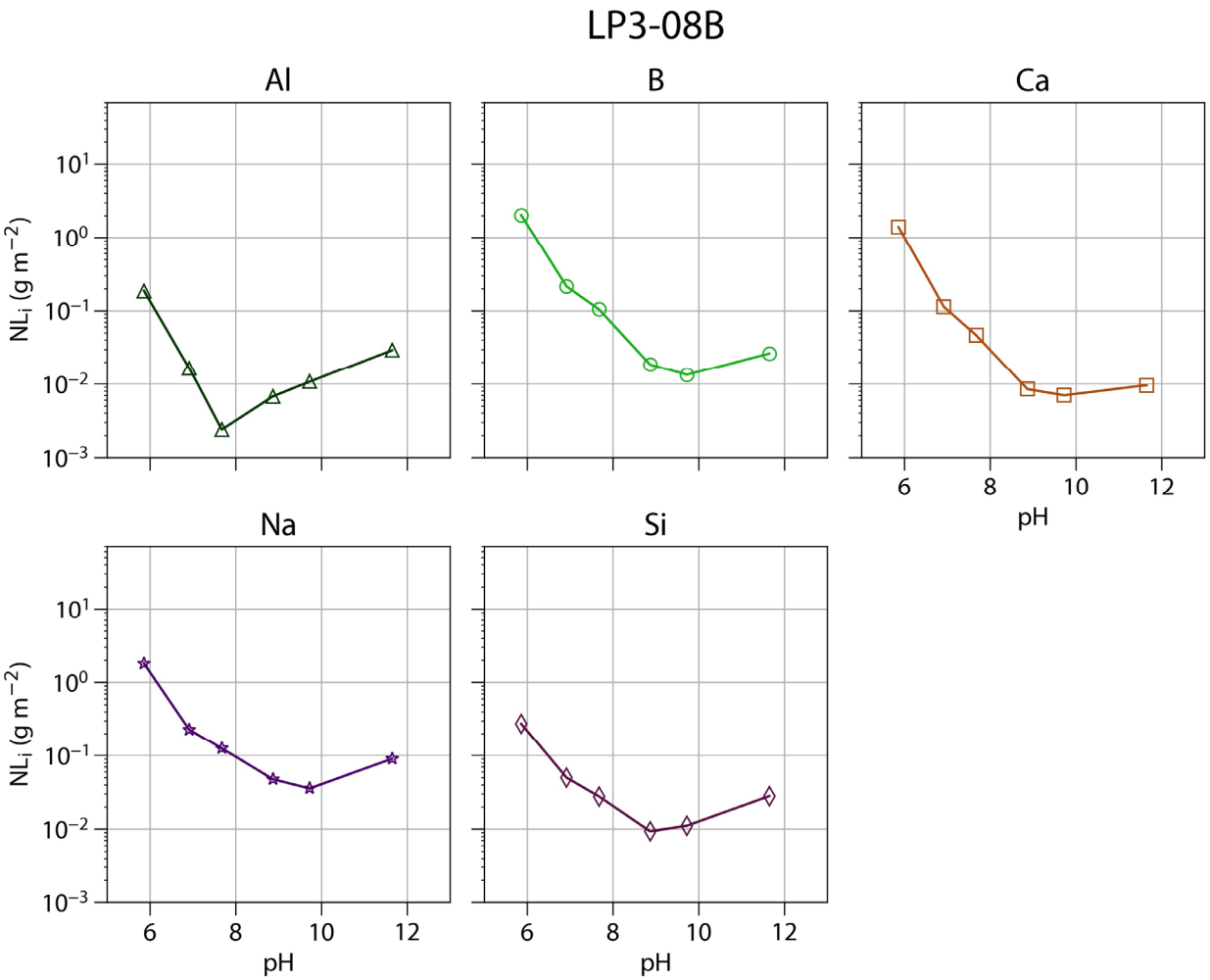

Figure B-23. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-08 (trial#2)

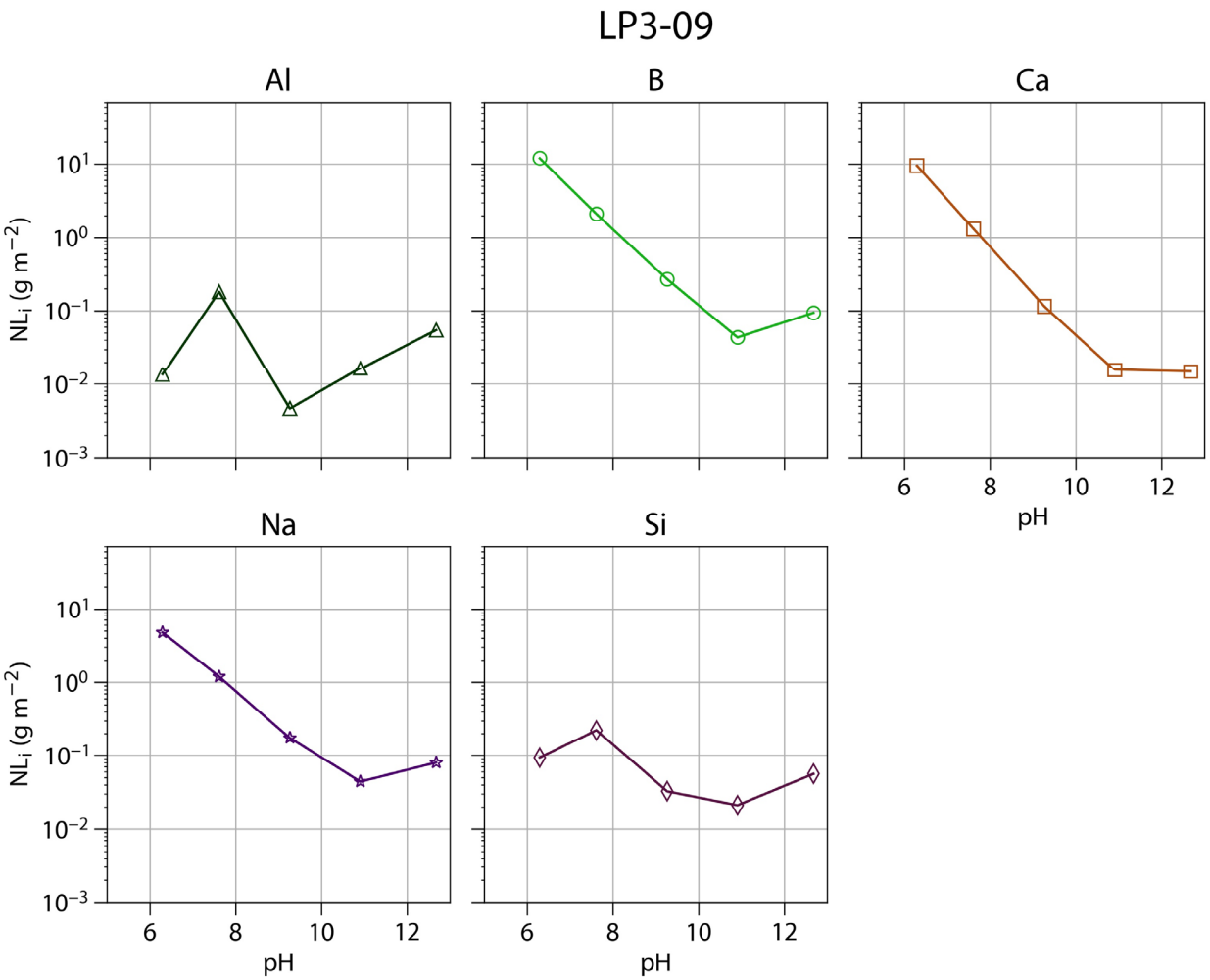

Figure B-24. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-09

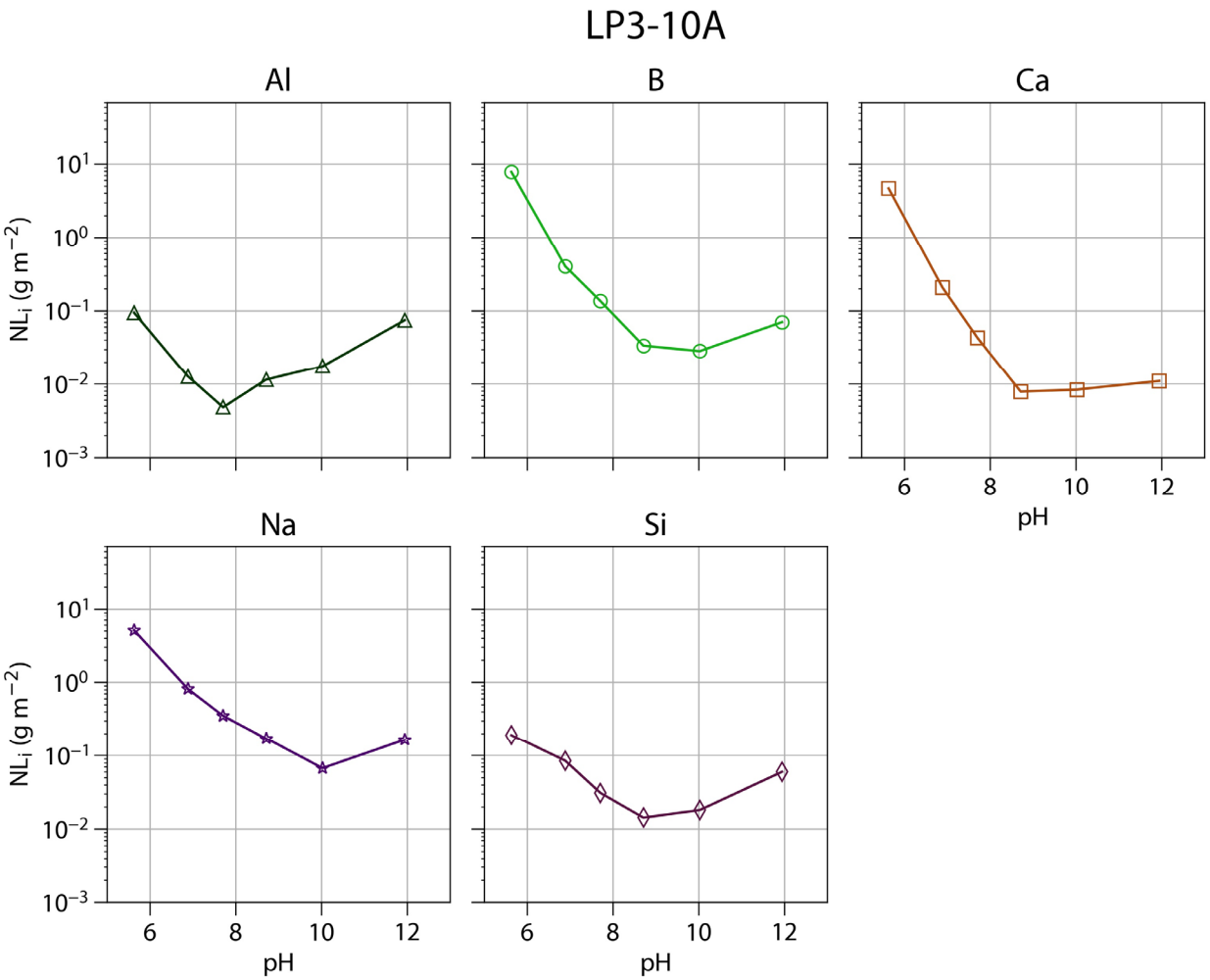

Figure B-25. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-10 (trial#1)

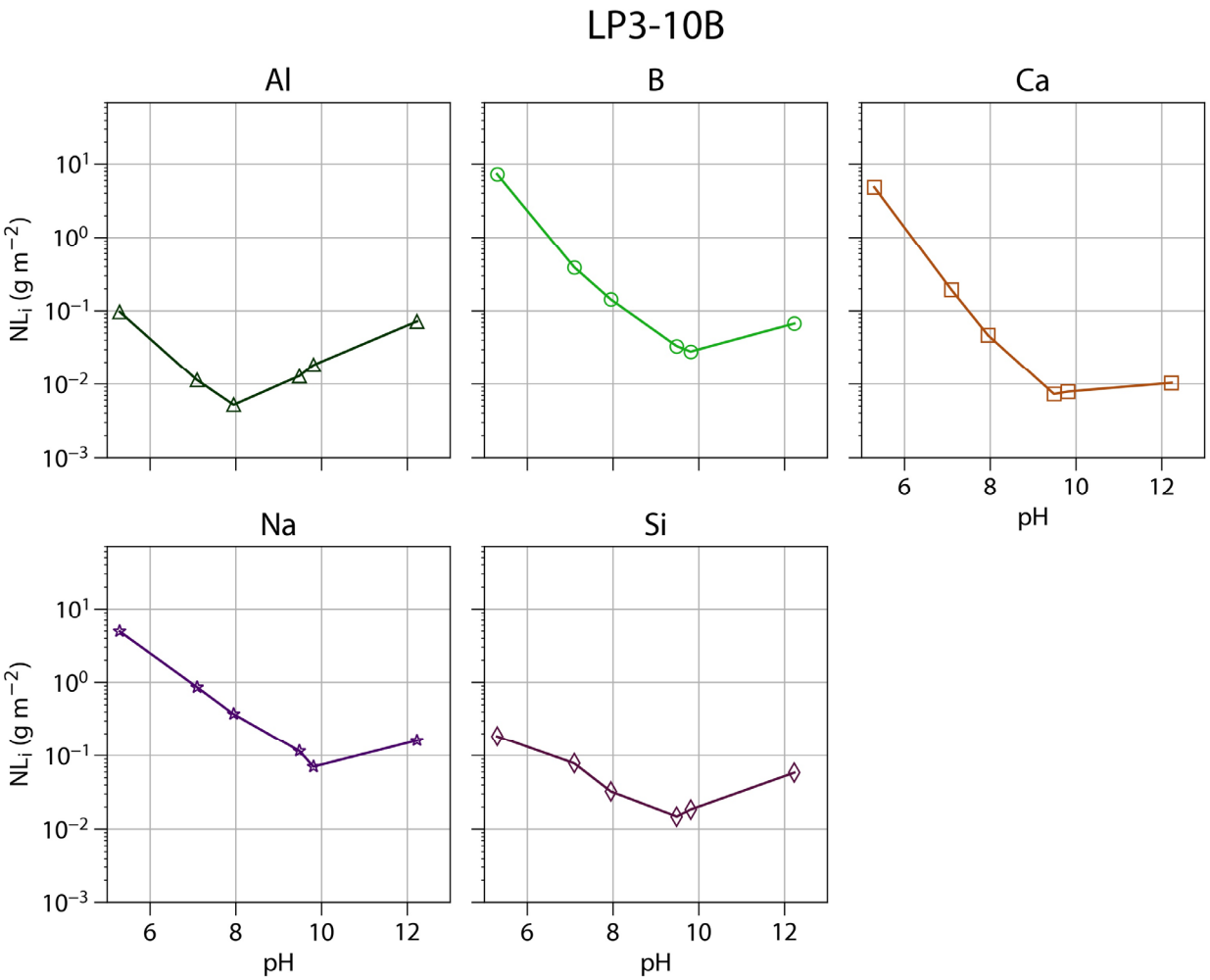

Figure B-26. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-10 (trial #2)

