Comment

GEOSPHERE

GEOSPHERE; v. 15, no. 2

https://doi.org/10.1130/GES02047.1

4 figures (2 are interactive); 1 table

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CITATION: Baker, L.L., Camp, V.E., Reidel, S.P., Martin, B.S., Ross, M.E., and Tolan, T.L., 2019, Alteration, mass analysis, and magmatic compositions of the Sentinel Bluffs Member, Columbia River flood basalt province: COMMENT: Geosphere, v. 15, no. 4, p. 1436–1447, https://doi.org/10.1130/GES02047.1.

Science Editor: Shanaka de Silva

Received 26 July 2018
Revision received 31 August 2018
Accepted 20 November 2018

Published online 2 July 2019

Alteration, mass analysis, and magmatic compositions of the Sentinel Bluffs Member, Columbia River flood basalt province: COMMENT

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INTRODUCTION

The Sentinel Bluffs Member is the youngest unit of the Grande Ronde Basalt, having typical physical characteristics of most large-volume Columbia River Basalt Group (CRBG) sheet flows. These flows have been studied for over 50 years, resulting in numerous scientific publications from academic institutions, federal and state regional mapping projects, more detailed mapping associated with the U.S. Department of Energy's (USDOE) Hanford Site (southeastern Washington State), and borehole investigations focused in part on petroleum exploration in the Columbia Basin. The compositional heterogeneity of individual flows, although small, has been interpreted by many workers as being the result of magma chamber processes (e.g., Wright et al., 1973; Wright and Swanson, 1980; Mangan et al., 1986; Hooper, 2000; Reidel, 2005) combined in places with surficial mixing as the flows are emplaced (e.g., Reidel and Tolan, 2013). Sawlan (2018) argues instead that flow heterogeneity in the Sentinel Bluffs Member is largely the result of previously unrecognized chemical alteration of basalt in contact with aquifers, accompanied by significant mass loss (up to 50%). The fact that nearly all CRBG lava flows are in contact with the confined aquifer system, even those in the anticlinal ridges, suggests that such alteration, if it existed, could be widespread.

The ramifications of widespread, hitherto undocumented alteration are compelling, in part because of the ambiguity such alteration would place on previous interpretations that rely on high-quality chemical data. Such data could be queried as problematic or invalid in a variety of studies that focus, for example, on chemostratigraphic correlations (e.g., Reidel, 2005; Conrey and Wolff, 2010; Vye-Brown et al., 2013; Martin et al., 2013; Wells et al., 2009) or on petrogenesis and evolution of the basalts (e.g., Davis et al., 2017; Blake et al., 2010; Martin, 1989; Wolff et al., 2008; Wolff and Ramos, 2013; Ramos et al., 2005, 2009, 2013; Tollstrup et al., 2002; Hooper, 1984; Hooper and Hawkesworth, 1993; Rodriguez and Sen, 2013; Thordarson and Self, 1998), as well as in paleomagnetic studies where the results are extremely sensitive to alteration especially in the case of iron minerals (e.g., magnetite, hematite) and rely on compositional data for flow identification (e.g., Wells et al., 1989; Hagstrum et al., 2010).

Consideration and scrutiny of the Sawlan (2018) study is both timely and relevant to workers on the CRBG, but it is also pertinent to workers on other basalt provinces where aquifers are present (e.g., Hawaii, the Snake River Plain [Idaho], and the Oregon High Lava Plains). We therefore embrace debate on the potential role of these aquifers in modifying primary magmatic compositions. It is our contention that careful sample collection of the CRBG has generated largely unaltered chemical compositions, and that the alteration hypothesis of Sawlan (2018) is incorrect due to flawed methodology. As a contribution to this debate, we (1) examine long-established procedures and sampling techniques that have allowed workers to avoid significant alteration when collecting and analyzing CRBG flows; (2) describe data collection of the Sentinel Bluffs Member and chemical variations that have been attributed to both magmatic processes and secondary alteration; and (3) discuss the concerns we have on the rationale and analytical methodology that form the basis for Sawlan's (2018) conclusions that compositional variations in the Sentinel Bluff Member are the result of chemical alteration from basalt-water interaction.

ESTABLISHED METHODOLOGY OF SAMPLE COLLECTION, THIN-SECTION ANALYSIS, AND XRF GEOCHEMISTRY

Since the first use of chemistry to characterize rocks, geologists have been concerned about the effects of alteration on the composition. This potential problem was recognized by early CRBG geologists who employed difficult wet chemical analysis. In the 1960s and 1970s when rapid X-ray fluorescence (XRF) analyses became available, geologists continued to be concerned about the potential of chemical differentiation of the CRBG lavas by secondary alteration.

Workers with decades of experience on the CRBG have established a standard procedure for sample collection to avoid alteration (e.g., Swanson and Wright, 1981). This involves an initial inspection for (1) signs of alteration and weathering, (2) significant fractures, and (3) vesicles that characterize flow tops and bases as well as vesicle cylinders and lenses in the more massive parts of flows. Hand samples are typically broken into smaller chips and inspected for signs of alteration, with altered or amygdule-rich chips removed prior to crushing. Sawlan (2018) appears to have applied a similar procedure. However, Reidel (2005) and Reidel and Tolan (2013) applied an additional important step where chips were washed in an ultrasonic cleaner to remove smaller particles that might otherwise escape notice and impact the analysis. It is important to note that even in areas like the Portland Basin (Oregon) where the basalt has undergone extreme alteration, good samples can still be obtained with care in collecting and preparation. Tolan et al. (2009a, 2009b) demonstrated how alteration in this region progresses from an absence of alteration in most core stones to the altered margins of basalt columns, as reflected in variations of whole-rock chemistry. Workers have learned from decades of field experience that excellent samples can often be obtained if one takes care and collects unaltered rock from the core of altered columns.

For well over 100 years, thin-section analysis of the CRBG lavas has been the universal method for determining the texture and mineralogy in each flow as well as the degree and type of alteration. The preservation of fresh, unaltered glass is commonly used as an indicator for the lack of alteration. Peacock and Fuller (1928) recognized two types of glass in the CRBG: tachylyte, a dark colored glass containing minute iron-oxide crystals; and sideromelane, a pure, vitreous glass typically generated by quenching. The alteration of both types is relatively easy to recognize in thin section. Unaltered glassy mesostasis in the CRBG is typically isotropic and has a rhyolitic composition (Lambert et al., 1989; U.S. Department of Energy, 1988). Horton (1986) found that incipiently altered glassy mesostasis is generally semi-opaque to dark brown. More intensely altered glass is typically orange-brown with variable birefringence, but can exhibit light-brown color or various shades of yellow-brown, drab olive green–brown, or red. These optical characteristics have allowed workers to discern the difference between fresh and incipiently altered glass when selecting samples for XRF analysis.