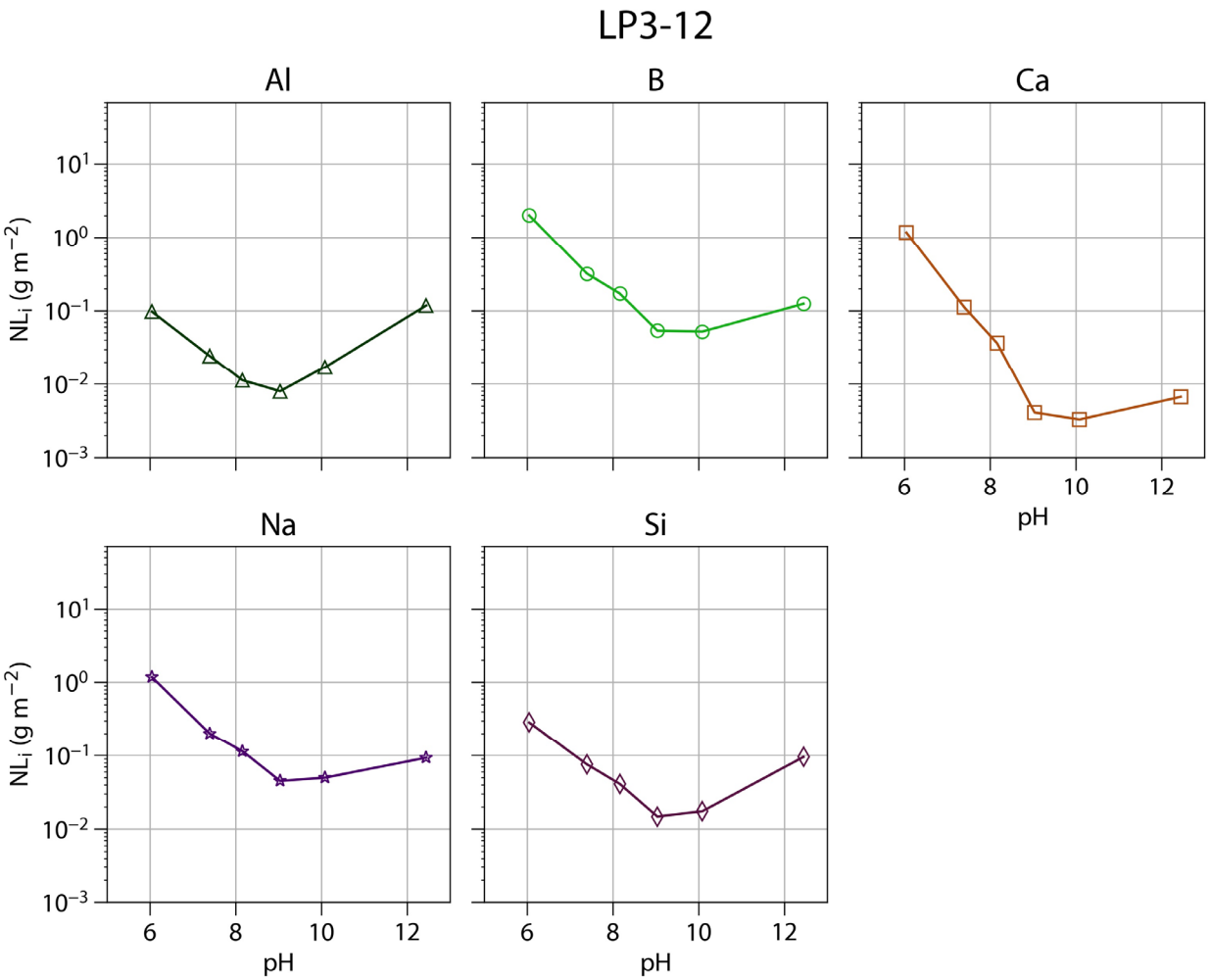

Figure B-27. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-12

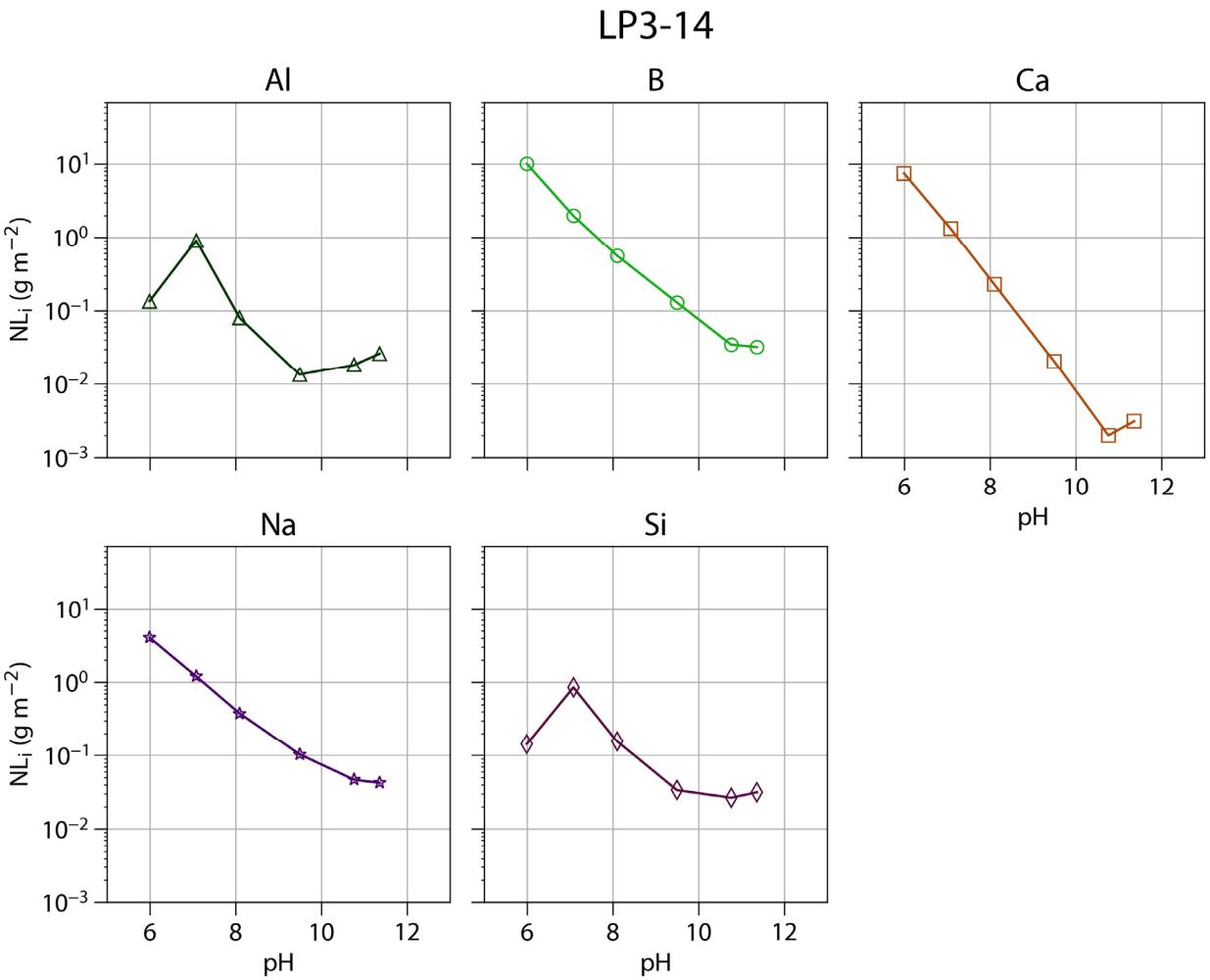

Figure B-28. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-14

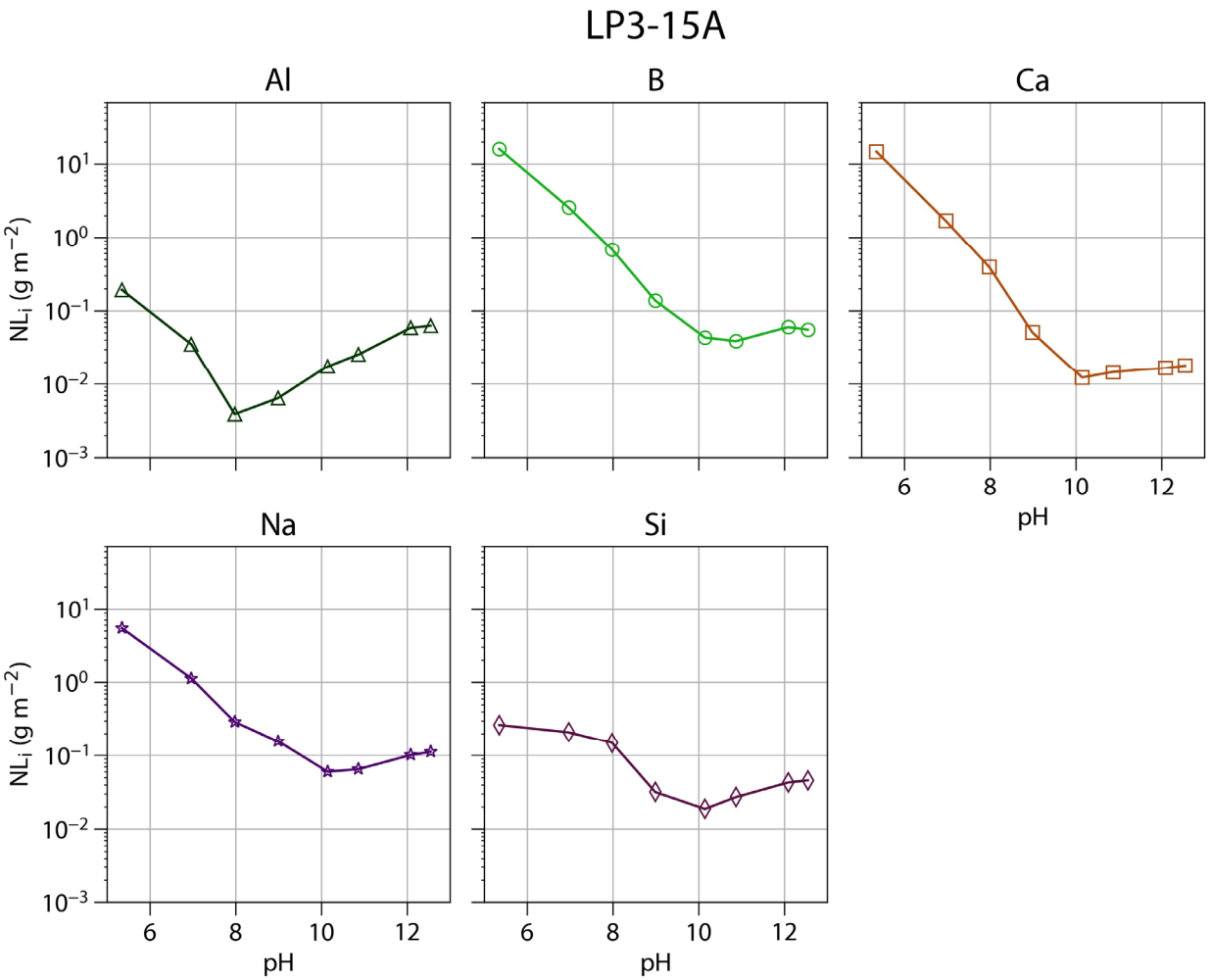

Figure B-29. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-15 (trial #1)

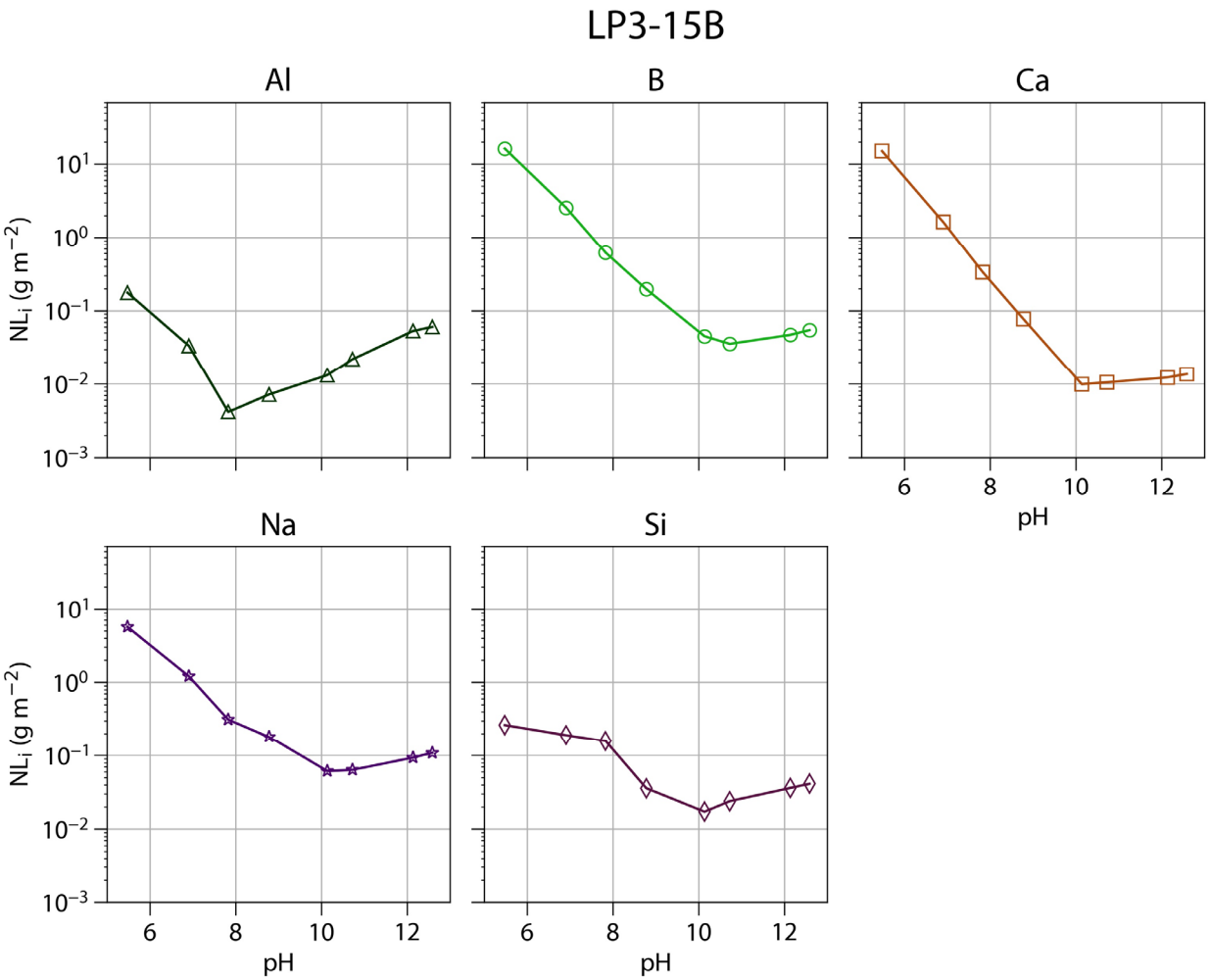

Figure B-30. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-15 (trial #2)

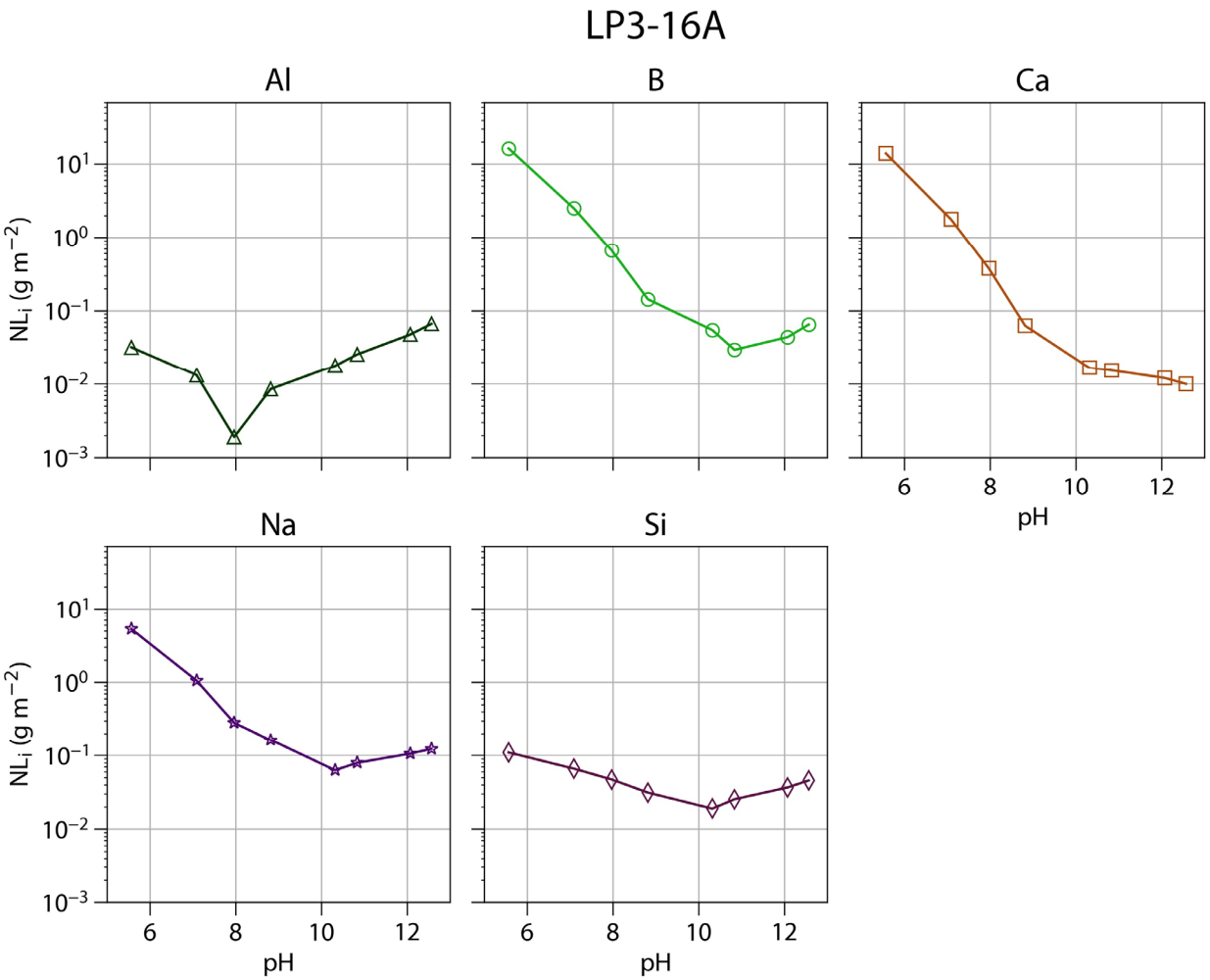

Figure B-31. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-16 (trial #1)

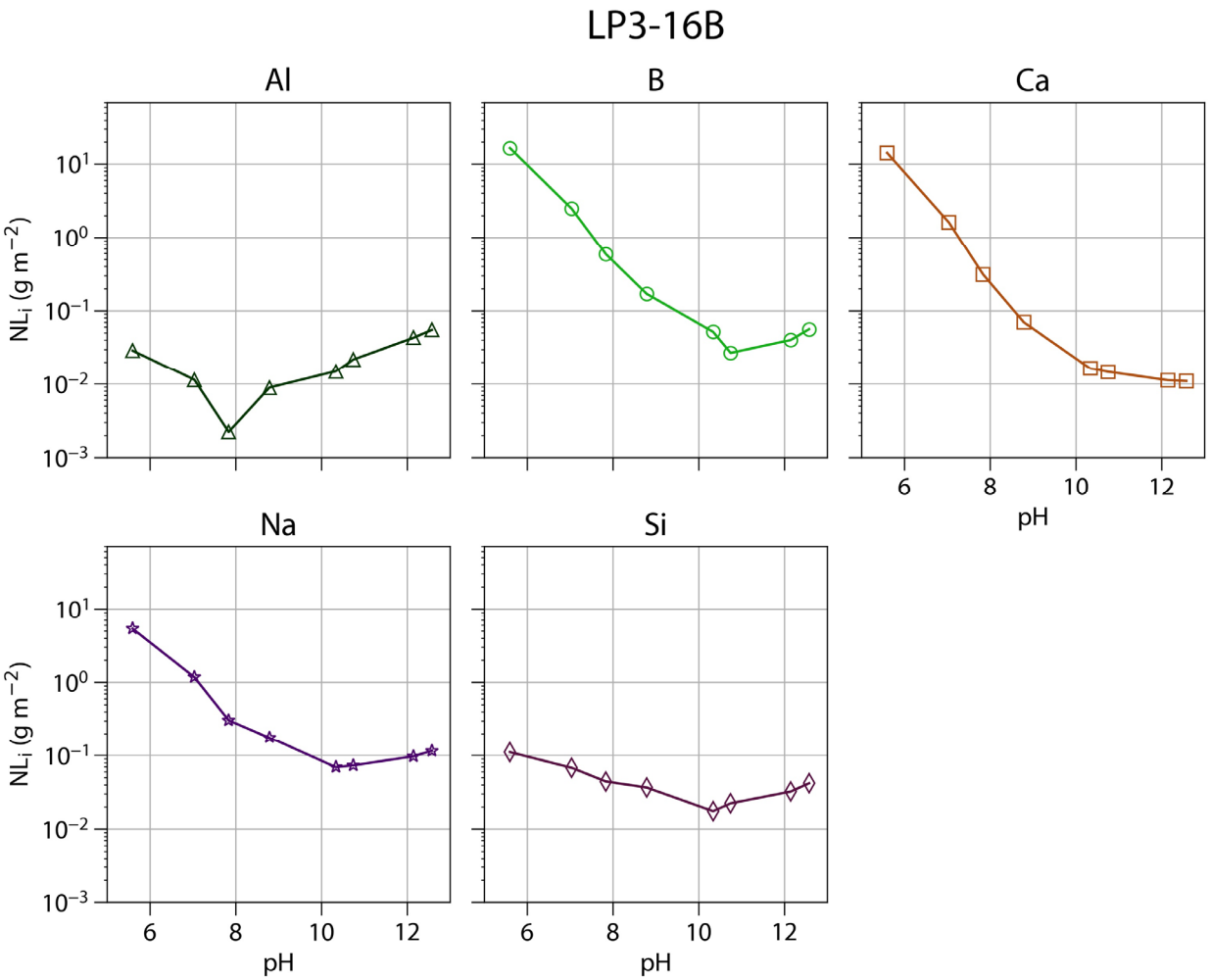

Figure B-32. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-16 (trial #2)

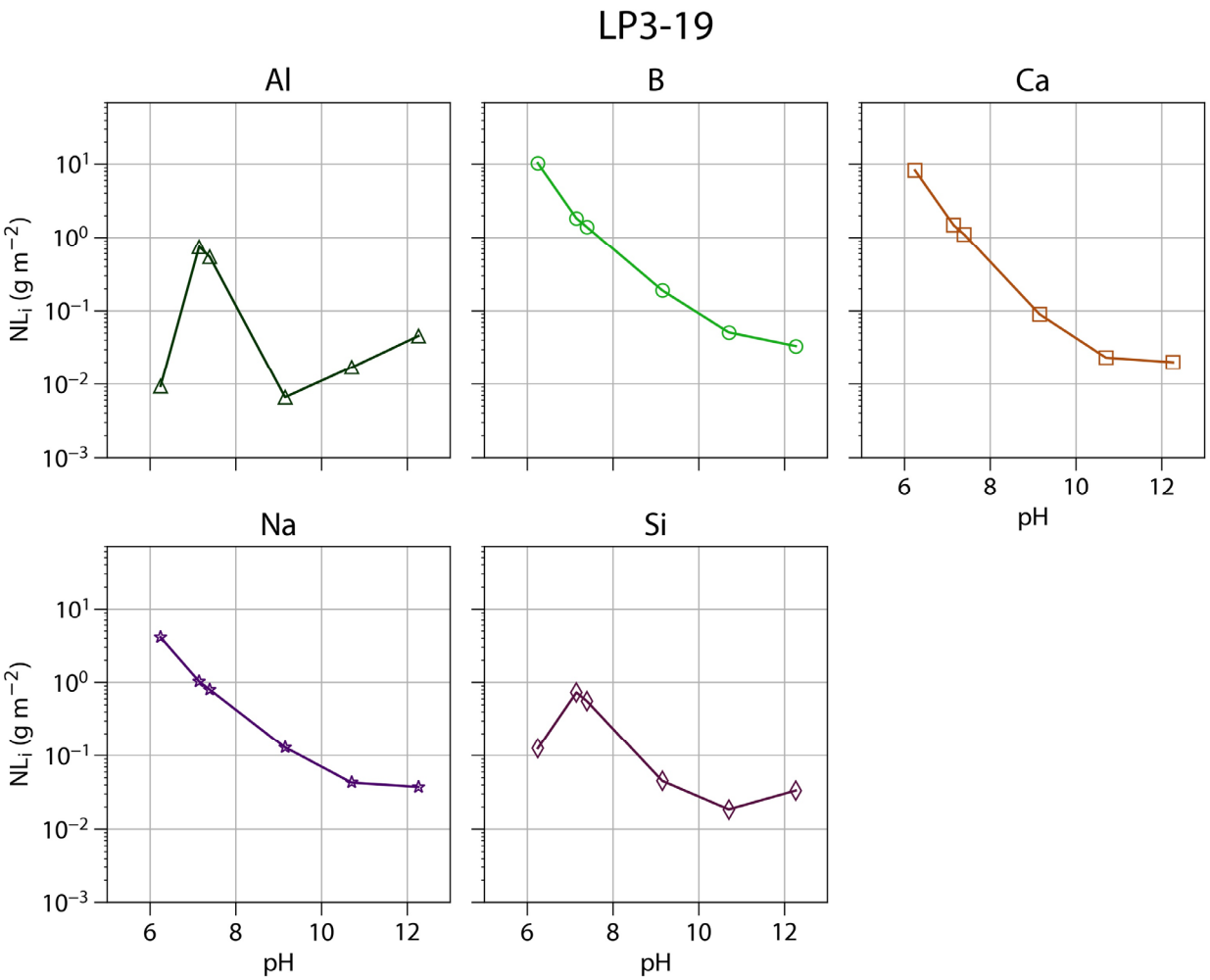

Figure B-33. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-19

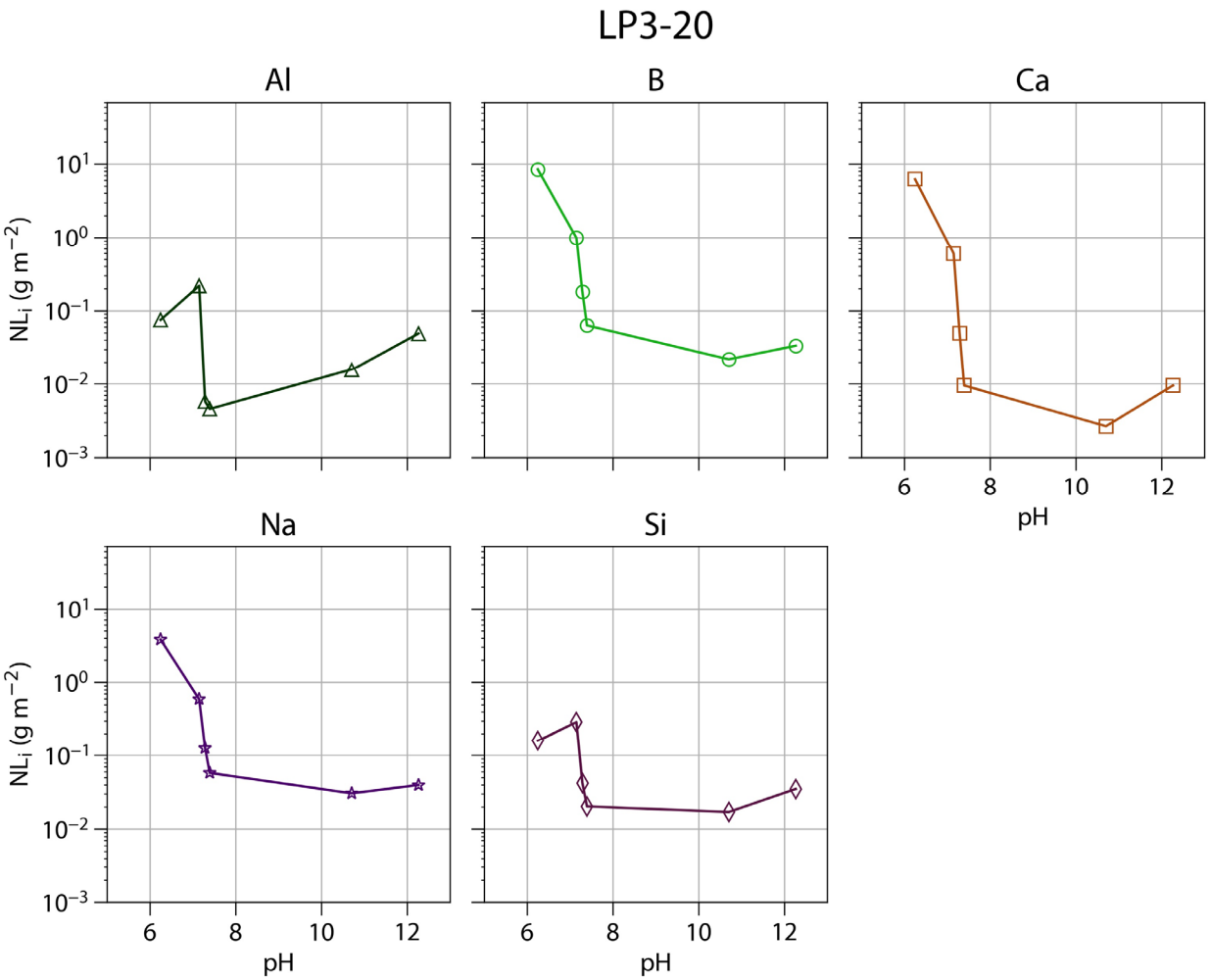

Figure B-34. EPA 1313 normalized elemental releases of Al, B, Ca, Na, and Si for LP3-20

Table B-1. Elemental releases for LP3 and join glasses (mg/L). This table is continued on the following page

	LP3-01-1		LP3-02		LP3-03		LP3-04 A		LP3-04 B		LP3-05mod6		LP3-06		LP3-07		LP3-08 A		LP3-08 B		LP3-09	
	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L	pH	conc mg/L
Al	6.171	4.05	5.85	0.562	5.901	20.65	5.5	6.99	5.51	6.82	6.303	22.75	5.572	1.87	5.804	11.95	5.993	13.809	5.865	14.013	6.296	1.17
Al	6.822	7.1	7.094	2.02	6.513	88.35	7.07	1.54	7.17	1.91	6.725	46.95	7.432	6.55	6.869	9.95	6.743	1.277	6.922	1.234	7.617	15.65
Al	7.725	42.55	7.813	36.25	7.199	13.05	8.26	0.24	8.19	0.27	8.097	1.58	7.899	3.04	8.988	0.317	7.546	0.169	7.679	0.18	9.267	0.402
Al	10.22	0.68	9.275	0.187	10.402	1.05	9.43	0.657	9.47	0.62	9.891	0.739	8.598	0.735	9.982	1.62	9.115	0.461	8.872	0.509	10.91	1.41
Al	10.715	1.27	10.117	0.91	12.643	2.71	10.3	0.959	10.32	0.986	10.153	1.12	10.0013	1.61	10.412	1.76	9.576	0.824	9.729	0.807	12.68	4.75
Al	11.969	4.41	12.338	8.89			10.76	1.3	10.85	1.305	12.478	4.99	12.467	5.52	12.548	3.9	12.384	2.476	11.652	2.192		
Al							11.97	6.46	12.02	5.86												
Al							12.53	7.59	12.54	6.879												
B	6.171	895.8	5.85	1069.8	5.901	302.8	5.5	1369.911	5.51	1371.313	6.303	283.8	5.572	596.8	5.804	624.8	5.993	91.473	5.865	91.452	6.296	660.8
B	6.822	492.8	7.094	463.8	6.513	111.8	7.07	231.88	7.17	236.383	6.725	92.5	7.432	25.1	6.869	103.8	6.743	9.395	6.922	9.677	7.617	116.8
B	7.725	164.8	7.813	139.8	7.199	34.5	8.26	68.03	8.19	60.969	8.097	21	7.899	17.2	8.988	6.52	7.546	5.019	7.679	4.721	9.267	14.8
B	10.22	7.87	9.275	25	10.402	0.8	9.43	24.65	9.47	25.451	9.891	2.22	8.598	7.61	9.982	2.41	9.115	0.874	8.872	0.835	10.91	2.41
B	10.715	5.19	10.117	6.86	12.643	2.24	10.3	14.16	10.32	13.611	10.153	2.3	10.0013	0.764	10.412	2.79	9.576	0.619	9.729	0.603	12.68	5.2
B	11.969	6.28	12.338	10			10.76	6.76	10.85	5.657	12.478	4.87	12.467	2.06	12.548	4.24	12.384	1.25	11.652	1.188		
B							11.97	8.47	12.02	6.879												
B							12.53	8.64	12.54	8.083												
Ca	6.171	726.91	5.85	1119.91	5.901	332.91	5.5	2429.836	5.51	2429.686	6.303	117.91	5.572	1679.91	5.804	1329.91	5.993	145.339	5.865	145.227	6.296	1449.91
Ca	6.822	278.91	7.094	268.91	6.513	127.91	7.07	283.23	7.17	263.796	6.725	39.71	7.432	63.91	6.869	178.91	6.743	11.322	6.922	11.822	7.617	200.91
Ca	7.725	76.31	7.813	71.91	7.199	32.51	8.26	71.36	8.19	61.649	8.097	3.69	7.899	42.21	8.988	7.7	7.546	5.317	7.679	4.835	9.267	17.51
Ca	10.22	0.326	9.275	4.53	10.402	0.226	9.43	23.08	9.47	18.995	9.891	0.094	8.598	16.91	9.982	2.01	9.115	0.927	8.872	0.883	10.91	2.37