SENTINEL BLUFFS DATABASE

In the late 1970s and 1980s, the USDOE's Basalt Waste Isolation Project (BWIP) targeted the Sentinel Bluffs Member as a potential deep-storage site for high-level nuclear waste. Sawlan (2018) describes Sentinel Bluffs samples collected by the BWIP and later investigations as being altered to variable degrees with mass loss of mobile elements. Thousands of analyses of the Sentinel Bluffs Member have been published by Mangan et al. (1985), Landon and Long (1989), Wright (1988), Wright et al. (1973, 1979, 1980, 1982, 1989), and Hooper (2000) to name just a few, but we focus here on the most recent and complete data sets of Reidel (2005) and Reidel and Tolan (2013).

The BWIP project ended in 1988 but investigations on the Sentinel Bluffs Member continued. Landon and Long (1989) recognized subtle but significant compositional variations in the Sentinel Bluffs flows based on analyses obtained on a Philips 1410 XRF spectrometer at Washington State University (WSU). Reidel (2005) reran these samples on WSU's newer equipment to ensure analytical consistency with newer data. Close scrutiny of the chemical data together with a reexamination of thin sections (polished) confirmed the original conclusion of Landon and Long (1989) that the Sentinel Bluffs analyses were obtained from fresh, unaltered samples representative of true magmatic compositions.

Reidel's (2005) study incorporated a large amount of chemical data from over 193 measured and collected stratigraphic sections. This comprehensive catalog included samples from new field sections and well borings together with a reexamination of older analyses (e.g., Hooper and Gillespie, 1996; Wright et al., 1979, 1980, 1982); the composite database included 2268 XRF analyses for major and trace elements (samples prior to 1989 were reanalyzed), 77 Instrumentation Neutron Activation Analyses (or INAA), and over 766 thin sections (polished and unpolished). In addition, 39 of the XRF samples were measured for water content (mean values of 0.93% H_2O^+; 0.73%, H_2O^-) and 20 samples for iron content (mean values of 9.26% FeO with 1σ of 0.61%; 2.52% Fe_2O_3 with 1σ of 0.68%). The H_2O content is comparable to that measured by Wright et al. (1979, 1980, 1982) and is a good measure of hydrothermal alteration, or lack thereof.

The thin-section database on Sentinel Bluffs Member flows included 406 samples from surface sections and 360 from continuous core. Samples were collected at various levels within individual flows, and thin sections were cut and evaluated with the intent of selecting unaltered samples for XRF analyses. Selected samples were point counted, and polished thin sections were used to determine mineral and glass compositions by microprobe analysis. Detailed thin-section analysis of the Sentinel Bluffs Member was used to describe the general petrography, but also to characterize alteration at a microscopic level. Alteration was an important consideration for the BWIP because the heat-generating waste was to be entombed in saturated confined aquifers of the Grande Ronde Basalt. Thus, numerous field and laboratory studies were performed to evaluate basalt-water interaction and how heated water reacts with basalt (e.g., U.S. Department of Energy, 1988, and references therein). Below is a brief summary of those studies.

STUDIES OF ALTERATION FROM PREVIOUS INVESTIGATIONS

Secondary alteration mainly occurs in flow tops and bottoms in contact with confined aquifers, and along cooling joints and tectonic fractures (e.g., McMillan et al., 1989; Horton, 1986; Lindberg, 1986, 1989; U.S. Department of Energy, 1988), as also noted by Sawlan (2018). The flow tops and bottoms form the confined aquifer system and have a typical hydraulic transmissivity of 10^{-6} m^2/s, but the flow interiors are regarded as aquitards with hydraulic transmissivity in the range of 10^{-12} m^2/s (e.g., U.S. Department of Energy, 1988, v. 2; Strait and Mercer, 1987, and references therein).

Over 3200 joint widths and mineral infillings from surface outcrops and core in mainly the Sentinel Bluffs Member and a Winter Water Member flow were described by Lindberg (1989) and Meints (1986). They determined that the majority of the joints in all flows were normal cooling joints with a median joint width of 0.14 mm. While the dense interior portion of these flows were replete with cooling joints, in their undisturbed state these joints were found to be typically 77% to >99% filled with secondary minerals (clay, silica, zeolite), and void spaces that do occur are typically not interconnected (U.S. Department of Energy, 1988; Lindberg, 1989). Thin-section studies show that the basalt from the dense flow interiors lacks evidence of microfracture and micropore systems that were suggested to be present by Benson and Teague (1982). The physical properties of the cooling joints within the dense interior portion of the basalt flow produce an essentially impermeable barrier to groundwater movement for all practical purposes (Newcomb, 1969; Oberlander and Miller, 1981; Davies-Smith et al., 1988; Lite and Grondin, 1988; U.S. Department of Energy, 1988; Lindberg, 1989; Wozniak, 1995; Tolan et al., 2009b; Burt et al., 2009; Vaccaro et al., 2009; Ely et al., 2011; Lite, 2013). The fact that CRBG dense flow interiors typically act as aquitards accounts for the confined behavior exhibited by most CRBG aquifers.

Project scientists (e.g., McKinley et al., 1986; Horton, 1986; U.S. Department of Energy, 1988; and references therein) worked under the rational assumption that a lack of mineral dissolution, oxidation rinds, or secondary mineralization in thin section, together with the presence of fresh glass, indicates an insignificant degree or lack of chemical modification by secondary alteration. Secondary minerals that formed by alteration in the flow interiors, away from flow tops and bottoms and in the internal fractures, typically comprise <1% of the mineralogy (McKinley et al., 1986; Reidel and Valenta, 2000, their appendix C, e.g., samples 399-39-84 3870, 399-39-84 3878, 399-39-84 3176, 399-39-84 3191). Horton (1986) found that the most abundant secondary mineral phases in the more altered regions of the Sentinel Bluffs Member are smectite, clinoptilolite, calcite, pyrite, and varieties of silica (i.e., opal-C, opal-CT, and quartz). Alteration rinds in the CRBG penetrate to a depth of ~0.75 cm, with a textural progression from fracture surface to unaltered basalt (McKinley et al., 1986; U.S. Department of Energy, 1988).

In order to assess the degree of alteration in basalt that lies in close proximity to zones of visible alteration (i.e., vesicular zones and fractures), Horton (1986) selected a series of samples for thin section study and XRF analysis (from Reidel and Valenta, 2000, their appendices C and B, respectively). For example, samples C4278, C4289, C4296, C4298, and C9014 were collected within and adjacent to cooling fractures. Thin-section studies showed alteration to secondary minerals along the fractures, but carefully chosen chips collected away from the alteration provided excellent analyses. In addition, samples C4270 and C4271 were collected adjacent to a vesicular zone of the Cohassett flow and showed some alteration near the vesicles in thin section, but carefully chosen chips away from the interior vesicular zone provided excellent analyses (Reidel and Valenta, 2000, their appendix B).

Experimental studies of synthetic groundwater–basalt interaction show that the glassy mesostasis is preferentially altered with only minor alteration of coexisting mineral phases (e.g., Allen et al., 1985; Lane et al., 1984; U.S. Department of Energy, 1988, and references therein). Fractures exhibit early hydrothermal alteration of the glassy mesostasis pyroxene, in preference to plagioclase and titanomagnetite in the mesostasis. Plagioclase-mesostasis boundaries and pyroxene-plagioclase boundaries are unaltered at the fracture surface, but pyroxene-mesostasis boundaries do show signs of alteration.

Although weathered sections of the CRBG have been documented (e.g., Takeuchi et al., 2007; Thomson et al., 2014; Hobbs and Parrish, 2016; Baker and Neill, 2017), the majority of flows have remained largely unweathered east of the Cascade Range where the rain-shadow effect has existed since the earliest of the flood-basalt eruptions (e.g., Takeuchi et al., 2007; Hobbs and Parrish, 2016). Wright et al. (1989) collected a large database of fresh sideromelane glass from water-quenched pillows, hyaloclastic debris, partially palagonitized vent spatter, and the selvage zones of dikes, all of which were used to study the initial composition of the first basaltic liquids to be erupted from a dike-vent system. Unaltered glass has also been found in Pele's tears from a Grande Ronde vent as well as in selvage zones from dikes (Reidel and Tolan, 1992). In addition, a large database of thin sections collected by students and researchers over the decades confirms the common appearance of preserved glass in the CRBG lavas east of the Cascades.

In contrast, the Portland-Willamette Basin west of the Cascades has experienced the most extreme cases of alteration in the CRBG. Storm development and high precipitation rates in this region have generated significant weathering in the CRBG lavas, culminating at locales like the Salem Hills (Oregon) where Sentinel Bluffs and Winter Water flows have degraded completely to ferruginous bauxite (Hoffman, 1981; Corcoran and Libbey, 1956; Libbey et al., 1945). These basalt remnants would have been mined for aluminum ore except for the high residual iron content left behind as the original flows weathered for 15 million years. These bauxite deposits average 36.3% Al_2O_3, 31.8% Fe_2O_3, and 5.9% SiO_2 (Hoffman, 1981), demonstrating the residual nature and lack of mobility of both Al and Fe. This observation lies in contrast to the conclusion of Sawlan (2018) that iron depletion is a significant part of the observed mass loss in the CRBG lavas.

Thin-section studies of basaltic and andesitic rocks in the Portland Basin show that the first constituent to decompose is interstitial glass and glassy mesostasis followed by pyroxene and plagioclase. Here, plagioclase has altered mainly to kaolinite and metahalloysite, but pyroxene and basaltic glass have altered mainly to nontronite, hematite, limonite, and amorphous clay. Iron-bearing opaque minerals remain nearly unaltered and contribute iron to the residuum (e.g., Jackson, 1974; Hoffman, 1981; Martly, 1983; Eggleton et al., 1987). Alteration and weathering of basalts result in the progressive depletion of MgO in addition to SiO_2, Na, La, Sm, and Lu, and the relative enrichment of

Al_2O_3, Fe_2O_3, TiO_2, Sc, Cr, Th, and Hf due to their immobility in aqueous phases. Other elements like La, Sm, Ce, Hf, Th, Sc, Co, and MnO form a third group that displays sympathetic geochemical variation in concentration or depletion relative to parent material concentration within the laterite profiles. Silica depletion, the key process in the development of ferruginous bauxite, similar to the bauxite at Salem Hills, is dependent on removal of leachate by the flushing action of rainwater and groundwater (Hoffman, 1981).

CHEMOSTRATIGRAPHIC CONSISTENCY OF LARGE-VOLUME SHEET FLOWS

An important consideration for recognizing flow heterogeneity due to magmatic processes, as opposed to alteration effects, lies in the distribution of flows that maintain their compositional range over their entire area of exposure. Conversely, alteration at a contact with an aquifer or weathering at the surface should be a site-specific process localized to those distinct domains.

The large amount of petrochemical data from many stratigraphic sections allows us to test such compositional variations in Sentinel Bluffs Member flows from dike source to flow terminus. Most of the dikes and vent systems for these flows are known. Mapped dikes have been analyzed for the basalts of McCoy Canyon, Stember Creek, Spokane Falls, and Museum (Reidel et al., 1992).

An excellent example of dike-to-vent-to-flow has been documented by Hooper and Gillespie (1996) for the Stember Creek flow. They recognized the Stember Creek vent at the continuation of the dike and mapped the flow throughout their study area in the Blue Mountains of southeastern Washington. A review of their analyses clearly shows essentially homogeneous flow compositions extending over a wide area, rather than localized anomalies that would be expected from weathering.

A second independent example comes from the Moscow-Pullman groundwater study along the border of Washington and Idaho, where Conrey and Wolff (2010) and Conrey et al. (2013) analyzed well cuttings from the basalt aquifers. Their studies showed that only one Sentinel Bluffs Member flow is present with precise and consistent compositions identical to that of the basalt of Spokane Falls (Reidel, 2005). The samples were selected and analyzed by Conrey and Wolff (2010) in their laboratory at WSU, which allowed them to visually recognize any alteration during sample preparation. In a more recent study, Bush and Dunlap (2017) and Bush et al. (2016) verified the conclusions of Conrey and Wolff (2010). This consistency demonstrates that the analyses represent rock magmatic compositions that were unaffected by aquifer alteration. This flow as well as the basalts of Stember Creek, Museum, and McCoy Canyon can be traced west of the Cascades into the Portland Basin (e.g., Wells et al., 2009; Evarts and Fleck, 2017; Evarts et al., 2013; Tolan and Reidel, unpublished mapping for USGS [Menlo Park, California, USA]).