	LP3-01-1		LP3-02		LP3-03		LP3-04A		LP3-04B		LP3-05mod6		LP3-06		LP3-07		LP3-08A		LP3-08B		LP3-09	
		conc		conc		conc		conc		conc		conc		conc		conc		conc		conc		conc
	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L
Ca	10.715	0.259	10.117	1.57	12.643	0.318	10.3	7.559	10.32	7.004	10.153	0.0784	10.0013	2.03	10.412	1.06	9.576	0.716	9.729	0.728	12.68	2.25
Ca	11.969	0.495	12.338	1.07			10.76	4.76	10.85	3.726	12.478	0.261	12.467	4.16	12.548	0.98	12.384	1.023	11.652	0.997		
Ca							11.97	5.07	12.02	4.093												
Ca							12.53	5.73	12.54	4.086												
Na	6.171	1709.54	5.85	1829.54	5.901	1079.54	5.5	1925.279	5.51	1969.974	6.303	885.54	5.572	1679.54	5.804	1649.54	5.993	568.049	5.865	573.102	6.296	1539.54
Na	6.822	1129.54	7.094	1019.54	6.513	476.54	7.07	385.05	7.17	465.617	6.725	334.54	7.432	112.54	6.869	420.54	6.743	69.123	6.922	72.242	7.617	387.54
Na	7.725	406.54	7.813	335.54	7.199	160.54	8.26	114.47	8.19	125.678	8.097	82.34	7.899	79.24	8.988	40.24	7.546	41.501	7.679	39.835	9.267	56.44
Na	10.22	23.14	9.275	64.34	10.402	10.54	9.43	63.98	9.47	58.481	9.891	13.94	8.598	38.14	9.982	21.34	9.115	14.932	8.872	15.089	10.91	14.24
Na	10.715	17.24	10.117	21.54	12.643	16.84	10.3	48.37	10.32	45.828	10.153	13.44	10.0013	8.83	10.412	24.24	9.576	11.33	9.729	11.313	12.68	25.84
Na	11.969	19.24	12.338	32.54			10.76	25.46	10.85	23.255	12.478	24.24	12.467	14.74	12.548	33.14	12.384	29.267	11.652	28.676		
Na							11.97	39.79	12.02	35.588												
Na							12.53	45.06	12.54	44.677												
Si	6.171	90.8	5.85	39.3	5.901	100	5.5	40.84	5.51	42.63	6.303	139	5.572	39	5.804	37.6	5.993	123.955	5.865	125.176	6.296	35
Si	6.822	46.1	7.094	42.5	6.513	401	7.07	28.51	7.17	30.98	6.725	263	7.432	45.2	6.869	64.5	6.743	22.618	6.922	22.786	7.617	83.2
Si	7.725	202	7.813	169	7.199	96	8.26	24.01	8.19	25.19	8.097	34.2	7.899	30	8.988	8.1	7.546	12.993	7.679	12.694	9.267	12.2
Si	10.22	9.65	9.275	13.4	10.402	5.42	9.43	22.42	9.47	19.497	9.891	6.88	8.598	13.4	9.982	4.93	9.115	4.132	8.872	4.264	10.91	7.91
Si	10.715	9.58	10.117	8.16	12.643	15.7	10.3	21.39	10.32	19.846	10.153	8.37	10.0013	5.2	10.412	6.15	9.576	5.17	9.729	5.092	12.68	21
Si	11.969	18.5	12.338	31.7			10.76	23.62	10.85	22.053	12.478	28.9	12.467	15.3	12.548	20	12.384	13.553	11.652	12.829		
Si							11.97	29.81	12.02	27.309												
Si							12.53	32.69	12.54	30.891												

Table B-1 (cont.). Elemental releases for LP3 and join glasses (mg/L). This table is continued on the following page

	LP3-10A		LP3-10B		LP3-12		LP3-14		LP3-15A		LP3-15B		LP3-16A		LP3-16B		LP3-19		LP3-20	
		conc		conc		conc		conc		conc		conc		conc		conc		conc		conc
	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L
Al	5.634	8.726	5.3	8.955	6.05	12.25	5.995	10.05	5.35	15.82	5.48	14.5	5.57	2.17	5.6	1.955	6.251	0.713	5.296	5.09
Al	6.891	1.171	7.102	1.046	7.4	3.05	7.082	67.65	6.97	2.89	6.91	2.76	7.09	0.895	7.04	0.772	7.152	57.15	6.6	14.75
Al	7.711	0.446	7.956	0.481	8.163	1.41	8.099	5.94	7.98	0.319	7.83	0.342	7.97	0.129	7.84	0.151	7.398	41.25	8.466	0.387
Al	8.714	1.06	9.493	1.193	9.04	1	9.496	1	8.99	0.527	8.78	0.591	8.82	0.577	8.79	0.604	9.156	0.502	9.266	0.308
Al	10.029	1.625	9.815	1.688	10.088	2.15	10.761	1.36	10.15	1.437	10.14	1.083	10.32	1.206	10.34	1.01	10.705	1.29	10.271	1.06
Al	11.947	6.921	12.236	6.69	12.448	14.85	11.362	1.95	10.87	2.1	10.73	1.811	10.84	1.74	10.75	1.48	12.268	3.48	12.392	3.33
Al									12.09	4.85	12.14	4.38	12.08	3.24	12.15	2.925				
Al									12.55	5.17	12.59	4.982	12.57	4.52	12.58	3.741				
B	5.634	343.223	5.3	320.238	6.05	183.8	5.995	588.8	5.35	1000.263	5.48	1012.184	5.57	765.152	5.6	774.886	6.251	472.8	5.296	488.8
B	6.891	17.637	7.102	17.096	7.4	29.2	7.082	115.8	6.97	160.75	6.91	160.139	7.09	118.32	7.04	116.908	7.152	84.3	6.6	57.5
B	7.711	5.947	7.956	6.237	8.163	15.7	8.099	32.9	7.98	42.83	7.83	38.711	7.97	31.37	7.84	27.68	7.398	64	8.466	10.4
B	8.714	1.474	9.493	1.453	9.04	4.92	9.496	7.57	8.99	8.6	8.78	12.293	8.82	6.74	8.79	7.99	9.156	8.7	9.266	3.66
B	10.029	1.242	9.815	1.226	10.088	4.78	10.761	2.03	10.15	2.705	10.14	2.822	10.32	2.58	10.34	2.439	10.705	2.35	10.271	1.27
B	11.947	3.078	12.236	2.956	12.448	11.4	11.362	1.88	10.87	2.42	10.73	2.221	10.84	1.39	10.75	1.264	12.268	1.53	12.392	1.94
B									12.09	3.75	12.14	2.942	12.08	2.07	12.15	1.89				
B									12.55	3.46	12.59	3.415	12.57	3.06	12.58	2.654				
Ca	5.634	447.988	5.3	465.763	6.05	57.01	5.995	681.91	5.35	2179.641	5.48	2219.663	5.57	1773.298	5.6	1802.169	6.251	1069.91	5.296	505.91
Ca	6.891	19.82	7.102	18.38	7.4	5.42	7.082	120.91	6.97	250.47	6.91	241.064	7.09	226.3	7.04	205.591	7.152	192.91	6.6	48.71
Ca	7.711	4.087	7.956	4.443	8.163	1.77	8.099	20.81	7.98	58.33	7.83	49.588	7.97	48.07	7.84	39.591	7.398	143.91	8.466	4.01
Ca	8.714	0.753	9.493	0.698	9.04	0.198	9.496	1.87	8.99	7.51	8.78	11.432	8.82	7.98	8.79	8.884	9.156	11.71	9.266	0.769
Ca	10.029	0.8	9.815	0.753	10.088	0.159	10.761	0.182	10.15	1.81	10.14	1.465	10.32	2.13	10.34	2.086	10.705	3.01	10.271	0.216

	LP3-10A		LP3-10B		LP3-12		LP3-14		LP3-15A		LP3-15B		LP3-16A		LP3-16B		LP3-19		LP3-20	
		conc		conc		conc		conc		conc		conc		conc		conc		conc		conc
	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L
Ca	11.947	1.05	12.236	0.988	12.448	0.323	11.362	0.283	10.87	2.15	10.73	1.554	10.84	1.95	10.75	1.865	12.268	2.6	12.392	0.77
Ca									12.09	2.46	12.14	1.814	12.08	1.53	12.15	1.429				
Ca									12.55	2.65	12.59	2.015	12.57	1.27	12.58	1.395				
Na	5.634	1803.738	5.3	1740.864	6.05	393.54	5.995	1589.54	5.35	1842.186	5.48	1914.866	5.57	1943.222	5.6	1961.163	6.251	1399.54	5.296	1299.54
Na	6.891	288.91	7.102	305.46	7.4	67.04	7.082	474.54	6.97	383.34	6.91	410.858	7.09	388.67	7.04	431.792	7.152	351.54	6.6	201.54
Na	7.711	124.62	7.956	131.22	8.163	38.14	8.099	148.54	7.98	98.49	7.83	105.859	7.97	103.68	7.84	112.18	7.398	273.54	8.466	42.64
Na	8.714	60.912	9.493	40.8	9.04	15.04	9.496	40.74	8.99	53.25	8.78	61.251	8.82	59.93	8.79	64.75	9.156	44.24	9.266	19.74
Na	10.029	23.905	9.815	25.042	10.088	16.64	10.761	18.44	10.15	20.374	10.14	20.828	10.32	23.14	10.34	25.393	10.705	14.64	10.271	10.44
Na	11.947	58.31	12.236	57.132	12.448	31.14	11.362	16.64	10.87	22.1	10.73	21.588	10.84	29.15	10.75	26.941	12.268	12.74	12.392	13.54
Na									12.09	34.39	12.14	31.735	12.08	38.91	12.15	35.694				
Na									12.55	38.05	12.59	36.927	12.57	45.14	12.58	42.555				
Si	5.634	71.252	5.3	68.468	6.05	102	5.995	57.8	5.35	93.712	5.48	93.381	5.57	40.79	5.6	41.24	6.251	51.5	5.296	68
Si	6.891	31.627	7.102	29.13	7.4	27	7.082	345	6.97	74.46	6.91	68.053	7.09	24.56	7.04	25.19	7.152	300	6.6	123
Si	7.711	11.548	7.956	12.049	8.163	14.7	8.099	62.9	7.98	53.31	7.83	56.421	7.97	17.53	7.84	16.44	7.398	228	8.466	17.9
Si	8.714	5.325	9.493	5.513	9.04	5.3	9.496	13.6	8.99	11.31	8.78	12.788	8.82	11.62	8.79	13.616	9.156	18.5	9.266	8.64
Si	10.029	6.755	9.815	6.859	10.088	6.25	10.761	10.7	10.15	6.67	10.14	6.125	10.32	7.05	10.34	6.509	10.705	7.59	10.271	7.25
Si	11.947	22.288	12.236	21.762	12.448	34.1	11.362	12.7	10.87	9.65	10.73	8.513	10.84	9.45	10.75	8.308	12.268	13.7	12.392	14.9
Si									12.09	15.23	12.14	12.861	12.08	13.66	12.15	12.012				
Si									12.55	16.34	12.59	14.644	12.57	16.99	12.58	15.573				

Table B-1 (cont.). Elemental releases for LP3 and join glasses (mg/L). This table is continued on the following page

	Dike		D2B		D6B		D6B10N		D6B14N		D6B20N		D10B20N	
		conc		conc		conc		conc		conc		conc		conc
	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L
Al	5.014	2.577	5.831	1.533	5.81	3.91	5.945	2.759	5.239	9.103	5.472	1.074	5.56	3.954
Al	6.785	0.404	7.338	0.371	6.772	1.777	7.408	0.38	7.055	0.752	6.777	0.449	6.501	0.875
Al	8.046	0.04	8.141	0.03	7.817	0.452	7.901	0.072	7.447	0.11	8.601	0.446	8.207	0.291
Al	8.291	0.055	8.412	0.085	8.634	0.077	8.151	0.085	7.932	0.121	9.454	1.062	8.889	0.621
Al	8.609	0.112	9.128	0.836	8.707	0.137	9.389	0.176	8.825	0.206	10.007	0.601	9.418	0.82
Al	12.096	6.68	12.288	4.334	11.741	14.861	12.185	8.574	11.978	3.591	11.987	10.175	12.328	12.786
B			5.831	0.215	5.81	1.082	5.945	2.422	5.239	7.436	5.472	9.823	5.56	681.245
B			7.338	0.096	6.772	1.016	7.408	0.726	7.055	3.565	6.777	1.694	6.501	355.294
B			8.141	0.034	7.817	0.56	7.901	0.414	7.447	2.556	8.601	1.089	8.207	4.628
B			8.412	0.058	8.634	0.153	8.151	0.387	7.932	1.354	9.454	0.898	8.889	3.204
B			9.128	0.063	8.707	0.122	9.389	0.419	8.825	0.729	10.007	0.695	9.418	3.149
B			12.288	0.642	11.741	1.621	12.185	3.148	11.978	4.098	11.987	6.329	12.328	15.866
Ca	5.014	2.891	5.831	2.566	5.81	6.037	5.945	8.274	5.239	22.295	5.472	18.793	5.56	878.043
Ca	6.785	1.381	7.338	1.259	6.772	3.816	7.408	2.665	7.055	7.134	6.777	2.491	6.501	397.431
Ca	8.046	0.692	8.141	0.463	7.817	2.407	7.901	1.411	7.447	3.506	8.601	2.073	8.207	4.333
Ca	8.291	0.763	8.412	0.859	8.634	0.794	8.151	1.359	7.932	3.384	9.454	2.161	8.889	3.13
Ca	8.609	1.052	9.128	1.047	8.707	0.652	9.389	1.249	8.825	2.266	10.007	1.492	9.418	3.019
Ca	12.096	4.345	12.288	3.467	11.741	3.369	12.185	4.996	11.978	5.663	11.987	5.267	12.328	2.014
Na	5.014	1.788	5.831	1.208	5.81	2.534	5.945	12.06	5.239	43.512	5.472	91.055	5.56	1309.738
Na	6.785	1.326	7.338	0.941	6.772	2.31	7.408	4.861	7.055	10.797	6.777	21.237	6.501	1048.392
Na	8.046	1.07	8.141	0.658	7.817	1.27	7.901	3.677	7.447	6.065	8.601	16.872	8.207	24.255

	Dike		D2B		D6B		D6B10N		D6B14N		D6B20N		D10B20N	
		conc		conc		conc		conc		conc		conc		conc
	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L	pH	mg/L
Na	8.291	0.974	8.412	0.876	8.634	0.802	8.151	3.288	7.932	7.676	9.454	15.268	8.889	18.422
Na	8.609	1.285	9.128	1.104	8.707	0.65	9.389	3.238	8.825	9.472	10.007	14.435	9.418	18.14
Na	12.096	8.262	12.288	8.057	11.741	6.75	12.185	17.657	11.978	27.788	11.987	52.565	12.328	76.565
Si	5.014	4.366	5.831	2.841	5.81	6.925	5.945	9.18	5.239	32.328	5.472	23.838	5.56	62.568
Si	6.785	2.141	7.338	2.533	6.772	4.844	7.408	2.902	7.055	4.509	6.777	5.445	6.501	56.029
Si	8.046	1.916	8.141	1.469	7.817	2.806	7.901	2.651	7.447	1.905	8.601	4.7	8.207	5.534
Si	8.291	1.861	8.412	2.171	8.634	1.693	8.151	2.497	7.932	2.546	9.454	4.291	8.889	5.424
Si	8.609	2.849	9.128	2.095	8.707	1.322	9.389	2.372	8.825	5.307	10.007	2.92	9.418	5.151
Si	12.096	24.075	12.288	21.466	11.741	26.235	12.185	31.815	11.978	31.142	11.987	44.33	12.328	57.398