In summary, these studies verify that Sentinel Bluffs compositions are regionally extensive, with individual flows having consistent compositions with minor magmatic differentiation across the 193 stratigraphic sections of the Sentinel Bluffs study area, which extends across the Columbia Basin and into western Oregon (Reidel, 2005; Reidel and Tolan, 2013).

CRBG FLOWS WITH STRONGLY HETEROGENEOUS COMPOSITIONS

Although many CRBG flows maintain largely homogeneous compositions, many other flows display significant chemical heterogeneity. The origin of several of these flows has been attributed to the simultaneous eruption and co-mingling and/or mixing of multiple flows during their emplacement (e.g., Reidel et al., 2018). Instead, Sawlan (2018) suggests that their chemical diversity is more likely derived from rock-water interaction and the alteration of chemically homogeneous flows. We disagree with this conclusion and summarize here the field evidence that supports an origin of multiple-flow injection and mixing.

Myers (1973) described the Huntzinger flow at the USDOE Hanford Site as having a diverse chemistry resulting from post-extrusive differentiation. Ward (1976) noted the unusual character of the flow in vertical section, with extreme enrichment from flow margins to center for SiO_2, FeO, Na_2O, K_2O, SrO, TiO_2, and P_2O_5, and corresponding depletion in Al_2O_3, CaO, MgO, Cr, Co, and Sc. Swanson et al. (1979) recognized the extreme chemical diversity of the Huntzinger flow but tentatively correlated it with the chemically homogeneous Asotin flow of eastern Washington because of its apparent stratigraphic position and gross chemical similarity.

The redefined CRBG stratigraphy of Swanson et al. (1979) formalized the Asotin Member (Huntzinger flow of Myers [1973] and Ward [1976]), Lapwai Member, and Wilbur Creek Member which were also recognized by Camp (1976) in the Lewiston Basin area of Idaho. In a series of geologic maps, Swanson et al. (1981) were able to trace each of these flows westward to the Pasco Basin in Washington where Reidel and Fecht (1987) correlated their stratigraphic position to the Huntzinger flow. Hooper (1985) made a compelling case for those three flows having originated by mixing in a cogenetic magma chamber. The first flow erupted was the Wilbur Creek Member. The magma chamber was later recharged by magma with Asotin chemistry and erupted as the Lapwai Member, with a mixture of Wilbur Creek chemistry and Asotin chemistry (Hooper, 1985), and then finally lava of Asotin composition erupted. Reidel and Fecht (1987) demonstrated that the extreme compositional heterogeneity observed by Myers (1973) and Ward (1976) in the Huntzinger flow consisted of variations from all three individual flows recognized in the Lewiston Basin by Camp (1976), Swanson et al. (1981), and Hooper (1985). These studies conclusively showed that the Huntzinger flow's compositional heterogeneity was a result of magmatic processes that occurred prior to eruption and during flow emplacement, and not the result of weathering or secondary alteration.

This well-grounded petrogenetic interpretation for the Huntzinger flow is identical to that of Reidel (2005) for the "Cohassett flow" of the Sentinel Bluffs Member. Like that of the Huntzinger flow, the composite chemical character of the Cohassett flow (Fig. 1) can be mapped to the east into distinctly separate flows of identical chemistry. These include the Stember Creek and Spokane Falls flows, each of which were erupted from separate mapped dikes. In a separate study, Philpotts and Philpotts (2005) performed a detailed analysis of the minerals, mineral orientations, and compositions of the Cohassett flow and recognized the same chemical heterogeneity of the flows at the same locations as Reidel (2005). Both studies presented petrological evidence that the heterogeneity was the product of magmatic differentiation, and specifically not the product of alteration. The degree of secondary alteration in the Sentinel Bluffs Member and in the Cohassett flow is examined more rigorously later in this Comment.

In still another example, Camp (1976), Swanson et al. (1979, 1980, 1981), and Hooper and Gillespie (1996) mapped two flows of the Umatilla Member in the Lewiston Basin area. Swanson et al. (1980, 1981) mapped the Umatilla Member from the Lewiston Basin to the Pasco Basin, where on the border of the Pasco Basin both flows were recognized as the Sillusi and Umatilla flows of Laval (1956). However, within the Pasco Basin, the Umatilla Member consists of only one flow containing compositions of both flows present in the Lewiston Basin (Reidel, 1998). Thus, the same processes that produced the Asotin-Huntzinger flows and the Sentinel Bluffs flows produced the Umatilla flows.

The evident heterogeneity in the Huntzinger-Asotin, Cohassett, and Umatilla flows is consistent with comingling and mixing of giant pahoehoe sheet flows associated with flow inflation. The most important field criterion for this conclusion lies in the map relationships of the composite inflated lobes comprising each mixed flow. These lobes have been traced eastward in all cases into clear and distinct separate flows. This latter relationship categorically precludes a model of secondary alteration but is consistent with a model of comingling and mixing of contemporaneous sheet flows during flow inflation.

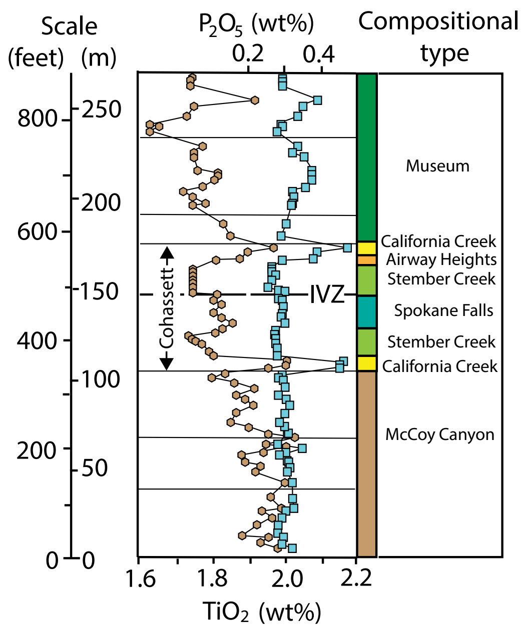

Figure 1. Vertical section of Sentinel Bluffs Member flows (Grande Ronde Basalt, Columbia River Basalt Group) and their TiO_2 (brown) and P_2O_5 (blue) contents from continuous core DC-6 at the U.S. Department of Energy Hanford Site. Cohassett—Cohassett flow (informal) of the Sentinel Bluffs Member, Grande Ronde Basalt. Modified from Reidel (2005). IVZ—internal vesicular zone.