Table B-2. Normalized mass loss for LP3 and join glasses (g/m^2). This table is continued on the following page

	LP3-01-1		LP3-02		LP3-03		LP3-04 A		LP3-04 B		LP3-05mod6		LP3-06		LP3-07		LP3-08 A		LP3-08 B		LP3-09	
	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})
Al	6.171	0.055	5.85	0.007	5.901	0.281	5.5	0.096	5.51	0.094	6.303	0.321	5.572	0.018	5.804	0.110	5.993	0.183	5.865	0.186	6.296	0.013
Al	6.822	0.096	7.094	0.026	6.513	1.200	7.07	0.021	7.17	0.026	6.725	0.663	7.432	0.064	6.869	0.092	6.743	0.017	6.922	0.016	7.617	0.180
Al	7.725	0.576	7.813	0.461	7.199	0.177	8.26	0.003	8.19	0.004	8.097	0.022	7.899	0.030	8.988	0.003	7.546	0.002	7.679	0.002	9.267	0.005
Al	10.22	0.009	9.275	0.002	10.402	0.014	9.43	0.009	9.47	0.009	9.891	0.010	8.598	0.007	9.982	0.015	9.115	0.006	8.872	0.007	10.91	0.016
Al	10.715	0.017	10.117	0.012	12.643	0.037	10.3	0.013	10.32	0.014	10.153	0.016	10.0013	0.016	10.412	0.016	9.576	0.011	9.729	0.011	12.68	0.055
Al	11.969	0.060	12.338	0.113			10.76	0.018	10.85	0.018	12.478	0.070	12.467	0.054	12.548	0.036	12.384	0.033	11.652	0.029	0	0.000
Al							11.97	0.089	12.02	0.081												
Al							12.53	0.104	12.54	0.095												
B	6.171	9.220	5.85	12.307	5.901	6.430	5.5	16.057	5.51	16.074	6.303	5.014	5.572	11.929	5.804	12.135	5.993	2.022	5.865	2.021	6.296	11.947
B	6.822	5.072	7.094	5.336	6.513	2.374	7.07	2.718	7.17	2.771	6.725	1.634	7.432	0.502	6.869	2.016	6.743	0.208	6.922	0.214	7.617	2.112
B	7.725	1.696	7.813	1.608	7.199	0.733	8.26	0.797	8.19	0.715	8.097	0.371	7.899	0.344	8.988	0.127	7.546	0.111	7.679	0.104	9.267	0.268
B	10.22	0.081	9.275	0.288	10.402	0.017	9.43	0.289	9.47	0.298	9.891	0.039	8.598	0.152	9.982	0.047	9.115	0.019	8.872	0.018	10.91	0.044
B	10.715	0.053	10.117	0.079	12.643	0.048	10.3	0.166	10.32	0.160	10.153	0.041	10.0013	0.015	10.412	0.054	9.576	0.014	9.729	0.013	12.68	0.094
B	11.969	0.065	12.338	0.115			10.76	0.079	10.85	0.066	12.478	0.086	12.467	0.041	12.548	0.082	12.384	0.028	11.652	0.026	0	0.000
B							11.97	0.099	12.02	0.081												
B							12.53	0.101	12.54	0.095												
Ca	6.171	9.537	5.85	13.690	5.901	4.737	5.5	15.296	5.51	15.295	6.303	3.115	5.572	10.268	5.804	9.080	5.993	1.394	5.865	1.393	6.296	9.496
Ca	6.822	3.659	7.094	3.287	6.513	1.820	7.07	1.783	7.17	1.661	6.725	1.049	7.432	0.391	6.869	1.222	6.743	0.109	6.922	0.113	7.617	1.316
Ca	7.725	1.001	7.813	0.879	7.199	0.463	8.26	0.449	8.19	0.388	8.097	0.097	7.899	0.258	8.988	0.053	7.546	0.051	7.679	0.046	9.267	0.115
Ca	10.22	0.004	9.275	0.055	10.402	0.003	9.43	0.145	9.47	0.120	9.891	0.002	8.598	0.103	9.982	0.014	9.115	0.009	8.872	0.008	10.91	0.016
Ca	10.715	0.003	10.117	0.019	12.643	0.005	10.3	0.048	10.32	0.044	10.153	0.002	10.0013	0.012	10.412	0.007	9.576	0.007	9.729	0.007	12.68	0.015
Ca	11.969	0.006	12.338	0.013			10.76	0.030	10.85	0.023	12.478	0.007	12.467	0.025	12.548	0.007	12.384	0.010	11.652	0.010	0	0.000

	LP3-01-1		LP3-02		LP3-03		LP3-04 A		LP3-04 B		LP3-05mod6		LP3-06		LP3-07		LP3-08 A		LP3-08 B		LP3-09	
	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})	pH	NL (g m^{-2})
Ca							11.97	0.032	12.02	0.026												
Ca							12.53	0.036	12.54	0.026												
Na	6.171	4.659	5.85	5.126	5.901	3.416	5.5	5.747	5.51	5.881	6.303	2.452	5.572	5.228	5.804	4.393	5.993	1.786	5.865	1.802	6.296	4.759
Na	6.822	3.078	7.094	2.856	6.513	1.508	7.07	1.149	7.17	1.390	6.725	0.927	7.432	0.350	6.869	1.120	6.743	0.217	6.922	0.227	7.617	1.198
Na	7.725	1.108	7.813	0.940	7.199	0.508	8.26	0.342	8.19	0.375	8.097	0.228	7.899	0.247	8.988	0.107	7.546	0.130	7.679	0.125	9.267	0.174
Na	10.22	0.063	9.275	0.180	10.402	0.033	9.43	0.191	9.47	0.175	9.891	0.039	8.598	0.119	9.982	0.057	9.115	0.047	8.872	0.047	10.91	0.044
Na	10.715	0.047	10.117	0.060	12.643	0.053	10.3	0.144	10.32	0.137	10.153	0.037	10.0013	0.027	10.412	0.065	9.576	0.036	9.729	0.036	12.68	0.080
Na	11.969	0.052	12.338	0.091			10.76	0.076	10.85	0.069	12.478	0.067	12.467	0.046	12.548	0.088	12.384	0.092	11.652	0.090	0	0.000
Na							11.97	0.119	12.02	0.106												
Na							12.53	0.135	12.54	0.133												
Si	6.171	0.247	5.85	0.112	5.901	0.234	5.5	0.112	5.51	0.117	6.303	0.351	5.572	0.106	5.804	0.106	5.993	0.271	5.865	0.274	6.296	0.094
Si	6.822	0.126	7.094	0.121	6.513	0.938	7.07	0.078	7.17	0.085	6.725	0.664	7.432	0.123	6.869	0.183	6.743	0.049	6.922	0.050	7.617	0.222
Si	7.725	0.551	7.813	0.482	7.199	0.225	8.26	0.066	8.19	0.069	8.097	0.086	7.899	0.082	8.988	0.023	7.546	0.028	7.679	0.028	9.267	0.033
Si	10.22	0.026	9.275	0.038	10.402	0.013	9.43	0.061	9.47	0.053	9.891	0.017	8.598	0.036	9.982	0.014	9.115	0.009	8.872	0.009	10.91	0.021
Si	10.715	0.026	10.117	0.023	12.643	0.037	10.3	0.058	10.32	0.054	10.153	0.021	10.0013	0.014	10.412	0.017	9.576	0.011	9.729	0.011	12.68	0.056
Si	11.969	0.050	12.338	0.090			10.76	0.065	10.85	0.060	12.478	0.073	12.467	0.042	12.548	0.057	12.384	0.030	11.652	0.028	0	0.000
Si							11.97	0.081	12.02	0.075												
Si							12.53	0.089	12.54	0.084												

Table B-2. (cont.) Normalized mass loss for LP3 and join glasses (g m^{-2}). This table is continued on the following page

	LP3-10A		LP3-10B		LP3-12		LP3-14		LP3-15A		LP3-15B		LP3-16A		LP3-16B		LP3-19		LP3-20	
	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})
Al	5.634	0.094	5.3	0.096	6.05	0.097	5.995	0.135	5.35	0.192	5.48	0.176	5.57	0.032	5.6	0.029	6.251	0.009	6.251	0.075
Al	6.891	0.013	7.102	0.011	7.4	0.024	7.082	0.907	6.97	0.035	6.91	0.033	7.09	0.013	7.04	0.011	7.152	0.748	7.152	0.218
Al	7.711	0.005	7.956	0.005	8.163	0.011	8.099	0.080	7.98	0.004	7.83	0.004	7.97	0.002	7.84	0.002	7.398	0.540	7.291	0.006
Al	8.714	0.011	9.493	0.013	9.04	0.008	9.496	0.013	8.99	0.006	8.78	0.007	8.82	0.008	8.79	0.009	9.156	0.007	7.398	0.005
Al	10.029	0.018	9.815	0.018	10.088	0.017	10.761	0.018	10.15	0.017	10.14	0.013	10.32	0.018	10.34	0.015	10.705	0.017	10.705	0.016
Al	11.947	0.075	12.236	0.072	12.448	0.118	11.362	0.026	10.87	0.025	10.73	0.022	10.84	0.026	10.75	0.022	12.268	0.046	12.268	0.049
Al									12.09	0.059	12.14	0.053	12.08	0.048	12.15	0.043				
Al									12.55	0.063	12.59	0.060	12.57	0.066	12.58	0.055				
B	5.634	7.819	5.3	7.295	6.05	2.006	5.995	10.071	5.35	16.004	5.48	16.195	5.57	16.246	5.6	16.452	6.251	10.255	6.251	8.454
B	6.891	0.402	7.102	0.389	7.4	0.319	7.082	1.981	6.97	2.572	6.91	2.562	7.09	2.512	7.04	2.482	7.152	1.828	7.152	0.995
B	7.711	0.135	7.956	0.142	8.163	0.171	8.099	0.563	7.98	0.685	7.83	0.619	7.97	0.666	7.84	0.588	7.398	1.388	7.291	0.180
B	8.714	0.034	9.493	0.033	9.04	0.054	9.496	0.129	8.99	0.138	8.78	0.197	8.82	0.143	8.79	0.170	9.156	0.189	7.398	0.063
B	10.029	0.028	9.815	0.028	10.088	0.052	10.761	0.035	10.15	0.043	10.14	0.045	10.32	0.055	10.34	0.052	10.705	0.051	10.705	0.022
B	11.947	0.070	12.236	0.067	12.448	0.124	11.362	0.032	10.87	0.039	10.73	0.036	10.84	0.030	10.75	0.027	12.268	0.033	12.268	0.034
B									12.09	0.060	12.14	0.047	12.08	0.044	12.15	0.040				
B									12.55	0.055	12.59	0.055	12.57	0.065	12.58	0.056				
Ca	5.634	4.669	5.3	4.854	6.05	1.173	5.995	7.475	5.35	14.696	5.48	14.966	5.57	13.898	5.6	14.124	6.251	8.188	6.251	6.221
Ca	6.891	0.207	7.102	0.192	7.4	0.111	7.082	1.325	6.97	1.689	6.91	1.625	7.09	1.774	7.04	1.611	7.152	1.476	7.152	0.599
Ca	7.711	0.043	7.956	0.046	8.163	0.036	8.099	0.228	7.98	0.393	7.83	0.334	7.97	0.377	7.84	0.310	7.398	1.101	7.291	0.049
Ca	8.714	0.008	9.493	0.007	9.04	0.004	9.496	0.020	8.99	0.051	8.78	0.077	8.82	0.063	8.79	0.070	9.156	0.090	7.398	0.009
Ca	10.029	0.008	9.815	0.008	10.088	0.003	10.761	0.002	10.15	0.012	10.14	0.010	10.32	0.017	10.34	0.016	10.705	0.023	10.705	0.003

	LP3-10A		LP3-10B		LP3-12		LP3-14		LP3-15A		LP3-15B		LP3-16A		LP3-16B		LP3-19		LP3-20	
	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)
Ca	11.947	0.011	12.236	0.010	12.448	0.007	11.362	0.003	10.87	0.014	10.73	0.010	10.84	0.015	10.75	0.015	12.268	0.020	12.268	0.009
Ca									12.09	0.017	12.14	0.012	12.08	0.012	12.15	0.011				
Ca									12.55	0.018	12.59	0.014	12.57	0.010	12.58	0.011				
Na	5.634	5.076	5.3	4.899	6.05	1.178	5.995	4.040	5.35	5.429	5.48	5.643	5.57	5.288	5.6	5.337	6.251	4.084	6.251	3.804
Na	6.891	0.813	7.102	0.860	7.4	0.201	7.082	1.206	6.97	1.130	6.91	1.211	7.09	1.058	7.04	1.175	7.152	1.026	7.152	0.590
Na	7.711	0.351	7.956	0.369	8.163	0.114	8.099	0.378	7.98	0.290	7.83	0.312	7.97	0.282	7.84	0.305	7.398	0.798	7.291	0.125
Na	8.714	0.171	9.493	0.115	9.04	0.045	9.496	0.104	8.99	0.157	8.78	0.181	8.82	0.163	8.79	0.176	9.156	0.129	7.398	0.058
Na	10.029	0.067	9.815	0.070	10.088	0.050	10.761	0.047	10.15	0.060	10.14	0.061	10.32	0.063	10.34	0.069	10.705	0.043	10.705	0.031
Na	11.947	0.164	12.236	0.161	12.448	0.093	11.362	0.042	10.87	0.065	10.73	0.064	10.84	0.079	10.75	0.073	12.268	0.037	12.268	0.040
Na									12.09	0.101	12.14	0.094	12.08	0.106	12.15	0.097				
Na									12.55	0.112	12.59	0.109	12.57	0.123	12.58	0.116				
Si	5.634	0.191	5.3	0.184	6.05	0.285	5.995	0.144	5.35	0.263	5.48	0.263	5.57	0.110	5.6	0.111	6.251	0.125	6.251	0.160
Si	6.891	0.085	7.102	0.078	7.4	0.075	7.082	0.858	6.97	0.209	6.91	0.191	7.09	0.066	7.04	0.068	7.152	0.730	7.152	0.290
Si	7.711	0.031	7.956	0.032	8.163	0.041	8.099	0.156	7.98	0.150	7.83	0.159	7.97	0.047	7.84	0.044	7.398	0.555	7.291	0.042
Si	8.714	0.014	9.493	0.015	9.04	0.015	9.496	0.034	8.99	0.032	8.78	0.036	8.82	0.031	8.79	0.037	9.156	0.045	7.398	0.020
Si	10.029	0.018	9.815	0.018	10.088	0.017	10.761	0.027	10.15	0.019	10.14	0.017	10.32	0.019	10.34	0.018	10.705	0.018	10.705	0.017
Si	11.947	0.060	12.236	0.058	12.448	0.095	11.362	0.032	10.87	0.027	10.73	0.024	10.84	0.025	10.75	0.022	12.268	0.033	12.268	0.035
Si									12.09	0.043	12.14	0.036	12.08	0.037	12.15	0.032				
Si									12.55	0.046	12.59	0.041	12.57	0.046	12.58	0.042				

Table B-2. (cont.) Normalized mass loss for LP3 glasses (g m^{-2}). This table is continued on the following page

	Dike		D2B		D6B		D6B10N		D6B14N		D6B20N		D10B20N	
	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})	pH	conc NL (g m^{-2})
Al	5.014	1.9E-02	5.831	1.3E-02	5.81	3.6E-02	5.945	2.7E-02	5.239	9.4E-02	5.472	1.1E-02	5.56	4.5E-02
Al	6.785	3.0E-03	7.338	3.1E-03	6.772	1.6E-02	7.408	3.7E-03	7.055	7.8E-03	6.777	4.7E-03	6.501	9.9E-03
Al	8.046	2.9E-04	8.141	2.5E-04	7.817	4.2E-03	7.901	7.1E-04	7.447	1.1E-03	8.601	4.7E-03	8.207	3.3E-03
Al	8.291	4.0E-04	8.412	7.1E-04	8.634	7.1E-04	8.151	8.3E-04	7.932	1.3E-03	9.454	1.1E-02	8.889	7.0E-03
Al	8.609	8.2E-04	9.128	7.0E-03	8.707	1.3E-03	9.389	1.7E-03	8.825	2.1E-03	10.007	6.4E-03	9.418	9.3E-03
Al	12.096	4.9E-02	12.288	3.6E-02	11.741	1.4E-01	12.185	8.4E-02	11.978	3.7E-02	11.987	1.1E-01	12.328	1.4E-01
B			5.831	2.1E-02	5.81	3.1E-02	5.945	7.3E-02	5.239	2.4E-01	5.472	3.2E-01	5.56	1.2E+01
B			7.338	9.3E-03	6.772	2.9E-02	7.408	2.2E-02	7.055	1.2E-01	6.777	5.6E-02	6.501	6.4E+00
B			8.141	3.3E-03	7.817	1.6E-02	7.901	1.3E-02	7.447	8.3E-02	8.601	3.6E-02	8.207	8.3E-02
B			8.412	5.6E-03	8.634	4.4E-03	8.151	1.2E-02	7.932	4.4E-02	9.454	3.0E-02	8.889	5.8E-02
B			9.128	6.1E-03	8.707	3.5E-03	9.389	1.3E-02	8.825	2.4E-02	10.007	2.3E-02	9.418	5.7E-02
B			12.288	6.2E-02	11.741	4.6E-02	12.185	9.5E-02	11.978	1.3E-01	11.987	2.1E-01	12.328	2.9E-01
Ca	5.014	2.3E-02	5.831	2.0E-02	5.81	5.2E-02	5.945	7.7E-02	5.239	2.2E-01	5.472	2.0E-01	5.56	1.0E+01
Ca	6.785	1.1E-02	7.338	1.0E-02	6.772	3.3E-02	7.408	2.5E-02	7.055	7.0E-02	6.777	2.6E-02	6.501	4.5E+00
Ca	8.046	5.5E-03	8.141	3.7E-03	7.817	2.1E-02	7.901	1.3E-02	7.447	3.4E-02	8.601	2.2E-02	8.207	4.9E-02
Ca	8.291	6.1E-03	8.412	6.9E-03	8.634	6.8E-03	8.151	1.3E-02	7.932	3.3E-02	9.454	2.3E-02	8.889	3.6E-02
Ca	8.609	8.4E-03	9.128	8.4E-03	8.707	5.6E-03	9.389	1.2E-02	8.825	2.2E-02	10.007	1.6E-02	9.418	3.4E-02
Ca	12.096	3.5E-02	12.288	2.8E-02	11.741	2.9E-02	12.185	4.6E-02	11.978	5.5E-02	11.987	5.6E-02	12.328	2.3E-02
Na	5.014	4.3E-02	5.831	4.0E-02	5.81	8.9E-02	5.945	8.2E-02	5.239	2.1E-01	5.472	3.1E-01	5.56	4.5E+00
Na	6.785	3.2E-02	7.338	3.1E-02	6.772	8.1E-02	7.408	3.3E-02	7.055	5.3E-02	6.777	7.2E-02	6.501	3.6E+00
Na	8.046	2.6E-02	8.141	2.2E-02	7.817	4.4E-02	7.901	2.5E-02	7.447	3.0E-02	8.601	5.8E-02	8.207	8.4E-02

	Dike		D2B		D6B		D6B10N		D6B14N		D6B20N		D10B20N	
	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)	pH	conc NL ($g\ m^{-2}$)
Na	8.291	2.3E-02	8.412	2.9E-02	8.634	2.8E-02	8.151	2.2E-02	7.932	3.7E-02	9.454	5.2E-02	8.889	6.4E-02
Na	8.609	3.1E-02	9.128	3.7E-02	8.707	2.3E-02	9.389	2.2E-02	8.825	4.6E-02	10.007	4.9E-02	9.418	6.3E-02
Na	12.096	2.0E-01	12.288	2.7E-01	11.741	2.4E-01	12.185	1.2E-01	11.978	1.4E-01	11.987	1.8E-01	12.328	2.6E-01
Si	5.014	8.6E-03	5.831	5.7E-03	5.81	1.4E-02	5.945	2.0E-02	5.239	7.4E-02	5.472	6.0E-02	5.56	1.6E-01
Si	6.785	4.2E-03	7.338	5.1E-03	6.772	9.8E-03	7.408	6.4E-03	7.055	1.0E-02	6.777	1.4E-02	6.501	1.5E-01
Si	8.046	3.8E-03	8.141	2.9E-03	7.817	5.7E-03	7.901	5.9E-03	7.447	4.4E-03	8.601	1.2E-02	8.207	1.4E-02
Si	8.291	3.7E-03	8.412	4.3E-03	8.634	3.4E-03	8.151	5.5E-03	7.932	5.8E-03	9.454	1.1E-02	8.889	1.4E-02
Si	8.609	5.6E-03	9.128	4.2E-03	8.707	2.7E-03	9.389	5.3E-03	8.825	1.2E-02	10.007	7.3E-03	9.418	1.3E-02
Si	12.096	4.8E-02	12.288	4.3E-02	11.741	5.3E-02	12.185	7.1E-02	11.978	7.2E-02	11.987	1.1E-01	12.328	1.5E-01

Table B-3. Calculated equivalent thickness for VHT and PCT of LP3 glasses (Lonergan et al. 2020).

Glass ID	Alteration rate (VHT) g m^{-2} d^{-1}	24 d VHT alteration thickness, µm	PCT-Na normalized release g m^{-2}	7 d PCT calculated equivalent layer thickness, µm
LAWPh3-01-1	>68.3	>645	3.734	1.409
LAWPh3-02	>77.8	>735	6.559	2.475
LAWPh3-03	>80.0	>775	3.257	1.229
LAWPh3-04	30.1	272	1.628	0.614
LAWPh3-05-mod6	241	640	5.190	1.958
LAWPh3-06	6.48	57.1	0.993	0.375
LAWPh3-07	>85.5	>775	1.168	0.441
LAWPh3-08	0.59	5.24	0.654	0.247
LAWPh3-09	55.7	501	1.653	0.624
LAWPh3-10	48.2	426	1.234	0.466
LAWPh3-12	78.6	211	0.960	0.362
LAWPh3-14	54.2	143	1.320	0.498
LAWPh3-15	33.5	86.8	1.475	0.557
LAWPh3-16	207	538	3.408	1.286
LAWPh3-19_mod1	38.9	103	1.092	0.412
LAWPh3-20	8.90	23.1	0.741	0.280

Table B-4. Calculated alteration layer thickness as a function of solution pH after tumbling for 48 hours. Alteration layer thickness is calculated from the normalized release of boron, as described by Equation 2.4.

LP3-01-1		LP3-02		LP3-03		LP3-04		LP3-05mod6		LP3-06		LP3-07		LP3-08	
pH	µm	pH	µm	pH	µm	pH	µm	µm	pH	µm	pH	pH	µm	pH	µm
6.171	3.69	5.85	4.92	5.901	2.57	Trial 1		2.01	5.572	4.77	5.804	5.804	4.85	Trial 1	
6.822	2.03	7.094	2.13	6.513	0.95	5.5	6.42	0.65	7.432	0.20	6.869	6.869	0.81	5.993	0.81
7.725	0.68	7.813	0.64	7.199	0.29	7.07	1.09	0.15	7.899	0.14	8.988	8.988	0.05	6.743	0.08
10.22	0.03	9.275	0.12	10.402	0.01	8.26	0.32	0.02	8.598	0.06	9.982	9.982	0.02	7.546	0.04
10.715	0.02	10.117	0.03	12.643	0.02	9.43	0.12	0.02	10.0013	0.01	10.412	10.412	0.02	9.115	0.01
11.969	0.03	12.338	0.05			10.3	0.07	0.03	12.467	0.02	12.548	12.548	0.03	9.576	0.01
						10.76	0.03							12.384	0.01
						11.97	0.04							Trial 2	
						Trial 2								5.865	0.81
						5.51	6.43							6.922	0.09
						7.17	1.11							7.679	0.04
						8.19	0.29							8.872	0.01
						9.47	0.12							9.729	0.01
						10.32	0.06							11.652	0.01
						10.85	0.03								
						12.02	0.03								
						12.54	0.04								

Table B-4 (cont.). Calculated alteration layer thickness as a function of solution pH after tumbling for 48 hours. Alteration layer thickness is calculated from the normalized release of boron, as described by Equation 2.4.

LP3-09		LP3-10		LP3-12		LP3-14		LP3-15		LP3-16		LP3-19		LP3-20	
pH	µm	pH	µm	pH	µm	µm	pH	µm	pH	pH	µm	pH	µm	pH	µm
6.296	4.78	Trial 1		6.05	0.80	5.995	4.03	Trial 1		Trial 1		6.251	4.10	6.251	3.38
7.617	0.84	5.634	3.13	7.4	0.13	7.082	0.79	5.35	6.40	5.57	6.50	7.152	0.73	7.152	0.40
9.267	0.11	6.891	0.16	8.163	0.07	8.099	0.23	6.97	1.03	7.09	1.00	7.398	0.56	7.291	0.07
10.91	0.02	7.711	0.05	9.04	0.02	9.496	0.05	7.98	0.27	7.97	0.27	9.156	0.08	7.398	0.03
12.68	0.04	8.714	0.01	10.088	0.02	10.761	0.01	8.99	0.06	8.82	0.06	10.705	0.02	10.705	0.01
		10.029	0.01	12.448	0.05	11.362	0.01	10.15	0.02	10.32	0.02	12.268	0.01	12.268	0.01
		11.947	0.03					10.87	0.02	10.84	0.01				
		Trial 2						12.09	0.02	12.08	0.02				
		5.3	2.92					12.55	0.02	12.57	0.03				
		7.102	0.16					Trial 2		Trial 2					
		7.956	0.06					5.48	6.48	5.6	6.58				
		9.493	0.01					6.91	1.02	7.04	0.99				
		9.815	0.01					7.83	0.25	7.84	0.24				
		12.236	0.03					8.78	0.08	8.79	0.07				
								10.14	0.02	10.34	0.02				
								10.73	0.01	10.75	0.01				
								12.14	0.02	12.15	0.02				
								12.59	0.02	12.58	0.02				

Table B-4 (cont.). Calculated alteration layer thickness as a function of solution pH after tumbling for 48 hours. Alteration layer thickness is calculated from the normalized release of boron, as described by Equation 2.4.

D2B		D6B		D6B10N		D6B14N		D6B20N		D10B20N	
pH	µm	pH	µm	µm	pH	µm	pH	pH	µm	pH	µm
5.831	0.01	5.81	0.01	5.945	0.03	5.239	0.09	5.472	0.12	5.56	4.62
7.338	0.00	6.772	0.01	7.408	0.01	7.055	0.04	6.777	0.02	6.501	2.41
8.141	0.00	7.817	0.01	7.901	0.00	7.447	0.03	8.601	0.01	8.207	0.03
8.412	0.00	8.634	0.00	8.151	0.00	7.932	0.02	9.454	0.01	8.889	0.02
9.128	0.00	8.707	0.00	9.389	0.00	8.825	0.01	10.007	0.01	9.418	0.02
12.288	0.02	11.741	0.02	12.185	0.04	11.978	0.05	11.987	0.08	12.328	0.11

Appendix C – Electron Probe Microanalysis (EPMA) Results

This appendix shows cross-sectional EPMA images of selected unreacted LP3 glasses and glasses after EPA 1313 testing at pH 5.5 and 12. Cross sections are provided for all of the select glasses after EPA 1313 testing at pH 5.5 whereas cross-sectional maps are only provided for unreacted glass and after EPA 1313 testing at pH 12 for LP3-07, LP3-08, and LP3-10. Based on the elemental maps, in particular Figure C-17, the alteration layers appear to be depleted in Na and enriched in Si compared to the parent glass. Most pH 5.5 alteration layers seem to delaminate. Micrographs of some unreacted glass particles are presented here to verify that the observed features on the pH 5.5 tests formed as a result of EPA 1313 testing. Alteration layers were not observable using EPMA for samples with a final pH of 12.