SYNOPSIS FROM PAST INVESTIGATIONS

We conclude that a rigorous and well-established methodology employed by most workers on the CRBG has resulted in a large chemical database with reliable analyses unaffected by significant secondary alteration. Moore et al. (2018), for example, examined Al-Ti variations in nearly 150 samples of Steens Basalt (the lowest formation of the CRBG) and found no obvious alteration by anoxic waters. Although Sawlan (2018) cites the Sentinel Bluffs Member as containing suspect or unreliable data due to alteration, specific studies of the Sentinel Bluffs Member demonstrate consistent flow compositions with a lack of both thin-section and chemical evidence of significant low-temperature alteration (Reidel, 2005; Reidel and Tolan, 2013). Sawlan (2018) cites the lack of petrographic evidence noted by other workers in rocks that appear to be chemically altered, and he concludes that groundwater alteration can only be identified by examining chemical criteria. His conclusions on the Sentinel Bluffs Member are therefore based solely on chemical data from 112 samples without reference to thin-section examination. It seems improbable that samples with fresh outcrop appearance and commonly glassy and unaltered mineralogy in thin section could simultaneously show chemical trends resulting from considerable mineral dissolution. Instead, all such chemical trends observed in the Sentinel Bluffs database are consistent with those found in all other CRBG lava flows and attributed to one or a combination of magmatic processes (i.e., fractional crystallization, crustal assimilation, partial melting, and magma mixing) and/or surficial processes associated with flow inflation.

DATA CORRECTION SCHEME OF SAWLAN (2018)

Sawlan (2018) argues that numerous previously published analyses of the CRBG do not represent true magmatic compositions because they were affected by alteration. Here we show that this conclusion is incorrect, and that the data correction scheme proposed by that study is based on a misapplication of standard techniques for assessing element loss during rock weathering.

The standard techniques in question are based on the assumption that some elements are immobile in the weathering environment and passively accumulate in the weathered residue. Although no elements are truly immobile, several elements including Al, Ti, Zr, and Nb display limited mobility and often behave in a relatively conservative manner. It is possible to estimate the degree to which a given element has been leached from a weathered sample by calculating mobility ratios (Brimhall et al., 1992; Anderson et al., 2002; Thomson et al., 2014):

$$\mathrm{MR} = \left(\frac{\frac{c_{j,w}}{c_{i,w}}}{\frac{c_{j,p}}{c_{i,p}}} \right) - 1, \quad (1)$$

where MR is mobility ratio, and c is the concentration of mobile element j or immobile element i in weathered sample w or unaltered parent sample p.

As may be seen from Equation 1, in order to determine the extent of leaching, it is necessary to compare the mobile element to an immobile element, and to compare the weathered sample to an unweathered parent sample. This treatment is necessary because it takes into account changes in bulk density and volume upon leaching. The data correction scheme proposed by Sawlan (2018) does not account for these changes, even though the degree of leaching proposed by that study for some samples would certainly lead to considerable volume and density changes. This alone invalidates the data correction methodology proposed by that study.

One standard way to assess the degree of weathering experienced by a given sample is by calculating various indices that use ratios of concentrations of relatively immobile elements to those of relatively mobile elements. With progressive leaching, mobile elements are removed from the rock whereas relatively immobile elements passively accumulate. The chemical index of alteration (CIA) is one such index:

$$\mathrm{CIA} = \left(c_{Al_2O_3}\right) / \left(c_{Al_2O_3} + c_{CaO} + c_{Na_2O} + c_{K_2O}\right), \quad (2)$$

where c is molar concentration in a given sample of the element indicated (Nesbitt and Young, 1982). CIA values are low for unaltered samples and increase progressively with alteration, as relatively immobile Al accumulates in the sample and relatively mobile alkalis are leached.

Babechuk et al. (2014) proposed the more basalt-specific mafic index of alteration (MIA), with variations indicating weathering under oxidizing (MIA-O) or reducing (MIA-R) conditions. These indices take into account the mobility of Mg, as well as the fact that Fe(II) is mobile under reducing conditions whereas Fe(III) is relatively immobile under oxidizing conditions:

$$\mathrm{MIA\text{-}R} = \left(c_{Al_2O_3}\right) / \left(c_{Al_2O_3} + c_{Fe_2O_3} + c_{MgO} + c_{CaO} + c_{Na_2O} + c_{K_2O}\right), \quad (3)$$

$$\mathrm{MIA\text{-}O} = \left(c_{Al_2O_3} + c_{Fe_2O_3}\right) / \left(c_{Al_2O_3} + c_{Fe_2O_3} + c_{MgO} + c_{CaO} + c_{Na_2O} + c_{K_2O}\right). \quad (4)$$

In Table 1, we show values for CIA, MIA-R, and MIA-O calculated from supplemental file 1 of Sawlan (2018), and for known weathered CRBG samples from several previous studies (Tolan et al., 2009b, their table 2; Thomson et al., 2014; Baker and Neill, 2017). Typical values for unaltered CRBG flows are CIA = 38–40, MIA-R = 25–26, and MIA-O = 39–40. The CIA for the weathered sample sequences increases progressively from the parent basalt, indicating leaching of Ca, Na, and K. Early weathering under reducing conditions where Fe is mobile produces increases in MIA-R values, and more extensive weathering under oxidizing conditions where Fe is immobile results in increased MIA-O values. By contrast with the samples known to be weathered, the index values for the Sawlan data indicate mostly unaltered basalt compositions, with the exception of known weathered sample 070915-1604-MS-a, which shows typical values for weathered basalts, and the further example of a subset of samples discussed below. The high value for MIA-O in sample 070915-1604-MS-a, as compared to the moderate value for MIA-R, suggests that this basalt was altered chiefly under oxidizing conditions at which Fe behaved in a relatively conservative manner.

Additional useful graphical presentations of basalt weathering include ternary plots showing typically mobile elements (alkalis, Mg, Fe under reducing conditions) and typically less-mobile elements (Al, Fe under oxidizing conditions). Figure 2A is an AF-CNK-M (Al_2O_3 + Fe_2O_3, CaO + Na_2O + K_2O, MgO) plot comparing data from supplemental file 1 of Sawlan (2018) to data from CRBG saprolites that underwent varying degrees of leaching under pedogenic conditions (Thomson et al., 2014; Baker and Neill, 2017; Tolan et al., 2009b). The saprolite data (red squares) display typical trends of early loss of Mg by leaching of mafic minerals, followed by loss of alkalis under oxidizing conditions with more extensive leaching. By contrast, with the exception of known weathered sample 070915-1604-MS-a, the data from Sawlan (2018) cluster tightly together with no evidence of the Mg loss typical of early CRBG weathering.