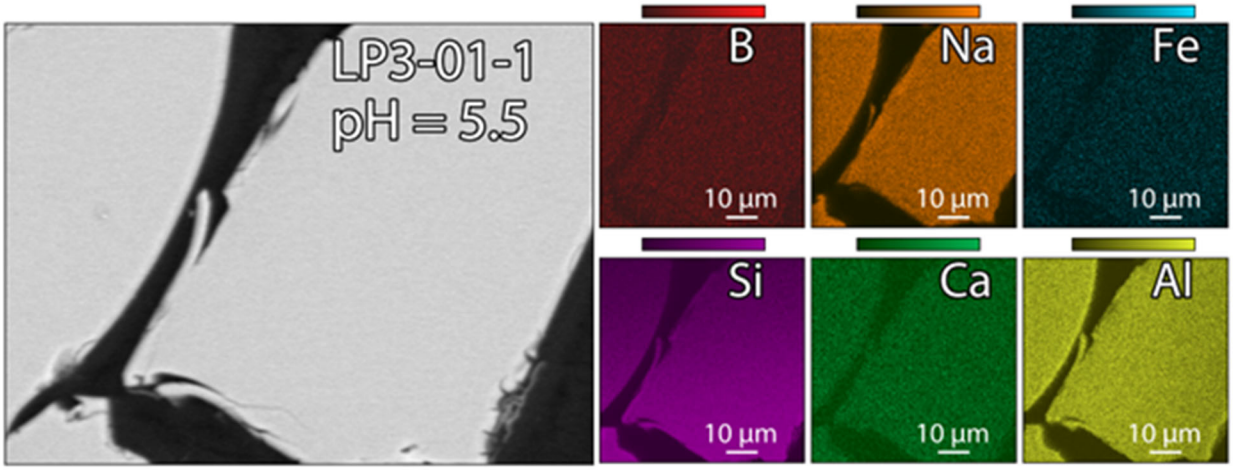

Figure C-1. LP3-01-1 after 2 Day EPA 1313 with pH = 5.5

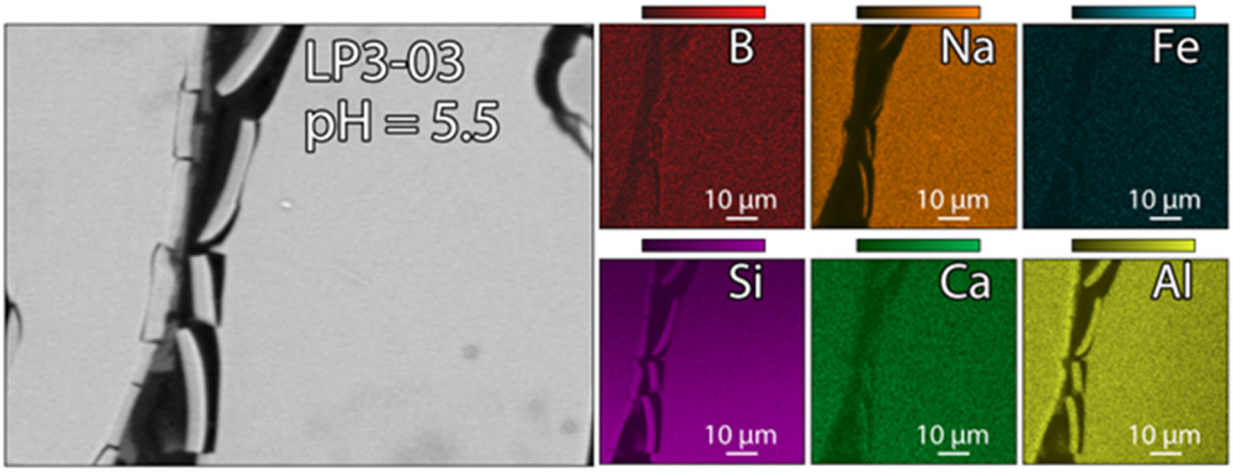

Figure C-2. LP3-03 after 2 Day EPA 1313 with pH = 5.5

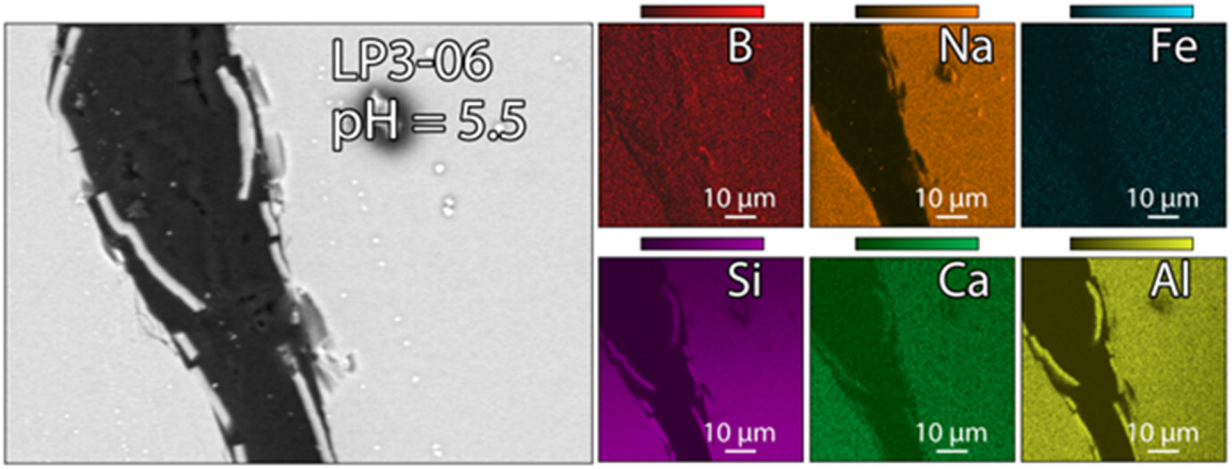

Figure C-3. LP3-06 after 2 Day EPA 1313 with pH = 5.5

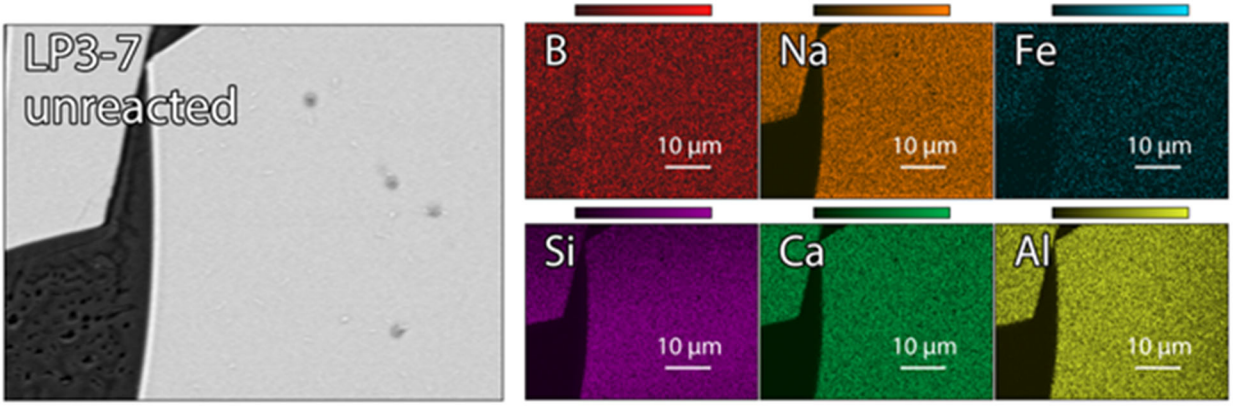

Figure C-4. LP3-07 unreacted glass

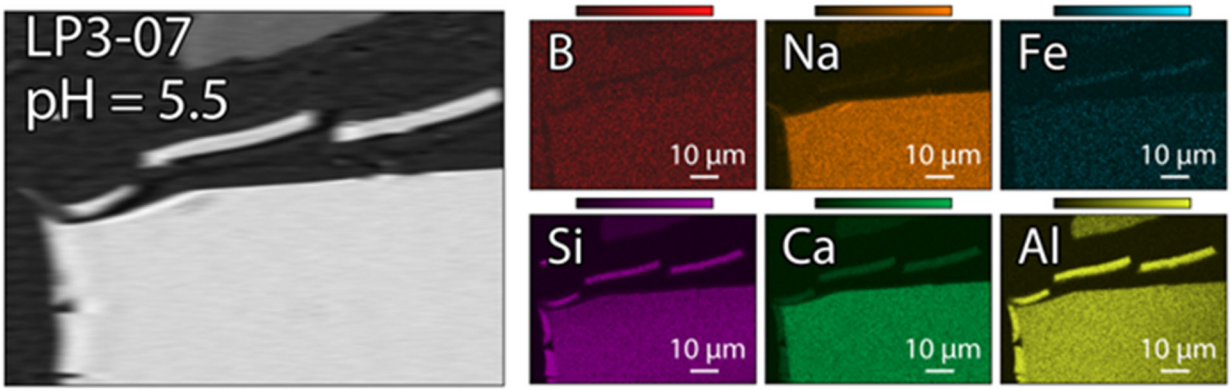

Figure C-5. LP3-07 after 2 Day EPA 1313 with pH = 5.5, map #1

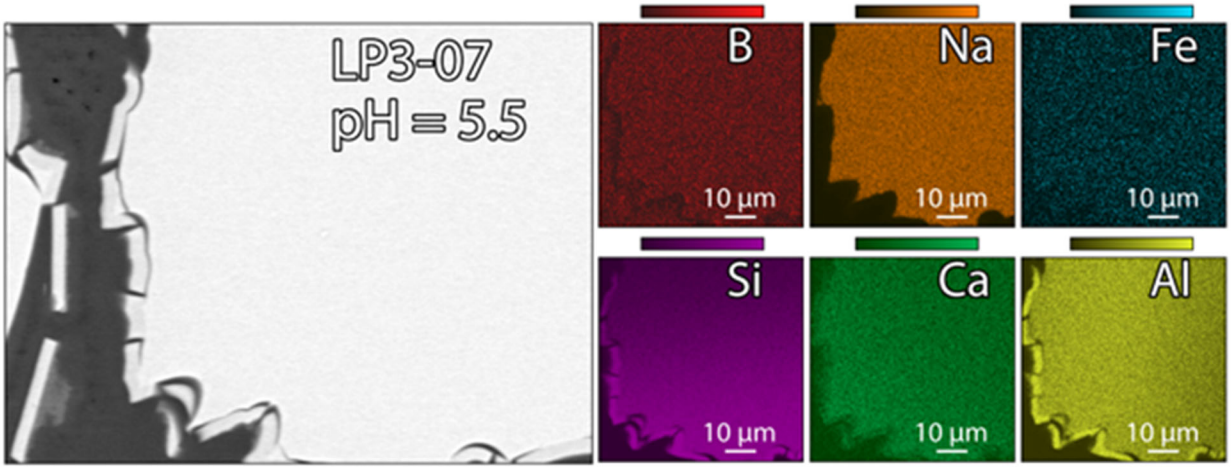

Figure C-6. LP3-07 after 2 Day EPA 1313 with pH = 5.5, map #2

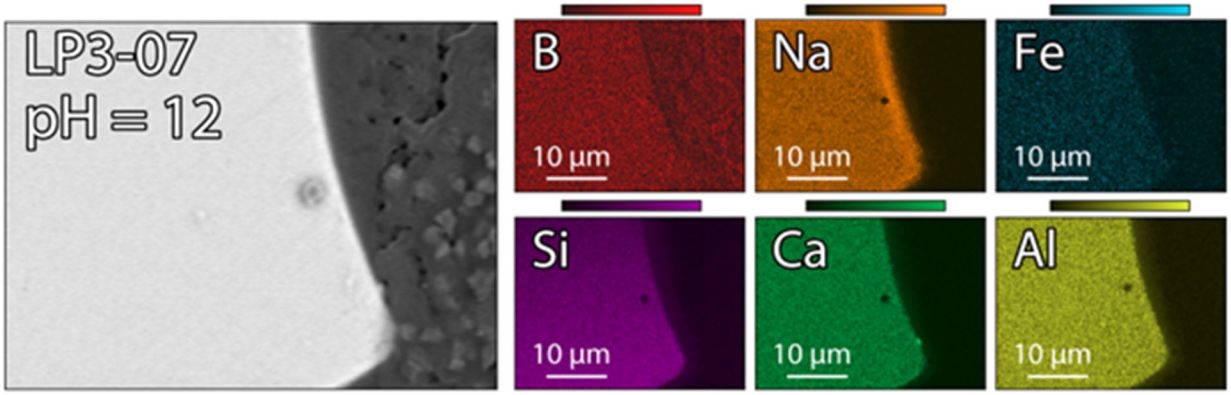

Figure C-7. LP3-07 glass after 2 Day EPA 1313 with pH = 12

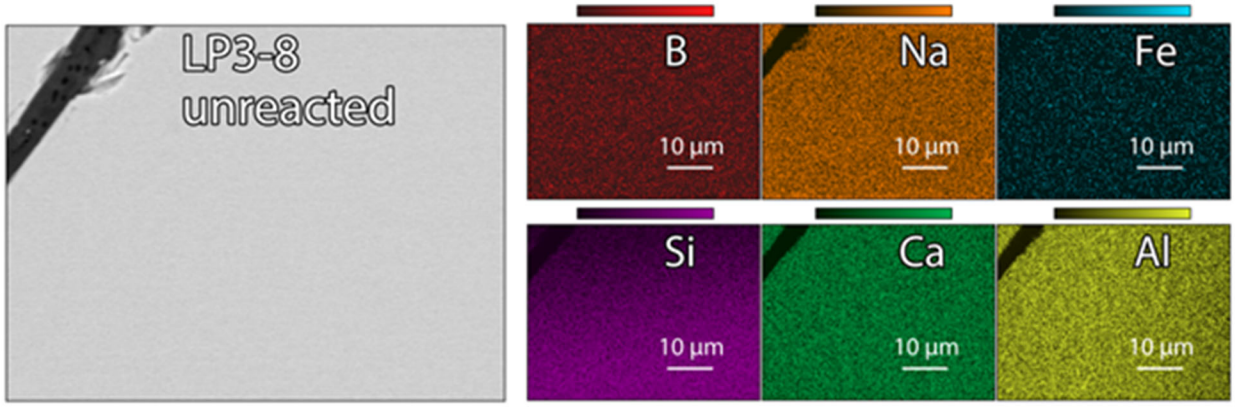

Figure C-8. LP3-08 unreacted glass

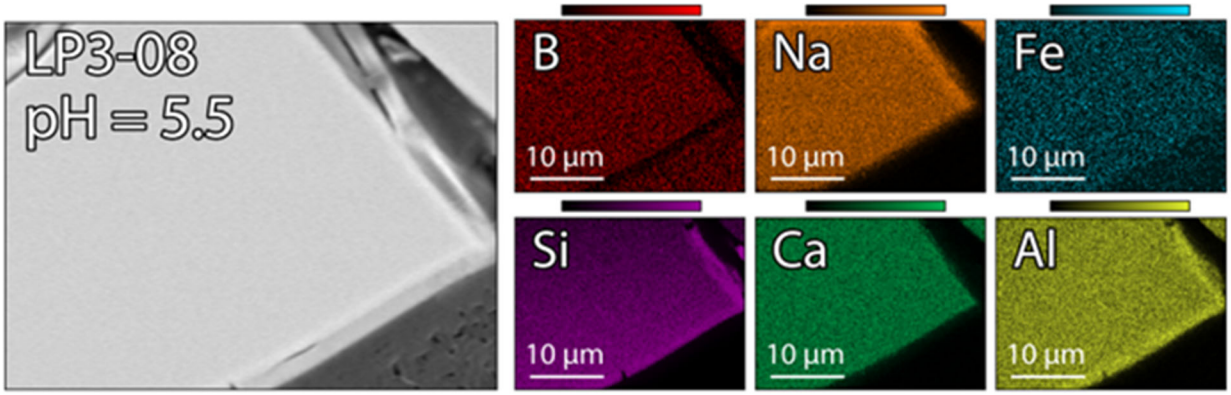

Figure C-9. LP3-08 glass after 2 Day EPA 1313 with pH = 5.5

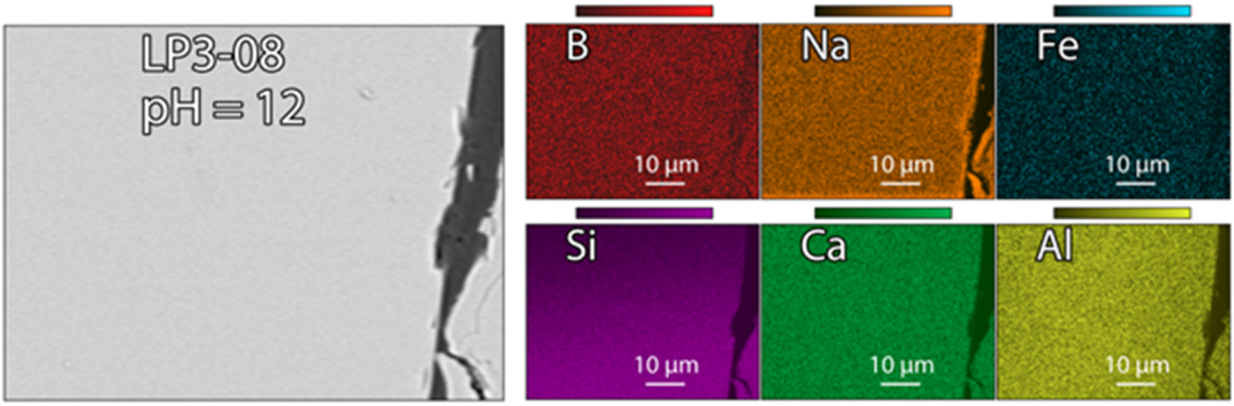

Figure C-10. LP3-08 glass after 2 Day EPA 1313 with pH = 12

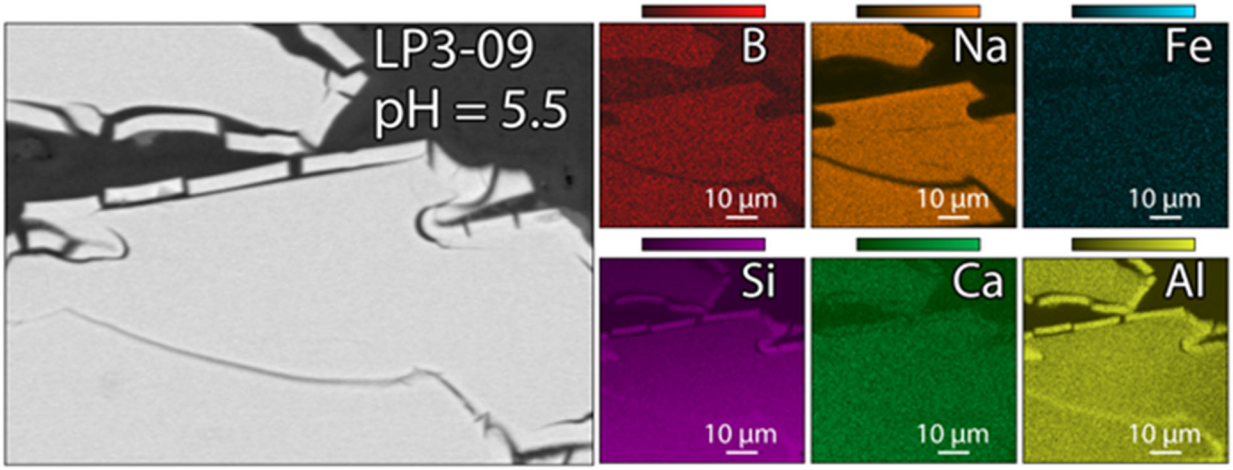

Figure C-11. LP3-09 after 2 Day EPA 1313 with pH = 5.5

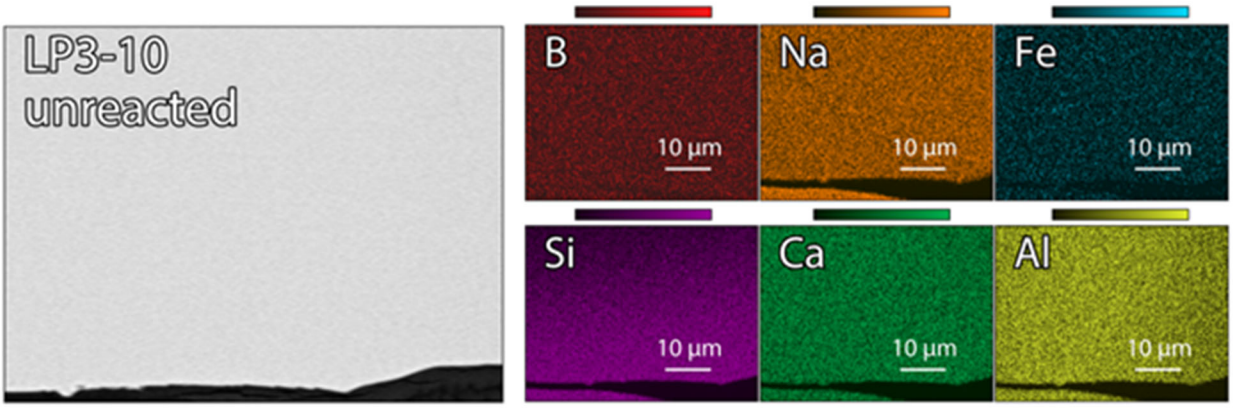

Figure C-12. LP3-10 unreacted glass

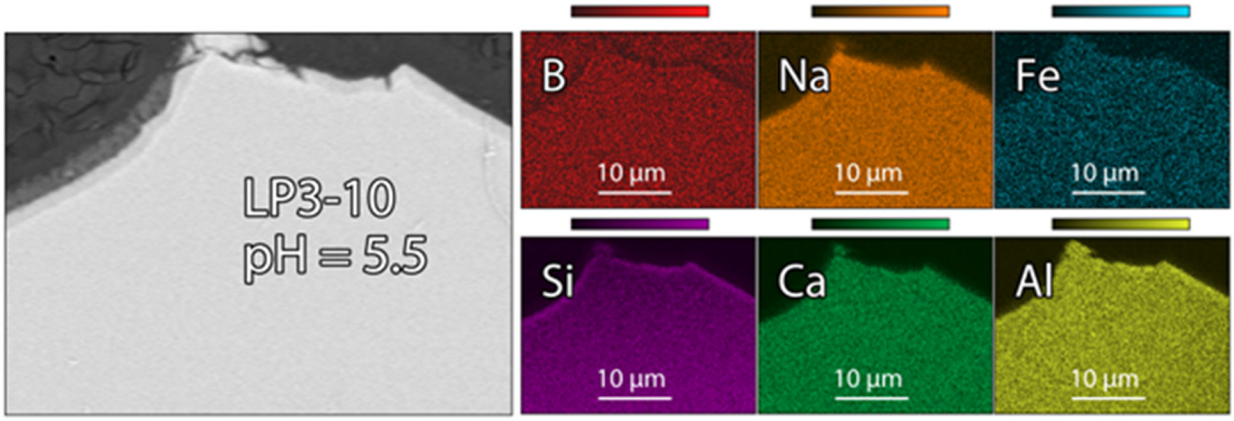

Figure C-13. LP3-10 glass after 2 Day EPA 1313 with pH = 5.5, map #1

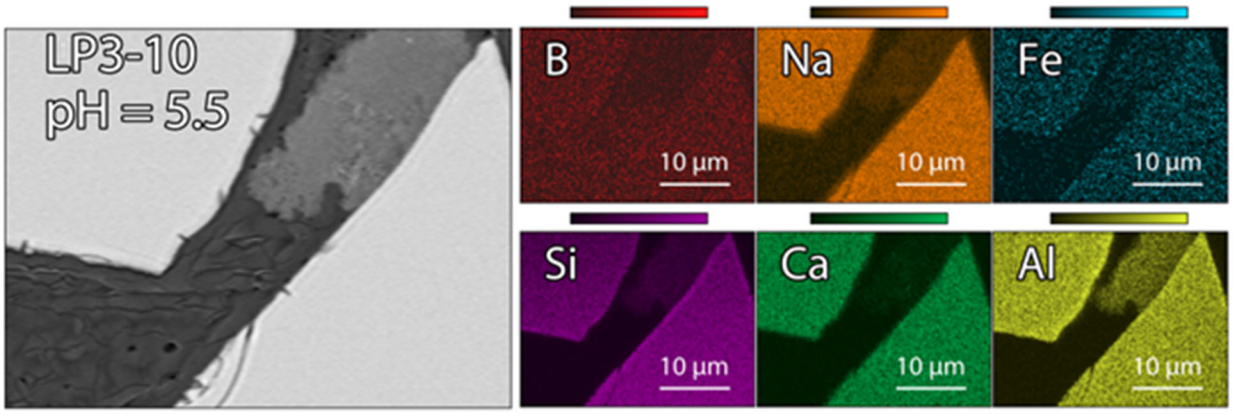

Figure C-14. LP3-10 glass after 2 Day EPA 1313 with pH = 5.5, map #2

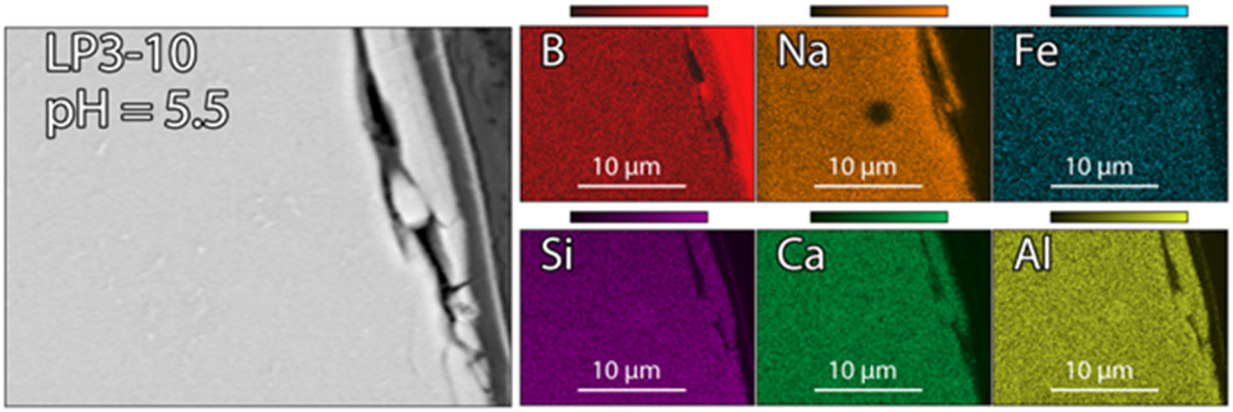

Figure C-15. LP3-10 glass after 2 Day EPA 1313 with pH = 5.5, map #3

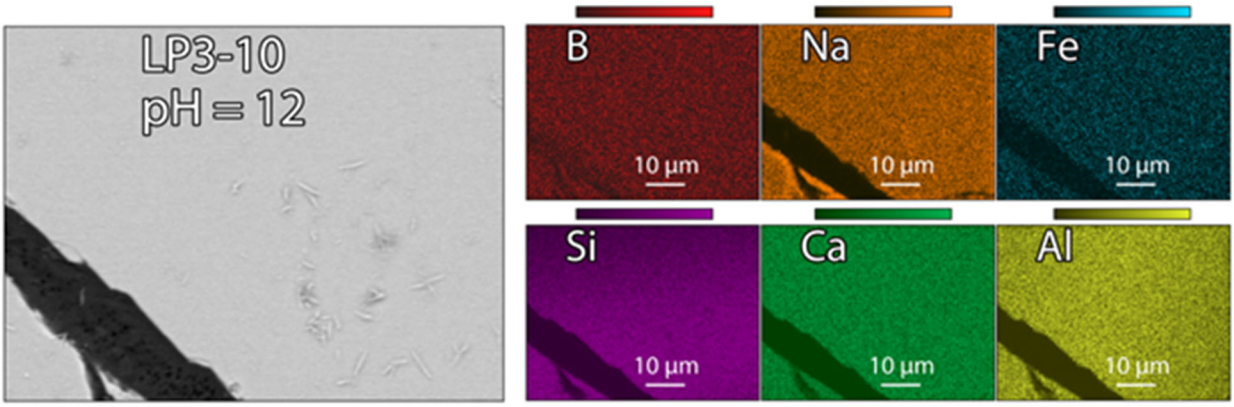

Figure C-16. LP3-10 glass after 2 Day EPA 1313 with pH = 12

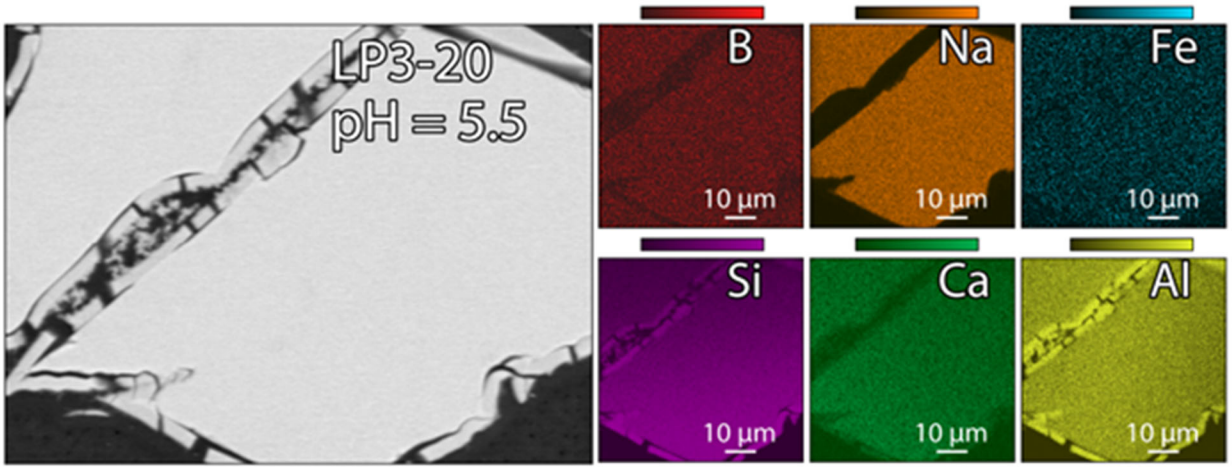

Figure C-17. LP3-20 after 2 Day EPA 1313 with pH = 5.5

Appendix D – Modeling of Normalized EPA 1313 Elemental Releases

This appendix presents the modeling results for the preliminary modeling of the elemental release during EPA 1313. First, the normalized elemental release rates for Na, Ca, and B were fit based on the general alteration rate equation of Strachan (2017), described in Section 3.4:

$$r = k_i \left[\exp\left(-\frac{E_{aH^+}}{RT}\right) a_H^{\eta_a} + \exp\left(-\frac{E_{aH_2O}}{RT}\right) + \exp\left(-\frac{E_{aOH^-}}{RT}\right) a_{OH}^{\eta_b}\right] \quad \text{(D.1)}$$

where, expressed more simplistically, a stepwise linear model was used to fit to data where they were averaged into the parameter "ln(Ave)", the ln(Ave) values:

$$ln(Ave) = \begin{cases} \sum b_i x_i + c_1 \text{pH}, & \text{pH} < c = 11.5 \\ \sum b_i x_i + c_2 \text{pH}, & \text{pH} \geq c = 11.5 \end{cases} \quad \text{(D.2)}$$

where x_i is mole fraction of oxide component i, b_i is the linear compositional term of oxide component i, c_1 is the pH-dependent rate term for the acidic range, c_2 is the pH-dependent rate term for the basic range, and c is the pH cutoff. A sweep of cutoff values was performed to determine the optimal value based on goodness-of-fit parameters, R^2 and root mean square error (RMSE). The optimum value was found to be near c =11.5, see Figure D-1.

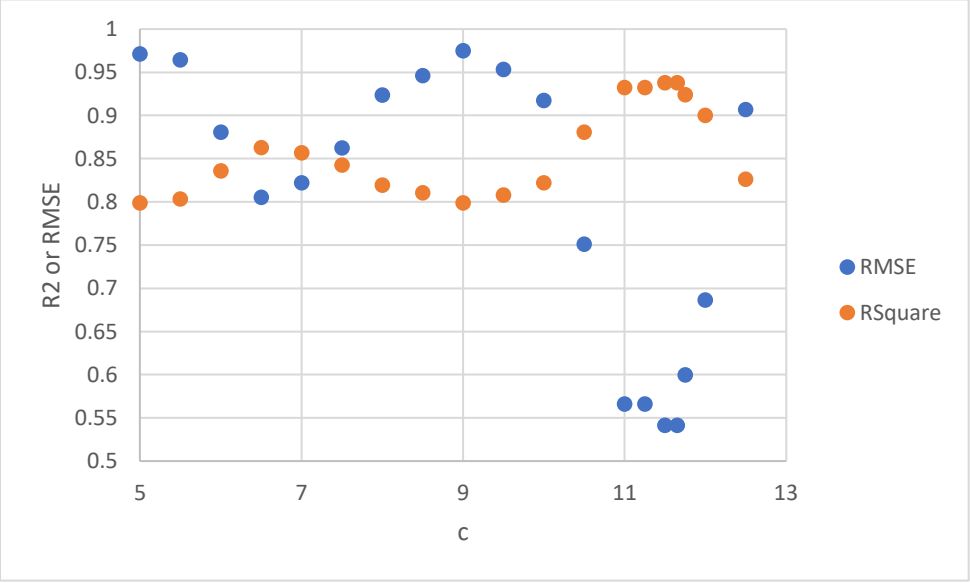

Figure D-1. Comparison of fitting parameter as a function of cutoff, c.

Figure D-2 compares the measured and modeled ln (Ave) values for the sixteen LP3 glasses and the seven join glasses using the optimized cutoff value from Figure D-1. In Figure D-2, data points shown in red deviated significantly from the stepwise linear model and were excluded from the fit. These datapoints corresponded to the join glasses (Dike, D2B, D6B, or D10B20N) and LP3-20 glass tested under alkaline

conditions (pH>11.5), under low pH conditions (pH<6.5). In Figure D-2, the simulated ln (Ave) versus pH yielded a non-physical trend suggesting the need for further modifications to the model.

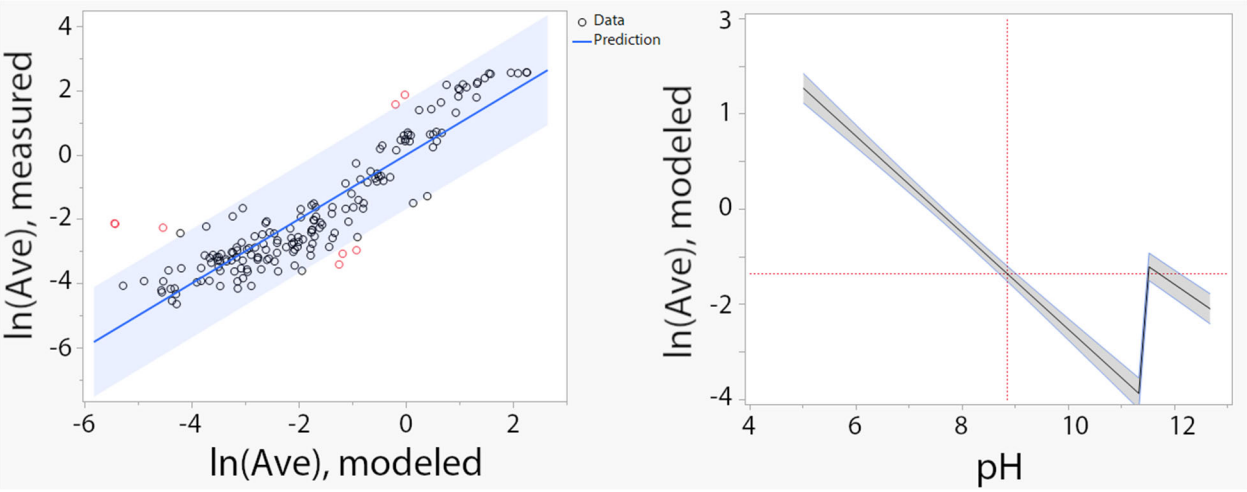

Figure D-2. (Left) Measured vs modeled ln (Ave) values for 16 LP3 glasses and 7 join glasses. (Right) Measured ln (Ave) values vs pH, based on stepwise linear model

Table D-1. Stepwise linear model coefficients fit to ln (Ave) data using Equation D.2.

Parameter	Estimate	Standard Error
b_{Al2O3}	-6.10	9.45
b_{B2O3}	16.20	3.38
b_{CaO}	13.74	3.72
b_{Fe2O3}	-6.25	32.86
b_{K2O}	32.78	9.95
b_{MgO}	3.02	7.94
b_{Na2O}	12.37	1.89
b_{SiO2}	3.43	2.47
b_{SnO2}	15.96	23.54
b_{V2O5}	33.31	20.14
b_{ZnO}	-41.77	28.43
b_{ZrO2}	0.42	23.52
b_{Others}	14.54	11.26
c_1	-1.01	0.05
c_2	-0.77	0.03