These calculations show that the majority of the basalt compositions in supplemental file 1 of Sawlan (2018) do not display any sign of having undergone significant alteration, with the exception of known weathered sample 070915-1604-MS-a. There is therefore no justification for the arbitrary adjustments proposed by Sawlan (2018) to chemical analyses of CRBG lavas.

A possible key to some geochemical processes in the Sentinel Bluffs lavas may be observed by replotting the data from Figure 2A on an A-CNK-FM (Al_2O_3, CaO + Na_2O + K_2O, Fe_2O_3 + MgO) plot (Fig. 2B). A trend in the data away from the FM apex indicates variation in either Fe or Mg content of the samples. It is apparent in Figure 2A that the Sentinel Bluffs samples of Sawlan (2018) did not undergo Mg loss, because there is no trend away from the M apex. Therefore, the trend away from the FM apex in Figure 2B must result from variation in Fe content of the samples. It appears likely that Sawlan (2018) would have interpreted this trend as indicating leaching of Fe from these samples under reducing conditions. However, Fe contents in these samples linearly correlate with Ti contents (Fig. 3, black circles). If this trend were due to Fe loss, then immobile Ti would have accumulated as a residual element, and Fe and Ti contents would be anticorrelated. Instead, the correlation of Fe and Ti contents appears to indicate some variation in the abundance of Fe-Ti oxides in these lavas. This variation is the source of most of the trend observable in the data points from Sawlan (2018) (black circles) in Figure 3.

TABLE 1. INDICES OF ALTERATION FOR KNOWN WEATHERED CRBG SAMPLES AND FOR SAMPLES FROM SAWLAN (2018)

	CIA	MIA-O	MIA-R
Tolan et al. (2009b) samples			
AU-2	39	40	25
WASH58005-130	40	41	25
WASH58005-110	42	43	29
DMW18A28-29	41	41	28
Canby10	41	41	28
WE-850	50	51	37
Thomson et al. (2014) samples			
LC-01	40	44	26
LC-02 RAT	40	44	27
LC-03 RAT	40	44	27
LC-04 RAT	44	48	31
LC-05	44	51	31
LC-06	44	48	32
LC-07A	45	49	33
LC-PALEOSOL	95	94	72
Baker and Neill (2017) samples			
T-BASALT	38	42	26
T 80 cm	52	56	40
T 60 cm	54	56	44
T 30 cm	54	56	43
Sawlan (2018) samples from their supplemental file 1			
080721-1914MS	39	39	25
080721-1807MS	39	39	25
080721-1701MS*	39	39	25
CB0119	39	41	25
100514-1258MS	39	40	25
100514-1258MS-a	39	40	25
100514-1106MS	39	41	25
100514-1106MS-a	39	41	25
100813-1501MS	39	41	25
100813-1501MS-a	39	41	25
100813-1501MS-b	39	41	25
100512-1730MS	39	40	25
100512-1624MS	39	40	24
100512-1624MS-a	39	40	24
100512-1349MS	39	40	25
100512-1349MS-a	39	40	25
100512-1349MS-b	39	39	25
100512-1256MS	39	39	25
100512-1256MS-a	39	39	25
100512-1051MS	39	40	25
100512-1051MS-a	39	39	25
100512-1051MS-b	39	39	25
Sawlan (2018) samples from their supplemental file 1 (*continued*)			
100511-1800MS	39	40	25
100511-1800MS-a	39	40	25
100511-1303MS	40	40	25
100511-1303MS-a	40	40	26
100511-1401MS	39	40	25
100511-1620MS	39	39	25
110427-1738MS	39	41	25
110427-1738MS-a	39	41	25
100924-1440MS	39	40	25
100924-1440MS-a	39	40	25
100924-1559MS	39	40	25
100924-1559MS-a	39	40	25
100924-1559MS-b	39	40	25
100924-1742MS	39	40	25
100924-1742MS-a	39	40	25
100925-1125MS	39	39	24
100925-1125MS-a	39	39	24
100925-1302MS*	40	39	27
100925-1302MS-a*	40	39	27
100930-1545MS	39	39	25
100930-1545MS-a*	39	39	26
100930-1545MS-b	39	39	25
070921-1455-MS*	39	39	26
070913-1726-MS*	41	40	28
070915-1604-MS-a	**96**	**92**	**58**
070915-1604-MS-c*	40	40	28
070915-1822-MS*	39	40	26
070915-1604-MS-d	39	41	25
070915-1604-MS-b	39	41	25
070919-1825-MS	39	41	25
070919-1453-MS	39	40	25
070917-1630-MS	39	40	24
070916-1811-MS	39	40	25
070908-1545-MS*	40	40	27
090921-1102MS	39	41	25
090921-1102MS-a	39	41	25
090921-1238MS	39	41	25
090921-1238MS-a	39	41	25
090921-1554MS	39	39	24
090921-1554MS-a	39	39	24
090921-1719MS	39	39	25
090921-1819MS	39	39	25
090921-1819MS-a	39	39	25
080531-1610MS	39	40	25
090616-1816MS-a*	39	38	26
Sawlan (2018) samples from their supplemental file 1 (*continued*)			
090616-1816MS-b*	39	39	26
080531-1715MS	39	40	25
090617-1010MS*	39	39	26
090616-1909MS*	39	38	26
100927-1642MS	39	41	25
100927-1642MS-b	39	41	25
100927-1642MS-a	39	41	25
100928-1505MS	39	40	25
100928-1505MS-a	39	40	25
110502-1242MS	39	40	25
110502-1242MS-a	39	40	25
100927-1823MS	39	39	25
100927-1823MS-a	39	39	25
100928-1808MS	39	39	25
100928-1808MS-a	39	39	25
100928-1840MS	39	39	25
100928-1840MS-a	39	39	24
100928-1657MS	39	39	25
100928-1657MS-a	39	39	25
110430-1712MS	39	39	24
110430-1712MS-a	39	39	24
100928-1248MS	40	40	25
100928-1248MS-a	40	40	25
100928-1248MS-b	40	40	25
100928-1102MS	40	40	26
100928-1102MS-a	40	40	26
080602-1824MS	38	39	25
080722-1922MS	39	39	25
110428-1137MS	40	40	25
110428-1137MS-a	40	40	25
06BV-G523	40	40	25
97SH-X49A	39	40	24
99SH-X98	39	40	25
00SH-X111	39	40	25
97LC-Q70	39	40	25
00LC-Q447	39	40	25
T1248	39	40	25
T3407*	39	39	26
T3421*	39	39	26
T3445*	40	39	27
070921-1250RW	39	40	25
070924-1633RW	39	40	25
070926-1227RW*	39	40	26
T2128	39	40	25
T2152	39	40	25

Notes: Alteration indices calculated using published analyses of known weathered samples from the Columbia River Basalt Group (CRBG) and for samples from Sawlan (2018) supplemental file 1. Indices include Chemical Index of Alteration (CIA) (Equation 2), Mafic Index of Alteration - Reducing (MIA-R) (Equation 3), and Mafic Index of Alteration - Oxidizing (MIA-O) (Equation 4). Asterisks indicate possibly altered samples from Sawlan (2018); these correspond to circled data points in Figures 3 and 4. The outlined sample 070915-1604-MS-a from Sawlan (2018) was noted in that study as having undergone advanced weathering.

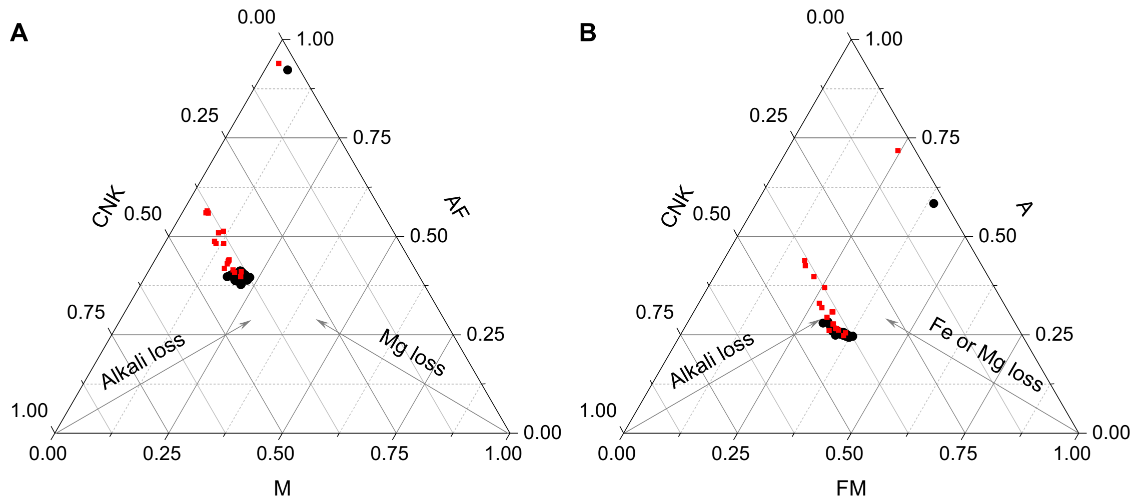

Figure 2. Ternary plots showing samples from Sawlan (2018) (black circles) and altered Columbia River Basalt Group (CRBG) samples (small red squares; Thomson et al., 2014; Baker and Neill, 2017; Tolan et al., 2009b). Arrows indicate the direction of trends that would be produced by leaching. A: AF-CNK-M plot ($Al_2O_3 + Fe_2O_3$, $CaO + Na_2O + K_2O$, MgO). B: A-CNK-FM plot (Al_2O_3, $CaO + Na_2O + K_2O$, Fe_2O_3 + MgO).

Approximately 18 of the 112 samples from Sawlan (2018) that are shown in Figure 3 plot off the Fe-Ti trend defined by most of the samples. These samples have Ti contents that fall within the range defined by other samples, but their Fe contents are low. These samples have been marked in Table 1 with an asterisk. Almost all of these samples have negative mobility ratio values for Fe, and this group of samples represents almost all of the samples in the database of Sawlan (2018) with negative Fe mobility ratios. Therefore, it appears possible that this subset of samples has undergone some alteration. Most of these samples also have anomalously low Mg contents (Sawlan, 2018, his table 1), suggesting possible minor dissolution of ferromagnesian minerals under reducing conditions that facilitated Fe loss. The MIA-R values (Table 1) are slightly elevated for approximately half of these samples, indicating the relative sensitivity of this index to leaching of Fe and Mg. These points are clearly identifiable on the A-CNK-FM graph, as shown in Figure 4, which shows only the portion of the ternary plot surrounding the cluster of data points in Figure 2B. In Figure 4, the circled data points from Sawlan (2018) are those that do not

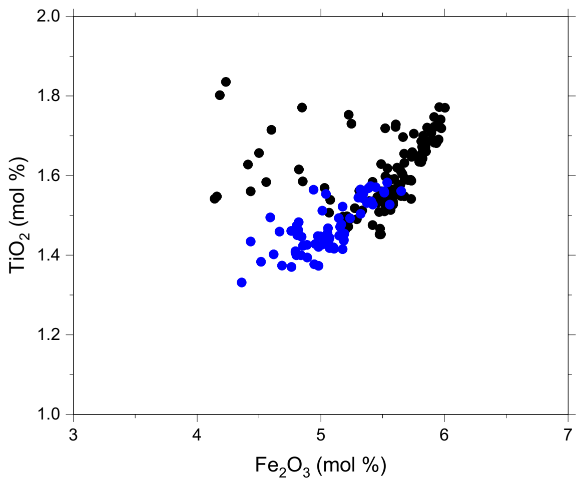

Figure 3. Graph of molar TiO_2 versus Fe_2O_3 contents in samples from Sawlan (2018) (black circles) and from Reidel (2005) (blue circles). To view the two layers of Figure 3 in the PDF version of this paper, open the PDF in Adobe Acrobat or Adobe Reader.

To view the layers while reading the full-text version of the paper, click http://doi.org/10.1130/GES02047.f3 to download a PDF of the figure.

To view the layers while reading the full-text version of the paper, click http://doi.org/10.1130/GES02047.f4 to download a PDF of the figure.

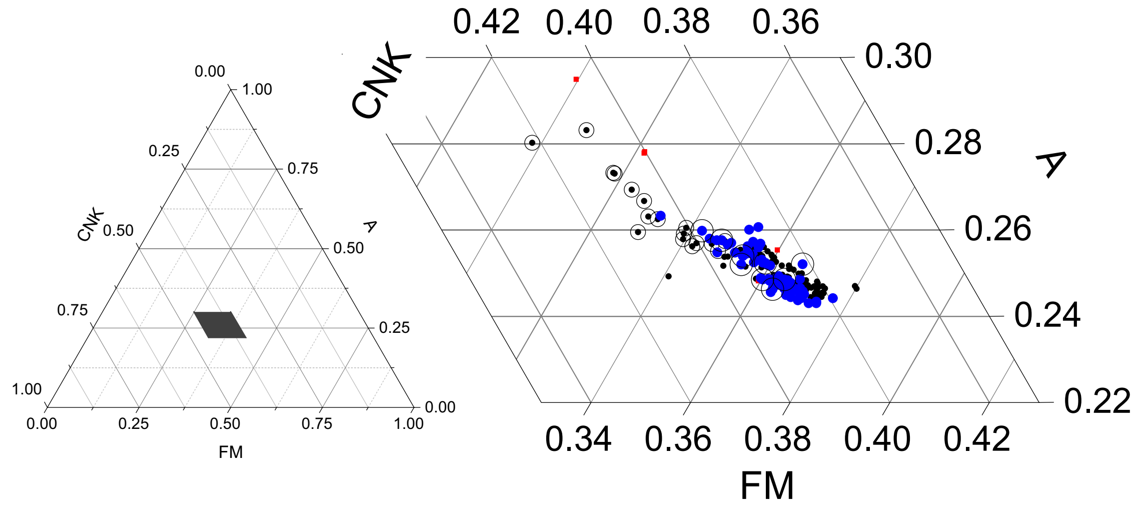

Figure 4. Section of A-CNK-FM ternary plot (Al_2O_3, CaO + Na_2O + K_2O, Fe_2O_3 + MgO) showing data as in Figure 2, cropped to zoom to the main cluster of data. Large open circles surround data points from Sawlan (2018) (black circles) that fall off of the Fe-Ti trend in Figure 3 and that are identified here as potentially altered due to Fe loss. Also shown are Sentinel Bluffs samples (Reidel, 2005) (blue circles); large open circles surround sample points that fall slightly to the left of the trend in Figure 3. To view the two layers of Figure 4 in the PDF version of this paper, open the PDF in Adobe Acrobat or Adobe Reader.

fall on the Fe-Ti trend in Figure 3. These samples follow the same general trend as the altered samples plotted in Figure 2B (red squares). Thus, our interpretation is that this subset of samples from Sawlan (2018) does indeed show signs of minor chemical weathering that are typical of the anoxic weathering trends observed at other CRBG localities (e.g., Thomson et al., 2014, their samples LC-02 and LC-03). Up to 10%–20% of total Fe and Mg appear to have been lost from the samples of Sawlan (2018) that we identify here as potentially altered. The majority of the samples examined by Sawlan (2018) show no indication of this alteration.

SENTINEL BLUFFS MEMBER TYPE SECTION AT SENTINEL GAP

As a final example, we look at data from Landon and Long (1989), Reidel (2005), and Reidel and Tolan (2013) on samples from the type section for the Sentinel Bluffs Member and Cohassett flow that are exposed at Sentinel Gap (Washington), a water gap where the Columbia River cuts through the Saddle Mountains. The Sentinel Bluffs section there includes all of the compositional types described by Reidel (2005): McCoy Canyon, California Creek, Airway Heights, Stember Creek, Spokane Falls, and Museum. Here, the Cohassett flow consists of the California Creek, Airway Heights, Stember Creek, and Spokane Falls compositional types. The data set consists of over 66 XRF analyses carried out at the Peter Hooper Geoanalytical Laboratory at Washington State University. In addition, 84 polished and unpolished thin sections accompanying the analyses were made from the same samples analyzed by XRF. The thin sections from Sentinel Gap show unaltered glass and mineral compositions except those previously noted, and all XRF samples have unnormalized major-element totals between 99% and 101%, consistent with fresh, unaltered basalt and magmatic compositions.

A linear correlation exists between Fe and Ti in these samples (Fig. 3, blue circles), indicating that the variation in Fe is likely due to variation in Fe-Ti oxide abundance, as suggested above for samples from Sawlan (2018). The Sawlan (2018) samples also plotted in this figure (black circles) have slightly higher overall Fe contents and show more indication of Fe leaching. A handful of the Sentinel Bluffs samples have Fe contents that fall slightly to the low-Fe side of the Fe-Ti trend, although generally not as far off the Fe-Ti trend as the samples of Sawlan (2018) identified as weathered.

The Sentinel Bluffs samples cluster tightly on an A-CNK-FM plot (Fig. 4, blue circles), indicating they have not undergone significant weathering or Fe loss. The samples that plot to the left of the Fe-Ti trend in Figure 3 are indicated in Figure 4 by large open black circles. Unlike altered samples from Sawlan (2018), these samples group with samples that fall on the Fe-Ti trend, indicating no discernable alteration of the Sentinel Bluffs samples at the type section.

Sawlan (2018) suggests that chemical variations among flows in the Sentinel Bluffs and other CRBG lavas are most likely due to chemical alteration of flow tops and bottoms. He emphasizes that chemically diverse flows like the Cohassett flow, which is incorporated in the Sentinel Gap section, are likely to be good examples of this process. Extensive leaching would be required to explain the observed variations in chemical composition among those flows, particularly for components such as P and K, which vary by several tens of percent. Such extensive leaching would preferentially remove mobile elements such as Mg, K, and Na, and would be immediately obvious on ternary plots such as those shown in Figures 2 and 4.

CONCLUSIONS

We agree with Sawlan (2018) that secondary alteration is a common feature in the CRBG lavas, particularly where wet conditions prevail west of the Cascade volcanic arc, and we appreciate his attempt to assess the degree of alteration and to better understand the mechanisms of element mobility. We disagree, however, on his methodology and his conclusion that secondary alteration is pervasive and responsible for the significant chemical diversity displayed in several regionally extensive flows of the CRBG such as those in the Sentinel Bluffs Member of Reidel (2005) and Reidel and Tolan (2013). Instead, we remain convinced that a long history of careful mapping, conscientious sample collecting, and close scrutiny of thin sections has resulted in a geochemical database of basalt compositions that show little effect of secondary alteration, but retain compositions indicative of their magmatic evolution and emplacement histories.

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